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Introduction to Radiochemistry

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BY

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AND

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To

JOYCE CLENNAM STEARNS

PREFACE

An increasing number of universities are offering courses in radioactivity for chemists. Very likely many teachers and students in these courses feel as we do that there has been no suitable textbook for this purpose. There is the very excellent *Manual of Radioactivity* by G. Hevesy and F. A. Paneth; however, advances in the science since its last edition, in 1938, have been more than any authors should have to expect in one decade. Moreover, no recent book on the subject has been written specifically for chemists. We have tried to prepare a textbook for an introductory course in the broad field of radiochemistry, at the graduate or senior undergraduate level, taking into account the degree of previous preparation in physics ordinarily possessed by chemistry students at that level.

We would like to offer definitions of terms, including radiochemistry, nuclear chemistry, tracer chemistry, and radiation chemistry that are heard increasingly today. Unfortunately, the meanings of some of these vary from laboratory to laboratory, and they are hardly used concisely at all. By one group nuclear chemistry is used to mean all applications of chemistry and nuclear physics to each other (including stable-isotope applications). However, to our minds nuclear chemistry emphasizes the reactions of nuclei and the properties of resulting nuclear species, just as organic chemistry is concerned with reactions and properties of organic compounds. We think of tracer chemistry as the field of chemical studies made with the use of isotopic tracers, including studies of the essentially pure tracers at extremely low concentrations. In the title of this book we have meant the term radiochemistry to include all the fields just described, but to exclude stable-isotope tracer applications. Radiation chemistry, which is not discussed in this text, deals with the chemical effects produced by nuclear and other like radiations, and although it involves some of the phenomena of radiochemistry it is really closely related to photochemistry.

Some comments on the order in which the subject matter is presented are perhaps appropriate. We believe that the sequence

of chapters after chapter VI is the logical one; the order of presentation of the material of the first five chapters is much more nearly a matter of individual choice. Our plan, which we have found quite teachable, is to use the historical background as a brief introduction to the concepts and terminology; this makes the going much easier in the succeeding topics. Chapter V actually follows logically after chapter I, and nothing in the arrangement of the material prevents its introduction there if preferred, but we feel that it is more effective first to present further descriptive information about atomic nuclei and nuclear reactions than to confront the student at this point with the quantitative treatment of growth and decay processes.

The development of the subject matter in this book has grown out of an introductory course in radiochemistry, first given in the informal "Los Alamos University" in the latter part of 1945 by the authors (principally G. F.) with the help of Drs. R. W. Dodson and A. C. Wahl, and offered each year since in the Department of Chemistry at Washington University, St. Louis, by one of us (J. W. K.). The formal teaching of radiochemistry to graduate students in chemistry at Washington University is divided into two one-semester courses. In the first course the subject matter of this book is covered as an introduction to radiochemistry; the second semester is a seminar which treats in some detail the applications of radioactivity to chemistry. The subject matter of this second course is covered in the new reference book *Radioactivity Applied to Chemistry*, edited by Prof. Wahl, and is reviewed only very briefly in chapters XI, XII and XIII of our text. The inclusion of these chapters in our book should not be taken as an effort to cover the field of tracer chemistry completely; rather it is meant to provide an introduction to the subject which should prove particularly useful in shorter courses in radiochemistry.

It is a pleasure to express our appreciation for assistance to a number of colleagues who have examined the manuscript and offered many valuable suggestions, including Professors A. C. Wahl, W. S. Koski, and M. D. Kamen; Mr. J. A. Miskel and Dr. N. Elliott, and particularly Prof. R. W. Dodson who in addition has contributed much to our understanding of the subject matter of chapter IX and to its formulation, through his lectures on the Statistical Nature of Radioactivity presented at the Los Alamos Laboratory. We are grateful both to the Brookhaven National

Laboratory and to Washington University for cooperation and hospitality during the writing of the text. We have also valued the assistance of Adrienne Kennedy and Gertrude Friedlander in the preparation of the manuscript and in proof-reading. Professors E. Feenberg, G. T. Seaborg, and E. Segrè were kind enough to look over the proofs and to point out a number of errors.

Throughout the book we have drawn upon the work and ideas of other investigators, but in keeping with its character as a textbook have not attempted to include more than a very few specific references. The references listed at the end of each chapter were selected to call attention to a large number of standard works and to introduce the student to some of the recent literature on specific topics. The exercises given at the end of each chapter are intended as an integral part of the course, and only with them does the text contain the variety of specific examples which we consider necessary for an effective presentation.

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Upton, N. Y.
St. Louis, Mo.
March 1949

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I. NATURALLY OCCURRING RADIOACTIVE SUBSTANCES

A. DISCOVERY OF RADIOACTIVITY

Becquerel's Discovery. The more or less accidental series of events which led to the discovery of radioactivity depended on two especially significant factors: (1) the mysterious X rays discovered about one year earlier by W. C. Roentgen produced fluorescence (the term phosphorescence was preferred at that time) in the glass walls of X-ray tubes, and in some other materials; and (2) Henri Becquerel had inherited an interest in phosphorescence from both his father and grandfather. The father, Edmund Becquerel (1820–1891), had actually studied phosphorescence of uranium salts, and about 1880 Henri Becquerel prepared potassium uranyl sulfate, $K_2UO_2(SO_4)_2 \cdot 2H_2O$, and noted its pronounced phosphorescence excited by ultraviolet light. Thus, in 1895 and 1896 when several scientists were seeking the connection between X rays and phosphorescence and were looking for penetrating radiation from phosphorescent substances, it was natural for Becquerel to experiment along this line with the potassium uranyl sulfate.

It was on February 24, 1896, that Henri Becquerel reported his first results: after exposure to bright sunlight crystals of the uranyl double sulfate emitted a radiation which blackened a photographic plate after penetrating black paper, glass, and other substances. During the next few months he continued the experiments, obtaining more and more puzzling results. The effect was as strong with weak light as with bright sunlight; it was found in complete darkness and even for crystals prepared and always kept in the dark. The penetrating radiation was emitted by other uranyl and also uranous salts, by solutions of uranium salts, and even by what was believed to be metallic uranium, and in each case with an intensity proportional to the uranium content. Proceeding by analogy with a known property of X rays Becquerel observed that the penetrating rays from uranium would discharge an electroscope. These results were all obtained in the early part

of 1896. Although Becquerel and others continued investigations for several years, the knowledge gained in this phase of the new science was summarized in 1898 when Pierre and Marie Sklodowska Curie concluded that the uranium rays were an atomic phenomenon characteristic of the element and not related to its chemical or physical state, and they introduced the name "radioactivity" for the phenomenon.

TABLE I-1

SOME URANIUM AND THORIUM MINERALS

Name	Composition	Uranium Content	Thorium Content	Color and Form
Uraninite (pitchblende)	Uranium oxide, UO_2 to U_3O_8, with rare-earth and other oxides	60–80%	0–10%	Grayish, greenish or brownish black; cubic, amorphous
Thorianite	Thorium and uranium oxides, (Th, U)O_2, with UO_3 and rare-earth oxides	4–40%	30–82%	Gray, brownish or greenish gray, black; cubic
Carnotite	Potassium uranyl vanadate, $K(UO_2)VO_4 \cdot nH_2O$	~45%		Yellow; hexagonal, rhombic
Monazite	Phosphates of the cerium earths and thorium, $CePO_4$ + $Th_3(PO_4)_4$		Up to 16%	Red, brown, yellowish brown; monoclinic
Pilbarite	Thorium lead uranate and silicate	~25%	~25%	Yellow
Autunite	Calcium uranyl phosphate, $Ca(UO_2)_2(PO_4)_2 \cdot 8H_2O$	~50%		Greenish yellow; rhombic
Thorite or orangite	Thorium orthosilicate, $ThSiO_4$		Up to 70%	Brown or black (thorite), orange–yellow (orangite); tetragonal

The Curies. Much new information appeared during the year 1898, mostly through the work of the Curies. Examination of other elements led to the discovery, independently by Mme. Curie

and G. C. Schmidt, that compounds of thorium emitted rays similar to the uranium rays. A very important observation was that some natural uranium ores were even more radioactive than pure uranium, and more active than a chemically similar "ore" prepared synthetically. The chemical decomposition and fractionation of such ores was the first exercise in radiochemistry and led immediately to the discovery of polonium—as a new substance observed only through its intense radioactivity—and of radium, a highly radioactive substance recognized as a new element and soon identified spectroscopically. The Curies and their coworkers had found radium in the barium fraction separated chemically from pitchblende (a dark almost black ore containing 60 to 90 per cent U_3O_8), and they learned that it could be concentrated from the barium by repeated fractional crystallization of the chlorides, the radium salt remaining preferentially in the mother liquor. By 1902 Mme. Curie reported the isolation of 100 mg of radium chloride spectroscopically free from barium and gave 225 as the approximate atomic weight of the element. (The work had started with about two tons of pitchblende, and the radium isolated represented about a 25 per cent yield.) Still later Mme. Curie redetermined the atomic weight to be 226.5 (the 1942 value is 226.05) and also prepared radium metal by electrolysis of the fused salt.

Becquerel in his experiments had shown that uranium, in the dark and not supplied with energy in any known way, continued for years to emit rays in undiminished intensity. E. Rutherford had made some rough estimates of the energy associated with the radioactive rays; the source of this energy was quite unknown. With concentrated radium samples the Curies made measurements of the resulting heating effect, which they found to be about 100 cal per hr per g of radium. The evidence for so large a store of energy not only caused a controversy among the scientists of that time but also helped to create a great popular interest in radium and radioactivity. (An interesting article in the *St. Louis Post-Dispatch* of October 4, 1903, speculated on this inconceivable new power, its use in war and as an instrument for destruction of the world.)

Early Characterization of the Rays. The effect of radioactive radiations in discharging an electroscope was soon understood in terms of the ionization of the air molecules, as J. J. Thomson and

others were developing a knowledge of this subject in their studies of X rays. The use of the amount of ionization in air as a measure of the intensity of radiations was developed into a more precise technique than the photographic blackening, and this technique was employed in the Curie laboratory, where ionization currents were measured with an electrometer. In 1899 Rutherford began a study of the properties of the rays themselves, using a similar instrument. Measurements of the absorption of the rays in metal foils showed that there were two components. One component was absorbed in the first few thousandths of a centimeter of aluminum and was named α radiation; the other was absorbed considerably in roughly 100 times this thickness of aluminum and was named β radiation. For the β rays Rutherford found that the ionization effect was reduced to the fraction $e^{-\mu d}$ of its original value when d centimeters of absorber were interposed; the absorption coefficient μ was about 15 cm^{-1} for aluminum and increased with atomic weight for other metal foils.

Rutherford at that time believed that the absorption of the α radiation also followed an exponential law and gave for it $\mu = 1600$ cm^{-1} in aluminum. About a year later Mme. Curie found that μ was not constant for α rays but increased as the rays proceeded through the absorber. This was a very surprising fact, since one would have expected that any inhomogeneity of the radiation would result in early absorption of the less penetrating components with a corresponding decrease in absorption coefficient with distance. In 1904 the concept of a definite range for the α particles (they were recognized as particles by that time) was proposed and demonstrated by W. H. Bragg. He found that several radioactive substances emitted α rays with different characteristic ranges.

The recognition of the character of the α and β rays as streams of high-speed particles came largely as a result of magnetic and electrostatic deflection experiments. In this way the β rays were seen to be electrons moving with almost the velocity of light. At first the α rays were thought to be undeviated by these fields. More refined experiments did show deflections; from these the ratio of charge to mass was calculated to be about half that of the hydrogen ion, with the charge positive, and the velocity was calculated to be about one-tenth that of light. The suggestion that the α particle was a helium ion immediately arose, and this

was confirmed after much more study. The presence of helium in uranium and thorium ores had already been noticed and was seen to be significant in this connection. A striking demonstration was later made, in which α rays were allowed to pass through a very thin glass wall into an evacuated glass vessel; within a few days sufficient helium gas appeared in the vessel to be detected spectroscopically.

Before the completion of these studies of the α and β rays, an even more penetrating new radiation, not deviated by a magnetic field, was found in the rays from radioactive preparations. The recognition of this γ radiation as electromagnetic waves, like X rays in character if not in energy, came rather soon. For a long time no distinction was made between the nuclear γ rays and some extranuclear X rays which often accompany radioactive transformations.[1]

Rutherford and Soddy Transformation Hypothesis. In the course of measurements of thorium salt activities Rutherford observed that the electrometer readings were sometimes quite erratic. During 1899 it was determined that the cause of this effect was the diffusion through the ionization chamber of a radioactive substance emanating from the thorium compound. Similar effects were obtained with radium compounds. Subsequent studies, principally by Rutherford and F. Soddy, showed that these emanations were inert gases of high molecular weight, subject to condensation at about $-150°C$. Another radioactive substance, actinium, had been separated from pitchblende in 1899, and it too was found to give off an active emanation.

The presence of the radioactive emanations from thorium, radium, and actinium preparations was a very fortunate circumstance for advancement of knowledge of the real nature of radioactivity. Essentially the inert gaseous character of these substances made radiochemical separations not only an easy process, but also one which forced itself on the attentions of these early investigators. Two very significant consequences of the early study of the emanations were: (1) the realization that the activity of radioactive substances did not continue forever but diminished

[1] In the nomenclature of this book concerning radioactive decay processes the term γ rays will include only nuclear electromagnetic radiation; accompanying X rays will be designated as such, even though this is not an entirely uniform practice in the current literature.

in intensity with a time scale characteristic of the substance; and (2) the knowledge that the radioactive processes were accompanied by a change in chemical properties of the active atoms. The application of chemical separation procedures, especially by W. Crookes and by Rutherford and Soddy, in 1900 and the succeeding years, revealed the existence of other activities with characteristic decay rates and radiations, notably uranium X which is separated from uranium by precipitation with excess ammonium carbonate (the uranyl carbonate redissolves in excess carbonate through formation of a complex ion), and thorium X which remains in solution when thorium is precipitated as the hydroxide with ammonium hydroxide. In each case it was found that the activity of the X body decayed appreciably in a matter of days, and that a new supply of the X body appeared in the parent substance in a similar time. It was also shown that both uranium and thorium, when effectively purified of the X bodies and other products, emitted only α rays, and that uranium X and thorium X emitted β rays.

By the spring of 1903 Rutherford and Soddy had reached an excellent understanding of the nature of radioactivity and published their conclusions that the radioactive elements were undergoing spontaneous transformation from one chemical atom into another, that the radioactive radiations were an accompaniment of these changes, and that the radioactive process was a subatomic change within the atom. However, it should be remembered here that the idea of the atomic nucleus did not emerge until eight years later and that in 1904 Bragg was attempting to understand the α particle as a flying cluster of thousands of more or less independent electrons.

Statistical Aspect of Radioactivity. In 1905 E. v. Schweidler used the foregoing conclusions as to the nature of radioactivity and formulated a new description of the process in terms of disintegration probabilities. His fundamental assumptions were that the probability p for a particular atom of a radioactive element to disintegrate in a time interval Δt is independent of the past history and the present circumstances of the atom; it depends only on the length of the time interval Δt and for sufficiently short intervals is just proportional to Δt; thus $p = \lambda \Delta t$, where λ is the proportionality constant characteristic of that species of radioactive atoms. The probability of the given atom not disinte-

grating during the short interval Δt is $1 - p = 1 - \lambda\Delta t$. If the atom has survived this interval, then its probability of not disintegrating in a second like interval is again $1 - \lambda\Delta t$. By the law for compounding such probabilities the probability for the given atom to survive the first interval and also the second is given by $(1 - \lambda\Delta t)^2$; for n such intervals this survival probability is $(1 - \lambda\Delta t)^n$. Setting $n\Delta t = t$, the total time, we have $\left(1 - \lambda\frac{t}{n}\right)^n$. Now the probability that the atom will remain unchanged after time t is just the value of this quantity when Δt is made indefinitely small; that is, it is the limit of $\left(1 - \lambda\frac{t}{n}\right)^n$ as n approaches infinity. Recalling that $e^x = \lim_{n\to\infty}\left(1 + \frac{x}{n}\right)^n$, we have $e^{-\lambda t}$ for the limiting value. If we consider not one atom, but a large initial number N_0 of the radioactive atoms, then the fraction remaining unchanged after time t we may take to be $N/N_0 = e^{-\lambda t}$, where N is the number of unchanged atoms at time t. This exponential law of decay is just that which had already been found experimentally for the simple isolated radioactivities.

Chapter IX will present a more detailed discussion of the statistical nature of radioactivity.

B. RADIOACTIVE DECAY AND GROWTH

In the preceding section we mentioned that the decay of a radioactive substance followed the exponential law $N = N_0e^{-\lambda t}$, where N is the (large) number of unchanged atoms at time t, N_0 is the number present when $t = 0$, and λ is a constant characteristic of the particular radioactive species. This will be recognized as the rate law for any monomolecular reaction, and, of course, this should be expected in view of the nature of the radioactive process. It may be derived if the decay rate, $-dN/dt$, is set proportional to the number of atoms present: $-dN/dt = \lambda N$. (This is to say that we expect twice as many disintegrations per unit time in a sample containing twice as many atoms, etc.) On integration the result is $\ln N = -\lambda t + a$, and the constant of integration a is evaluated from the limit $N = N_0$ when $t = 0$: $a = \ln N_0$. Combining these terms we have: $\ln N/N_0 = -\lambda t$, or $N/N_0 = e^{-\lambda t}$.

The constant λ is known as the decay constant for that radioactive species. As may be seen from the differential equation, it is the fraction of the number of atoms transformed per unit time, provided the time unit is chosen short enough so that only a small fraction of the atoms transform in that interval. In any case λ has the dimensions of a reciprocal time and is most often expressed in sec^{-1}. It is to be noticed that no attempt to alter λ through variation of ordinary experimental conditions, such as temperature; chemical change; pressure; gravitational, magnetic or electric fields; has ever given a detectable effect.[2]

The characteristic rate of a radioactive decay may very conveniently be given in terms of the half-life $t_{1/2}$, which is the time required for an initial (large) number of atoms to be reduced to half that number by transformations. Thus, at the time $t = t_{1/2}$ $N = N_0/2$, and:

$$\ln \tfrac{1}{2} = -\lambda t_{1/2}, \quad \text{or} \quad t_{1/2} = \frac{\ln 2}{\lambda} = \frac{0.69315}{\lambda}.$$

In practical work with radioactive materials the number of atoms N is not directly evaluated, and even the rate of change dN/dt is usually not measured absolutely. The usual procedure is to determine, through its electric, photographic, or other effect, a quantity proportional to λN; we may term this quantity the activity $\mathbf{A}$, with $\mathbf{A} = c\lambda N = c(-dN/dt)$. The coefficient c, which we may term the detection coefficient, will depend on the nature of the detection instrument, the efficiency for the recording of the particular radiation in that particular instrument, and the geometrical arrangement of sample and detector; a usual feature of the experimentation is careful precaution to keep all these factors under control. We may now write the decay law as it is commonly observed, $\mathbf{A} = \mathbf{A}_0 e^{-\lambda t}$.

The usual procedure for the treatment of data measuring $\mathbf{A}$ at successive times is to plot $\log \mathbf{A}$ vs. t; for this purpose semilog paper (with a suitable number of decades) is most convenient. Now λ could be found from the slope of the resulting straight line corresponding to the simple decay law; however, in this procedure there is a possibility for the confusion of units or of different logarithm bases. It is more convenient to read from the plot on

[2] A possible exception is mentioned in chapter VI, page 137, footnote 4.

semilog paper the time required for the activity to fall from any value to half that value; this is the half-life $t_{1/2}$.

In this discussion we have considered only the radioactivity corresponding to the transformation of a single atomic species; however, the atom resulting from the transformation may itself be radioactive, with its own characteristic radiation and half-life, as well as its own chemical identity. Indeed among the naturally occurring radioactive substances this is the more common situation, and eventually (in chapter V) we must treat quite complicated interrelated radioactive growths and decays. For the moment consider the decay of the substance uranium I, or U_I. This species of uranium is an α-particle emitter with $t_{1/2} = 4.51 \times 10^9$ years. The immediate product of its transformation is the radioactive substance uranium X_1, or UX_1, a β emitter with half-life 24.6 days. For this pair of substances, the parent uranium may be separated from the daughter atoms by precipitation of the daughter with excess ammonium carbonate, as already mentioned. The daughter precipitate will show a characteristic activity, which will decay with the rate indicated; that is, it will be half gone in 24.6 days, three fourths gone in 49.2 days, seven eighths gone in 73.8 days, etc. The parent fraction will, of course, continue its α activity as before, but will for the moment be free of the β radiations associated with the daughter. However, in time new daughter atoms will be formed, and the daughter activity in the parent fraction will return to its initial value with a time scale corresponding to the rate of decay of the isolated daughter fraction.

In an undisturbed sample containing N_1 atoms of U_I, a steady state is established in which the rate of formation of the daughter UX_1 atoms (number N_2) is just equal to their rate of decay. This means that $-dN_1/dt = \lambda_2 N_2$ in this situation, because the rate of formation of the daughter atoms is just the rate of decay of the parent atoms. Using the earlier relation we have then, $\lambda_1 N_1 = \lambda_2 N_2$, with λ_1 and λ_2 the respective disintegration constants. This is sometimes more convenient in terms of the two half-lives: $N_1/(t_{1/2})_1 = N_2/(t_{1/2})_2$. This state of affairs is known as secular equilibrium. No account is taken of the decrease of N_1 with time, since the fraction of U_I atoms transformed even throughout the life of the experimenter is completely negligible. In general, wherever a short-lived daughter results from the decay

of a very long-lived parent, this situation exists. The same relation, $\lambda_1 N_1 = \lambda_2 N_2 = \lambda_3 N_3$, etc., may be applied when several short-lived products arise from successive decays beginning with a long-lived parent, provided again that the material has been undisturbed (that is, no daughter substances removed or allowed to escape) for a long enough time for secular equilibrium to be established.

The concept of secular equilibrium suggests a convenient way to handle experimental data concerned with the rate of growth of a short-lived daughter substance in a freshly separated long-lived parent fraction. Because all the rates of decay are entirely independent of the chemical manipulations in the separation (say of UX_1 from U_I), the sum of the amounts of daughter UX_1 in the two fractions always continues at the constant value given by $\lambda_1 N_1 = \lambda_2 N_2$. Thus by the time the isolated daughter preparation is practically inactive the growth in the parent will have practically re-established the secular equilibrium condition. If measurements of the amount of daughter activity in the parent fraction are obtained as a function of the time t, then these activity values may be subtracted from the final value approached as t becomes long compared to $(t_{\frac{1}{2}})_2$, and the differences plotted on semilog paper vs. t to give a straight line like a decay curve. In fact this curve describes the decay of the isolated daughter fraction. In this way the daughter half-life may be obtained from its rate of growth in a very long-lived parent. (It may be useful here to emphasize that a plot directly of the growing daughter activity in the parent fraction, on either a linear or semilog basis, cannot give a straight line and in either case gives a curve approaching the secular equilibrium value as an asymptote. See figure V–3, page 113.)

C. RADIOACTIVE SERIES

Uranium, Thorium, and Actinium Series. All elements found in natural sources with atomic number greater than 83 (bismuth) are radioactive. They belong to chains of successive decays, and all the species in one such chain constitute a radioactive family or series. Three of these families include all the natural activities in this region of the periodic chart. One has U_I (mass 238 on the atomic weight scale) as the parent substance, and after 14 trans-

formations (8 of them by α-particle emission and 6 by β-particle emission) reaches a stable end product, radium G (lead with mass 206); this is known as the uranium series. (This series includes radium and its decay products; these are sometimes called the radium series.) Since the atomic mass is changed by four units in α decay and changed much less than one unit by β decay, the various masses found in members of the family differ by multiples of 4, and a general formula for the approximate masses is $4n + 2$, where n is an integer. Therefore, the uranium series is known also as the $4n + 2$ series. Figure I–1 shows the members and transformations of the uranium series. The existence of branching decays should be noticed; it is almost certain that very many more branchings would be found if sufficiently sensitive means of recognizing them were available.

Thorium (mass 232) is the parent substance of the $4n$ series, or thorium series, with lead of weight 208 as the stable end product. This series is shown in figure I–2. The $4n + 3$, or actinium, series has actino-uranium, AcU (uranium of mass 235), as the parent and lead of mass 207 as the stable end product. This series is shown in figure I–3.

The fairly close similarity of the three series to each other and in their relations to the periodic chart is interesting and helpful in remembering the decay schemes and nomenclature for the active bodies. Actually, these historical names may some day become obsolete, and the designations of chemical element and atomic mass become standard; already we are more familiar with U^{238}, U^{235}, and U^{234} than with U_I, AcU, and U_{II}. (This trend is favored by the fact that names like UX_1 and RaD do not immediately suggest that these substances are chemically like thorium and lead, respectively; also, in some of the early literature the nomenclature is different from current usage, which leads to some confusion. On the other hand, many of the historical names like RaA, RaB indicate immediately positions in the decay chain; and, further, the name Pa^{234} would not distinguish between UX_2 and UZ.)

The $4n + 1$ Series and Synthetic Transuranium Elements; Additional Natural Activities. In past years much comment has been given to the failure to find in nature a $4n + 1$ radioactive series; the most plausible presumption has been that no member of this series was sufficiently long-lived to survive the many years since

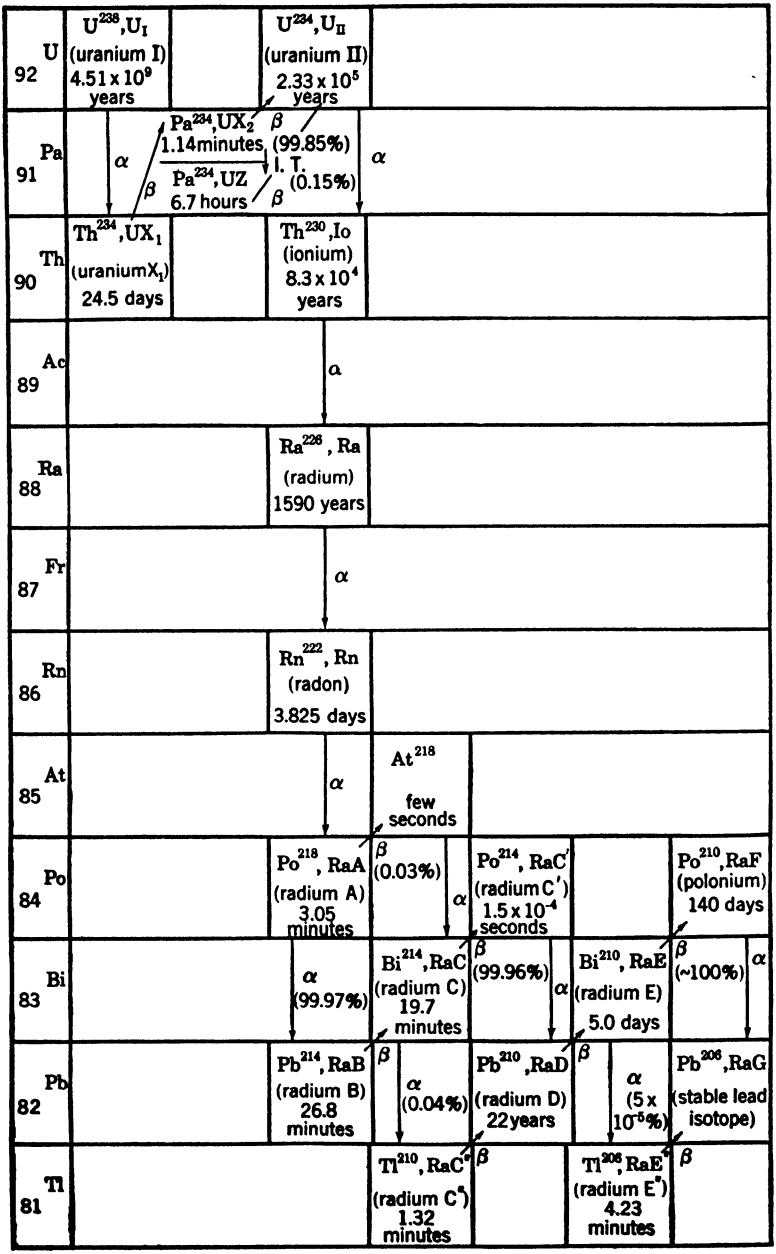


FIGURE I–1. The uranium series.

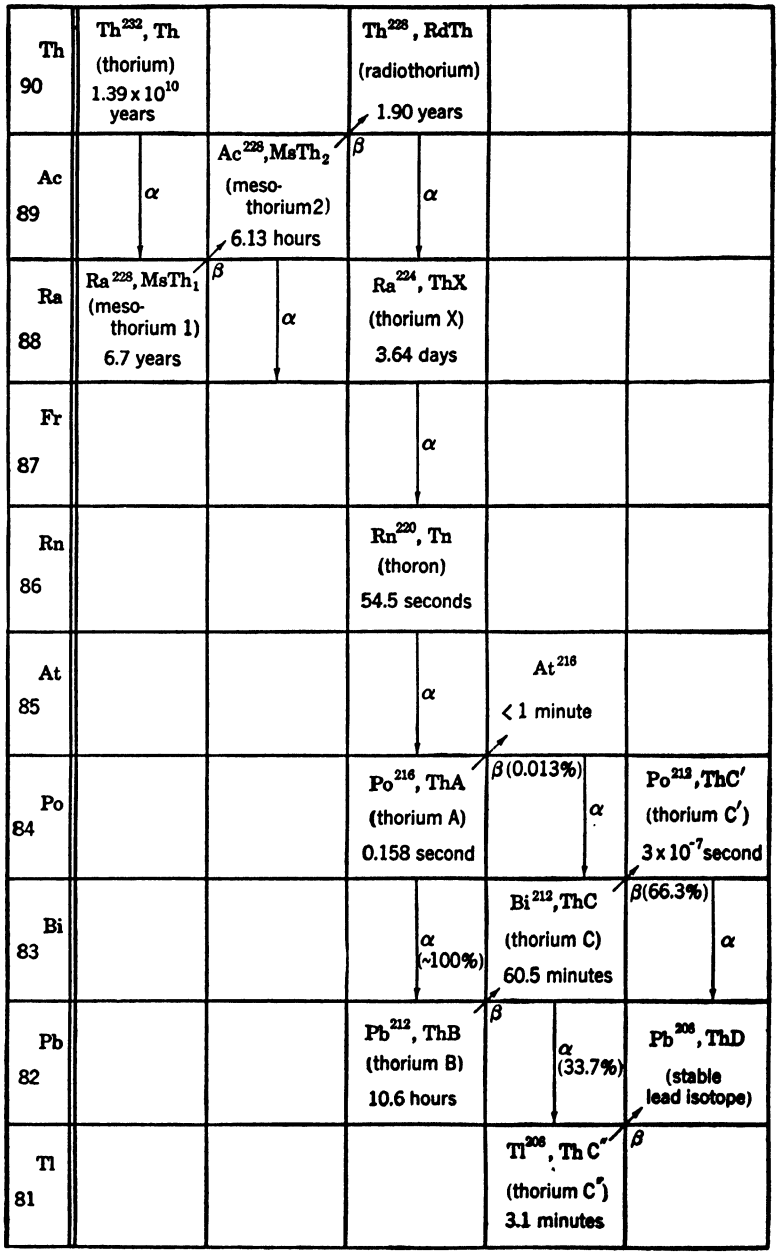


FIGURE I–2. The thorium series.

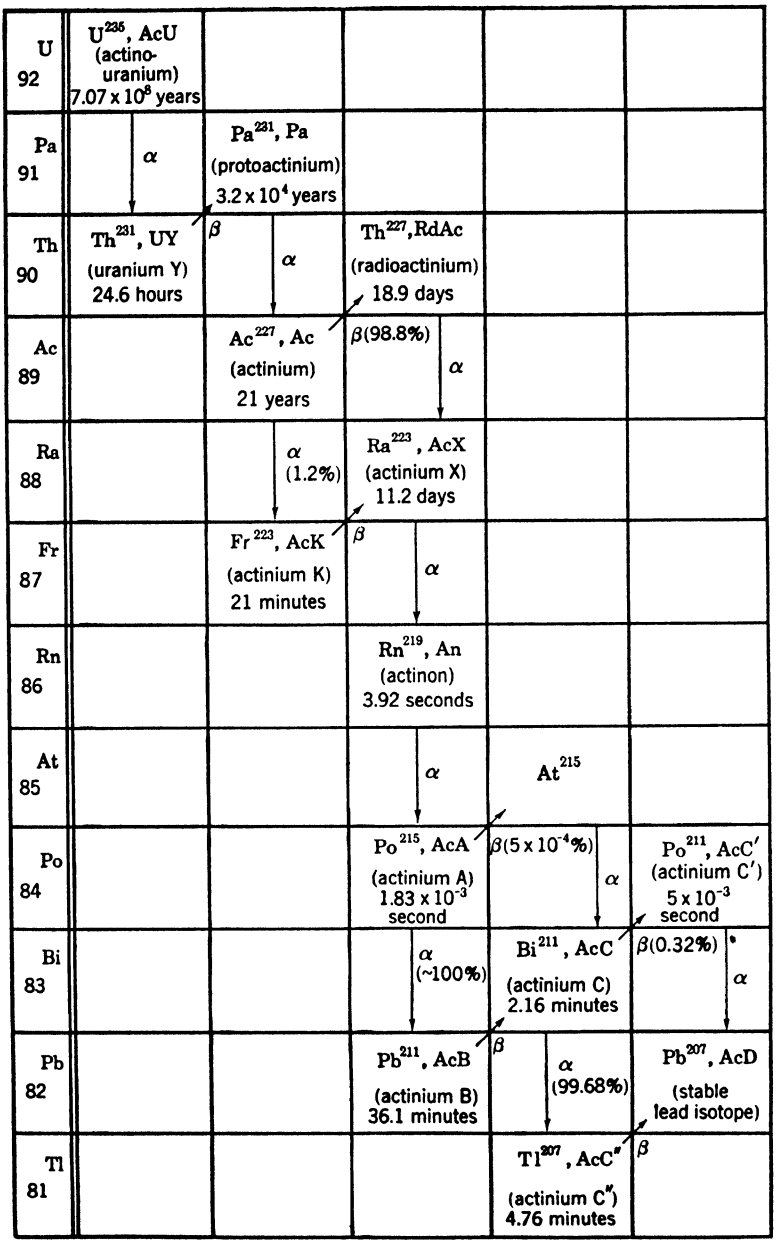


FIGURE I-3. The actinium series.

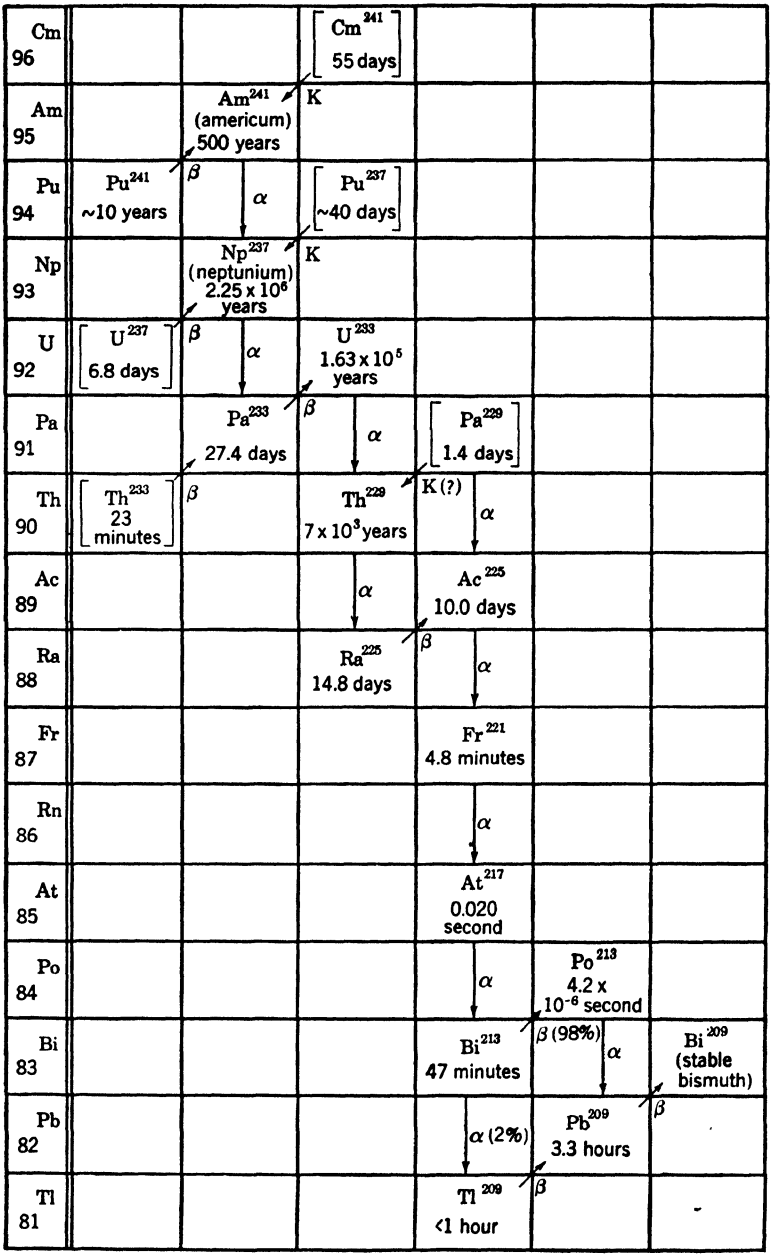


FIGURE I–4. The $4n + 1$ series.

it might have been formed. In the same way there has been speculation as to heavier members of the known families, with the assumption that the half-lives of any transuranium elements were short compared to geologic time. By recent artificial transmutation techniques a rather well-developed $4n + 1$ series including several transuranium species has been prepared and investigated. This series is displayed in figure I–4. The properties of several other transuranium species may be found in table A in the appendix.

Several investigators at one time or another have examined essentially all the remaining known elements for evidences of naturally occurring radioactivity, and with some positive results, first in the case of potassium and later in a number of others. The properties of the known natural radioactivities other than those of the three radioactive families are collected in table I–2.

TABLE I-2

ADDITIONAL NATURALLY OCCURRING RADIOACTIVE SUBSTANCES

Active Substance	Type of Disintegration	Half-life	Relative Isotopic Abundance	Stable Disintegration Products
K^{40}	β, K	4.5×10^8 years	0.012%	Ca^{40}, A^{40}
Rb^{87}	β	6.0×10^{10} years	27.2%	Sr^{87}
Sm^{152}	α	2.5×10^{11} years	26.6%	Nd^{148}
Lu^{176}	β, K	2.4×10^{10} years	2.5%	Hf^{176}, Yb^{176}
Re^{187}	β	4×10^{12} years	62%	Os^{187}

It may be seen that the samarium radioactivity is the only case of α disintegration; the type of decay listed as K is discussed in chapter VI, section B. In attempts to extend the search for new radioactivities to very low intensity levels difficulty arises from the general background of detectable radiations present in every laboratory. In part this general background is due to presence of traces of uranium, thorium, potassium, etc., and in large part to the cosmic radiation of unknown origin. The cosmic rays reach every portion of the earth's surface; their intensity is greater at high altitudes but persists measurably even in deep caves and mines. The magnitude of the background effect is indicated in the discussion of radiation-detection instruments in chapter VIII, page 193.

Age of the Earth. As already suggested, the existence of the radioactive substances uranium and thorium gives some information concerning the time that may have passed since the genesis of these elements, and perhaps of all elements. Clearly conditions as we know them today cannot have existed for a time very long compared to the half-lives, 4.51×10^9 years, 7.07×10^8 years, and 1.39×10^{10} years for U_I, AcU, and Th, respectively. Minerals have been studied with the object of determining their age in relation to radioactive constants by at least four methods; these are discussed in the following paragraphs.

1. *Intensity of Coloration of Pleochroic Haloes.* Many types of radiation are capable of producing coloration or discoloration in glass, quartz, mica, and a number of similar materials. Intense sources of α particles are effective in producing colorations in a short time, and even exceedingly small amounts of uranium or thorium are capable of producing visible effects within geological time intervals when present as a minute inclusion in a mineral such as mica. The range of coloration from α particles is of the order of a few thousandths of a centimeter in mica, and the characteristic α-particle ranges for the various decay products cause the production of tiny concentric shells of varying coloration. Examined in thin sections under a microscope, these appear as circular areas known as pleochroic haloes; in polarized light the colors change with the plane of polarization. The many observed radii of the color bands have been fairly well correlated with the known α-particle ranges in mica of decay products of uranium or thorium, and in this way the nature of the inclusion is established. The amount of inclusion may be judged roughly from its size in the microscope field. Attempts have been made to evaluate the amount of radiation required for a particular degree of coloration and thus to establish a geologic time scale for these mica samples. Effects such as reversal of the intensity of coloration caused by overexposure (analogous to photographic solarization) must be taken into account, and no accurate results can be claimed.

2. *Ratio of Uranium to Helium Content.* Once an atom of U_I disintegrates, the chain of successive decays soon (say in less than about a million years) produces eight α particles. Because the ranges of these particles are very short in dense matter, most of the resulting helium atoms (the helium ions at rest are easily capable of acquiring two electrons by oxidizing almost any sub-

stance) may be trapped in the interior of the rock. In favorable cases, with very impervious fine-grained rocks and a low helium concentration (pressure) from small uranium contents, this helium has been retained throughout the geologic ages and now serves as an indicator of the fraction of uranium transformed since the formation of the ore. The thorium content of the rock also is a source of helium, and this must be taken into account. Very sensitive methods of assay for helium, uranium, and thorium are available and have permitted determinations on rocks with uranium and thorium contents below 1 part per million, and on metallic iron meteorites (where loss of helium in any process short of melting seems quite unlikely). The ages found, usually to be taken as lower limits, range up to a little over 2000 million years. For some meteorites larger values have been found; however, evidence has been presented recently which suggests that a considerable part of the helium in meteorites may have resulted from the action of cosmic radiation and that some meteorites may, therefore, be considerably younger than indicated.

3. *Ratio of Uranium or Thorium to* RaG *or* ThD *Content.* Lead (RaG and ThD) is a stable end product of the disintegration of uranium and thorium and provided there is no other source of lead in an ore may be used as a quantitative indicator of disintegration. This lead method might be expected to be more reliable than the helium method since lead is not so likely to have been lost by slow diffusion; however, it is still quite possible that a lead–uranium or lead–thorium ratio has been changed by leaching or some other process. The distinction between these lead decay products (Pb^{206} from U_I and Pb^{208} from Th) and ordinary lead is made in a satisfactory way by mass spectrographic analysis; it is usually presumed that absence of Pb^{204} establishes the absence of ordinary lead. Age determinations from uranium-to-Pb^{206} and from thorium-to-Pb^{208} ratios do not always agree, but values ranging up to roughly 3000 million years are indicated.

4. *Ratio of Uranium Lead to Actinium Lead.* Probably the best method so far devised involves determination of the ratio of Pb^{206} to Pb^{207}. This method should be free of many experimental errors and is less sensitive to chemical or mechanical loss of either uranium or lead than method 3. The Pb^{206}/Pb^{207} ratio is an indicator of age because U_I and AcU decay at different rates. From mass spectrographic measurements of this ratio a convinc-

ing age scale of minerals can be established. Samples of two ores from a region known to be geologically very old (Huron Claim monazite and uraninite) have ages close to 2000 million years.

These times of about 2×10^9 years are of the same order as the "age of the universe" estimated from the red-shift phenomenon. (This disputed hypothesis assigns to each star system a velocity which would give the observed shift towards the red of all spectral lines as a Doppler effect; the result is an exploding universe model with the "age" as the time calculated for the systems to have achieved their present separation distances starting from the point of common origin.)

It may be noted that studies of relative abundances of radioactive species yield some other information on ancient time scales. It has been carefully demonstrated that the ratio of AcU to U_I is the same (1:139) for uranium samples from various sources; this is plausible if all our uranium had been formed at the same time; even if it had been formed in the same way at different times, the different rates of decay would leave in the younger samples more of the shorter-lived component. In the same way, the concentration of the radioactive K^{40} in potassium from various terrestrial and meteoric sources is constant, giving evidence for a common genesis of elements in our solar system. More careful attention to the distribution of isotopes, especially in meteorites, may give new knowledge of the time between genesis of the elements and formation of the discrete rock phases, since any comparatively short-lived element naturally included in one phase could give rise later by radioactive transformation to an "unnatural" inclusion in that phase. In considerations of this kind it may not be justified to neglect nuclear reactions of other types; for example it has been suggested that an era of high neutron flux may have at one time considerably upset the pattern of elementary and isotopic abundances.

Quite recently it has been demonstrated that one action of neutrons associated with the cosmic radiation is the continuing production of radioactive carbon (C^{14}), mostly in the upper regions of the earth's atmosphere. The half-life of about 5000 years for this substance is no doubt short compared to the duration of the cosmic irradiation; therefore, we may expect a steady-state concentration (analogous to the situation in secular equilibrium)

of this radioactivity in all carbon of the living carbon cycle. It may be possible to date archeological objects, bones, mummies and the like, through measurements of the level of carbon radioactivity, as decreased by radioactive decay since the specimen was removed from participation in the life cycle.

EXERCISES

1. One hundred milligrams of Ra would represent what percentage yield from exactly 2 tons of a pitchblende ore containing 75 per cent U_3O_8? *Answer:* 26 per cent.

2. Calculate the rate of energy liberation (in calories/hr) for 1.00 g of pure radium free of its decay products. What can you say about the actual heating effect of an old radium preparation?

Answer to first part: 25 cal/hr.

3. A certain active substance (which has no radioactive parent) has a half-life of 8.0 days. What fraction of the initial amount will be left after (*a*) 16 days, (*b*) 32 days, (*c*) 4 days, (*d*) 83 days? *Answer:* (*a*) 0.25.

4. How long would a sample of radium have to be observed before the decay amounted to 1 per cent? (Neglect effects of radium A, B, C, etc., on the detector.)

5. Find the number of disintegrations of uranium I atoms occurring per minute in 1 mg of ordinary uranium, from the half-life of U_I, $t_{1/2} = 4.51 \times 10^9$ years.

6. How many β disintegrations occur per second in 1.00 g of pitchblende containing 70 per cent uranium? You may assume that there has been no loss of radon from the ore. *Answer:* 53,000 per sec.

7. Estimate the age of a rock which is found to contain 5×10^{-5} cc of helium at standard temperature and pressure and 3×10^{-7} g of uranium per gram. *Answer:* 1.2×10^9 years.

8. By artificial transmutation techniques a number of new radioactive species have been produced in the region of the natural radioactive families. These may be classified into the four families according to mass and are known as collateral members of the families. Find all of these in table A in the appendix and place them in figures I–1, I–2, etc., so as to indicate their decay relationships to other members of the series.

9. Calculate the age of a mineral which shows a Pb^{206}-to-Pb^{207} ratio of 14.0 and is essentially free of Pb^{204}. *Answer:* 1×10^9 years.

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II. ATOMIC NUCLEI

A. ATOMIC STRUCTURE

Rutherford's Nuclear Model of the Atom. At the time the phenomenon of radioactivity was discovered the chemical elements were regarded as unalterable; that is, they were thought to retain their characteristic properties throughout all chemical and physical processes. This view became untenable when it was recognized that radioactive processes involved the disintegration of elements and the formation of other elements. Thus it became clear about 1900 that atoms, until then regarded as the indivisible building blocks of the elements, must have some structure and that it must be possible for the atoms of one element to be transformed into those of another, with the emission of radiations. However, it was not until 1911 that the nuclear model of the atom which is now generally accepted was proposed by Rutherford.

Rutherford was led to propose this model by experimental results on the scattering of α particles in matter. He discovered the scattering phenomenon when he noticed that a collimated beam of α particles was spread out in passing through a thin layer of matter. A quantitative study by H. Geiger of the scattering in very thin foils showed that single encounters between α particles and atoms resulted in scattering of the α particles through very small angles. On the basis of this information the probability of larger scattering angles resulting from multiple scattering processes in thicker foils could be calculated statistically, and this probability falls off very rapidly with increasing angle. When experiments of Geiger and E. Marsden showed that scattering angles of 90° or more were much more frequent than could be accounted for by multiple scattering, Rutherford proposed that these large angles were due to a special type of single scattering process.

To explain the scattering of α particles through large angles in single processes Rutherford postulated an atomic model in which the positive charge resides in a small massive nucleus and negative charge of the same magnitude is distributed over a sphere of

atomic dimensions. The large deflection of an α particle from its path was then supposed to result from Coulomb repulsion between the α particle and the positively charged nucleus of an atom. With this simple assumption and the additional restriction that the nucleus is so heavy as to be considered at rest during the impact Rutherford set up the conditions for conservation of momentum and energy and derived from these his famous scattering formula,

$$n(\theta) = n_0 \cdot \frac{Nd}{r^2}\left(\frac{Ze^2}{M_\alpha v_\alpha{}^2}\right)^2 \cdot \frac{1}{\sin^4 \frac{\theta}{2}},$$

where $n(\theta)$ is the number of scattered α particles falling on a unit area at a distance r from the scattering point when the angle between the directions of the initial and scattered α particles is θ; n_0 is the incident number of particles, d the thickness of the scatterer, N the number of nuclei per unit volume of scatterer and Ze the charge per nucleus. M_α and v_α are the mass and initial velocity of the α particle.

Rutherford's hypothesis thus predicted that the number of scattered particles per unit area was proportional to the thickness of the scatterer and to the square of the nuclear charge, and inversely proportional to the square of the α-particle energy and to the fourth power of the sine of half the scattering angle. The number of scattered particles has been carefully measured as a function of scattering angle, α-particle energy, and thickness of scatterer, and the results have been found to be in excellent agreement with the Rutherford formula provided heavy elements were used as scatterers. For light scatterers, that is, for the case where the nuclei of the scatterer cannot be considered at rest during the impact, a more complicated expression must be substituted for the Rutherford formula, and if this is done the agreement between theory and experiment is again satisfactory.

This experimental verification of the scattering formula led to a general acceptance of Rutherford's picture of the atom as consisting of a small positively charged nucleus, containing practically the entire mass of the atom, and surrounded by a distribution of negatively charged electrons. In addition, the scattering law made it possible to study the magnitude of the nuclear charge in the atoms of a given element, because the scattering intensity

depends on the square of the nuclear charge. It was by the method of α-particle scattering that nuclear charges were first determined, and this work led to the suggestion that atomic number was identical with the nuclear charge (expressed in units of the electronic charge e). This suggestion was subsequently confirmed by H. G. Moseley's work on the X-ray spectra of the elements.

Bohr's Theory of Electron Orbits. In the preceding section we have seen that according to Rutherford's hypothesis an atom consists of a small positively charged nucleus and a "cloud" of electrons surrounding it. Since the charge on the nucleus is an integral multiple Z of the electronic charge e, the number of electrons surrounding the nucleus of a neutral atom must also be equal to Z. This number Z is known as the atomic number.

Classical mechanics and electrodynamics cannot account for the stability of a system consisting of a heavy positive nucleus surrounded by moving electrons. According to classical theory such an atom would lose energy continuously because the electrons, being accelerated in the Coulomb field of the nucleus, would emit electromagnetic radiation. In 1913 N. Bohr introduced a quantum theory of atomic structure. He postulated that an atom could exist only in certain discrete energy states corresponding to particular circular orbits of the electrons around the nucleus, and that it could lose or gain energy only in transitions from one of these quantum states to another. The monochromatic radiation absorbed or emitted in such a transition should then be related to the energy difference ΔE between the two energy states by the quantum relation $\Delta E = h\nu$, where ν is the frequency of the radiation and h is Planck's constant. In considering the simplest atom, that of hydrogen, Bohr found he could obtain good agreement with observed frequencies in the hydrogen spectrum if he made the assumption that the electron was restricted to those orbits whose angular momenta were whole multiples of $h/2\pi$.

The angular momentum of an electron of mass m and velocity v traveling in a circular orbit of radius a is mva, and Bohr's quantum condition is, therefore, given by

$$mva = \frac{nh}{2\pi}, \tag{II-1}$$

where n is an integer. An additional condition, which follows from classical mechanics, is that the centripetal force due to the Coulomb attraction between electron and nucleus must equal the centrifugal force due to the electron's motion in its orbit. This condition is expressed by

$$\frac{Ze \cdot e}{a^2} = \frac{mv^2}{a}. \tag{II-2}$$

By solving both equations II–1 and II–2 for v and equating the expressions we get

$$a = \frac{n^2h^2}{4\pi^2 mZe^2}. \tag{II-3}$$

Thus, the radius of each Bohr orbit of the electron in the hydrogen atom is characterized by the so-called principal quantum number n. In the lowest or normal energy state of the hydrogen atom, $n = 1$, or the radius of the electron orbit is $h^2/4\pi^2 mZe^2 \approx 5.3 \times 10^{-9}$ cm.

Bohr's original theory was very successful in accounting for the main features of the hydrogen spectrum. However, the splitting of many of the hydrogen lines into several components could not be explained on this basis, and no quantitative agreement was obtained for the spectra of more complex elements. In the years following 1913 Bohr's theory underwent a series of refinements. The motion of the nucleus was taken into account through replacement, in Bohr's equations, of the electron mass m by the reduced mass of the system $\frac{Mm}{M+m}$, where M is the mass of the nucleus. A. Sommerfeld generalized the theory by introducing elliptical as well as circular orbits, with the nucleus at one focus. This introduced a second quantum condition restricting the eccentricities of the ellipses to certain allowed values. A third quantum condition allowing only a limited number of orientations of an atom in an external magnetic field was found to explain the Zeeman effect, that is, the splitting of spectral lines into several closely spaced components under the influence of a magnetic field. Finally the concept of electron spin was introduced, and the condition that an electron can have its spin-momentum vector oriented either parallel or antiparallel to its

orbital angular momentum vector represents a fourth quantum condition.

Summarizing we can say that each electron orbit in an atom is characterized by four quantum numbers:

1. The principal quantum number n (related to the average distance from the nucleus).
2. The azimuthal quantum number l (related to the orbital angular momentum).
3. The magnetic quantum number m_l.
4. The spin quantum number m_s.

It turns out that these quantum numbers are not independent of each other, but that for each value of n only certain values of the other quantum numbers are possible: for any n, l can have any integral value from 0 to $n - 1$; for any l, m_l can have any integral value from $-l$ to $+l$ including 0; and m_s can be either $-\frac{1}{2}$ or $+\frac{1}{2}$.

As an illustration the possible combinations of quantum numbers for $n = 1$ and $n = 2$ are given in table II–1. Also included

TABLE II–1

POSSIBLE COMBINATIONS OF QUANTUM NUMBERS FOR $n = 1$ AND $n = 2$

n	l	m_l	m_s	Energy Level Designation
1	0	0	$+\frac{1}{2}$ or $-\frac{1}{2}$	$1s$
2	0	0	$+\frac{1}{2}$ or $-\frac{1}{2}$	$2s$
2	1	-1	$+\frac{1}{2}$ or $-\frac{1}{2}$	$2p$
2	1	0	$+\frac{1}{2}$ or $-\frac{1}{2}$	$2p$
2	1	$+1$	$+\frac{1}{2}$ or $-\frac{1}{2}$	$2p$

are the designations of the orbits or energy states used in discussions of atomic structure and spectroscopy. The states corresponding to $l = 0, 1, 2, 3$ are referred to as s, p, d, f states, respectively, and the numerical value of n precedes the letter. Another terminology, taken over from X-ray spectroscopy, refers to the electron orbits characterized by $n = 1, 2, 3, 4, 5, 6$, etc., as the K, L, M, N, O, P, etc., shells.

Building Up of the Periodic Table. It follows from the exclusion principle formulated by W. Pauli in 1925 that in a system such as an atom no more than one electron can be in an energy state characterized by the same values of the four quantum numbers

n, l, m_l, and m_s. This principle, together with the very plausible assumption that the normal state of an atom always corresponds to the electron configuration of lowest energy, determines the distribution of electrons in the quantum levels of any given atom. In hydrogen the electron is normally in the orbit characterized by $n = 1$, $l = 0$ (a $1s$ orbit); in helium the two electrons are both in the $1s$ orbit but with opposite spin orientations. In lithium two electrons are again in the $1s$ orbit, but the third one has to go to the next higher energy state, the $2s$ state ($n = 2$, $l = 0$). In beryllium the $1s$ and $2s$ orbits are filled, from boron to neon the six $2p$ electrons are being added, in sodium the eleventh electron goes into the $3s$ orbit, etc. An irregularity occurs at potassium ($Z = 19$) where one might expect the nineteenth electron to go to the $3d$ orbit ($1s$, $2s$, $2p$, $3s$, $3p$ being filled), but actually the nineteenth electron in the normal atom appears in the $4s$ state. Only after the second $4s$ electron is also added (in calcium), do the $3d$ orbits fill (from scandium to copper). A similar situation occurs between rubidium and silver. These irregularities in the filling of the electron shells can be understood qualitatively in terms of the different eccentricities of the various orbits in a given shell. For any given n the orbits become increasingly eccentric with decreasing l; the largest value of l always corresponds to a circular orbit. Thus, although an electron in a $4s$ orbit is on the average further away from the nucleus than a $3d$ electron, its orbit penetrates inside some of the inner shells, while the $3d$ electron is rather well shielded from the positively charged nucleus by the inner electrons. Thus, it is plausible that in a particular atom an electron in a $4s$ orbit may actually have a somewhat lower potential energy than it would have in a $3d$ orbit. Such qualitative arguments as this are borne out by approximate quantitative calculations of the electron energies in the various orbits. The distribution of electrons among the quantum states can now be reasonably well calculated for the lowest state of most elements in the periodic table, and the results relate remarkably well to much of the chemical behavior of the elements. Thus, to name only a few examples, the similarity between elements in the same column of the periodic table can now be ascribed to the similarity in their electron structures in the outermost shells; valence can be correlated with the number of electrons in the outer shell; the striking similarity between the fourteen rare

earths can be accounted for by the fact that they differ only in the number of electrons in the $4f$ shell (which is well inside the atom) while the population of the $5s$, $5p$, $5d$, and $6s$ shells is practically the same for all of them.

Wave Mechanics. So far we have discussed atomic structure in terms of the picture introduced by Bohr in 1913. Actually this has been superseded by another approach to the problem which may be harder to visualize, but which is of much wider applicability and gives results that are quantitatively in better accord with experimental observations. This is the wave-mechanical approach developed by E. Schroedinger.

In 1923 L. de Broglie had pointed out that Bohr's quantum condition (equation II–1) followed directly from the assumptions that an electron of mass m and velocity v had associated with it a phase wave of wave length $\lambda = h/mv$ and that the orbit circumference must be an integral multiple of λ. In pursuing further the idea of wave properties associated with material particles Schroedinger arrived at his wave equation for the representation of material systems. In solving this equation for the hydrogen atom one finds that proper solutions exist only for certain values of the energy of the system, and these so-called eigen values (or proper values) agree closely with the energy values expected from spectroscopic data, even in those cases where the older Bohr picture does not give satisfactory agreement with experimental results. Furthermore, the quantum restrictions on the allowed energy states, which were somewhat artificially introduced into the Bohr theory, appear naturally in the solution of the wave equation.

A physical interpretation has been given by M. Born to the wave function ψ which is the solution of Schroedinger's equation for a given system such as an electron in an atom. He assumes that ψ is the amplitude of the wave associated with the electron and that the square of this amplitude at any point in space represents the fraction of the total time that the electron spends there. Thus according to wave mechanics the notion of well-defined orbits for the electrons has to be abandoned, because only the time average of the positions of the electrons can be derived from the equations. This is also in accord with the Heisenberg uncertainty principle which states that the inherent uncertainty Δx in the position of a particle whose momentum is known within an

accuracy Δp is such that the product $\Delta p \cdot \Delta x$ is of the order of magnitude of h.

However, it should be noted that it is still entirely legitimate to speak of the energy states or quantum states of bound electrons, and that everything said in the preceding section about the relation of atomic structure to the building up of the periodic system is still essentially valid provided the well-defined electron orbits are replaced by smeared-out probability distributions.

B. NUCLEAR STRUCTURE

Proton–Neutron Model. Having discussed, at least qualitatively, the arrangement of the external electrons in atoms we shall now return to a consideration of the atomic nuclei whose existence was revealed by the α-particle-scattering experiments. We have seen that a nucleus is a small positively charged particle whose mass accounts for almost the entire atomic mass and whose charge is equal in magnitude but opposite in sign to the sum of the charges of all the electrons in the neutral atom. The dimensions of nuclei are of the order of 10^{-12} to 10^{-13} cm while atomic dimensions, as determined, for example, from gas kinetics, are about 10^{-8} cm. Nuclei, therefore, are very much more dense than ordinary matter; the density of nuclear matter is in the neighborhood of 10^{14} g per cm^3 or 10^8 tons per cm^3.

According to present ideas nuclei consist of protons and neutrons. The simplest nucleus is that of the common hydrogen atom; a single proton constitutes this nucleus. A proton, therefore, carries a positive charge equal in magnitude to the charge on an electron, 4.8025×10^{-10} electrostatic unit (esu). The mass of a proton is approximately equal to that of a hydrogen atom and, therefore, nearly equal to one on the atomic weight scale. The neutron is an uncharged particle whose mass is very nearly equal to but slightly greater than the proton mass.

The nature of the forces which hold neutrons and protons together in nuclei is still not well understood. It is clear that the force cannot be simple electric (Coulombic) attraction, because the neutron carries no charge. Gravitational forces are too weak by very many orders of magnitude to account for nuclear binding. Many experimental facts indicate that nuclear forces have a very

short range, in fact, a range somewhat smaller than nuclear dimensions. The type of force which is now rather generally believed to act between nucleons (a collective term for protons and neutrons) is a so-called exchange force, that is, a force which holds the nucleons together through a continuous exchange of some constituent particles between them. These particles are referred to as mesons; this name was chosen because they are believed to be intermediate in mass between electrons and protons.[1] The exact properties ascribed to them vary in the different meson theories. According to the theory of exchange forces nuclear binding is to be pictured as a resonance phenomenon somewhat analogous to the binding of the two hydrogen atoms in a hydrogen molecule through the sharing or exchange of the pair of electrons between the two atoms.

Atomic Number and Mass Number. The number of protons in a nucleus is called the atomic number Z and ordinarily determines the chemical properties of the element. The atomic numbers of the known elements range from 1 for hydrogen to 96 for curium. The number of neutrons in the nucleus is sometimes called the neutron number N. Nuclei are known with values of N from 0 to 147.

The total number of nucleons (neutrons plus protons) in the nucleus of a given atomic species is called its mass number A; this is the whole number nearest the atomic weight of that particular atom. Mass numbers are known in the range 1 to 242. The difference $N - Z$ (or $A - 2Z$) between the number of neutrons and protons in a nucleus is referred to as the neutron excess or isotopic number.

The symbol used to denote a nuclear species is the chemical symbol of the element with the atomic number as a left subscript and the mass number as a superscript, usually to the right, for example, ${}_2\mathrm{He}^4$, ${}_{27}\mathrm{Co}^{59}$, ${}_{92}\mathrm{U}^{235}$. The atomic number is often omitted because it is uniquely determined by the chemical symbol.

Isotopes, Isobars, Isotones, and Isomers. Atomic species of the same atomic number, that is, belonging to the same element,

[1] In some laboratories the name mesotron is preferred. Intermediate mass particles have been found in cosmic-ray studies, and there is evidence for the existence of more than one type. See also chapter VI, page 134.

but having different mass numbers are called isotopes.[2] In the nuclei of the different isotopes of a given element the same number of protons is combined with different numbers of neutrons. For example, a $_{17}Cl^{35}$ nucleus contains 17 protons and 18 neutrons, whereas a $_{17}Cl^{37}$ nucleus contains 17 protons and 20 neutrons. Deuterium, a rare isotope of hydrogen, has a nucleus containing one proton and one neutron.

Atomic species having the same mass number but different atomic numbers are called isobars. A few examples of isobars are $_{32}Ge^{76}$ and $_{34}Se^{76}$; $_{52}Te^{130}$, $_{54}Xe^{130}$ and $_{56}Ba^{130}$; $_{80}Hg^{204}$ and $_{82}Pb^{204}$.

Atomic species having the same number of neutrons but different mass numbers are sometimes referred to as isotones. For example, $_{14}Si^{30}$, $_{15}P^{31}$, and $_{16}S^{32}$ are isotones because they all contain 16 neutrons per nucleus.

Among the natural radioactive bodies discussed in chapter I there are two, UX_2 and UZ, which have the same mass number as well as the same atomic number, but differ in their radioactive properties. This is an example of nuclear isomerism. Although UX_2 and UZ had been known for several years the phenomenon of nuclear isomerism did not receive much attention until another pair of isomers, Br^{80}, was discovered among artificially produced radioactive species in 1937. Some 70 pairs of nuclear isomers are now known. Nuclear isomers are regarded as different energy states of the same nucleus, each having different radioactive properties. The upper state is always radioactive; the lower state may be stable or radioactive. Isomerism is discussed more fully in chapter VI.

Comparison of the Proton–Neutron with the Older Proton–Electron Hypothesis. Before the discovery of the neutron there existed the idea that nuclei were composed of protons and electrons. In this model the nucleus contained enough protons to account for its approximate mass (one proton for each unit of atomic weight) and enough electrons to reduce the net positive charge to the

[2] For some years the word isotope has been used also in a broader sense to signify any particular nuclear species characterized by its A and Z values. The word nuclide has recently been suggested [T. P. Kohman, *Am. J. Phys* **15**, 356 (1947)], with the following definition: "a species of atom characterized by the constitution of its nucleus, in particular by the number of protons and neutrons in its nucleus."

proper value; thus the neutral atom was to contain as many total electrons as protons, with some in the known atomic orbits or energy levels and the remainder within the nucleus. This model now not only is unfashionable but also presents difficulties which are not easy to resolve, when compared with the proton–neutron nuclear model. One difficulty is that the electron is "too large to be in the small space of the nucleus"; we will consider two aspects of the question of the size of the electron.

With the assumption of unlimited validity for Coulomb's law (probably an improper assumption) it is clear that the electron cannot have zero radius since its potential energy would then be infinite. We may use the known mass of the electron to set an upper limit to this potential energy through the mass-energy relation $E = mc^2$, with the rest mass of the electron $m = 9.11 \times 10^{-28}$ g and the velocity of light $c = 3.00 \times 10^{10}$ cm sec^{-1}. Imagine the assembly of the electronic charge e ($= 4.80 \times 10^{-10}$ esu) from infinitesimal units dq of charge; take q for the charge already collected within the radius R. The (repulsive) force on dq at a distance r from the center of the electron under construction is given by Coulomb's law $f = -q\,dq/r^2$. The energy required for assembly of the electron of radius R is then

$$E = -\int_{q=0}^{q=e}\int_{r=\infty}^{r=R} \frac{q\,dq}{r^2}\,dr = \frac{e^2}{2R}.$$

From this,

$$R = \frac{e^2}{2E} = \frac{(4.80 \times 10^{-10})^2}{2 \times 9.11 \times 10^{-28} \times (3.00 \times 10^{10})^2} = 1.4 \times 10^{-13}\,\text{cm}.$$

This figure is not to be believed as a literal radius of the electron; often the quantity e^2/E (that is, twice the preceding value) is considered merely as a convenient unit of length and termed the "classical radius of the electron."

The length just obtained is of the order of nuclear dimensions. However, if the electron is to be thought of as being within the nucleus its de Broglie wave length $\lambda = h/mv$ must be of the order of nuclear dimensions; this requires a very high momentum and consequently high kinetic energy. For example, to make $\lambda = 10^{-12}$ cm the required total energy from the necessary relativistic formula is

$$E = mc^2 \approx mvc = \frac{hc}{\lambda} = \frac{6.62 \times 10^{-27} \times 3.00 \times 10^{10}}{10^{-12}}$$
$$= 2.0 \times 10^{-4} \text{ erg.}$$

This energy is 125 million electron volts (125 Mev) [3] and is far in excess of the energies known to be associated with nuclear changes involving one nucleon. On the other hand, a similar calculation for a neutron or proton (with mass $M = \sim 1.66 \times 10^{-24}$ g) from a satisfactory nonrelativistic formula gives the kinetic energy,

$$E = \frac{1}{2} Mv^2 = \frac{h^2}{2M\lambda^2} = 0.13 \times 10^{-4} \text{ erg} = 8.3 \text{ Mev.}$$

This value is just in the range of experimental energy changes for the addition or removal of one nucleon. On the basis of this argument the proton–neutron model of nuclear structure is more acceptable than the proton–electron model.

As may be seen in table II–3, the particles, proton, neutron, and electron, all have spin values of ½; that is, in each case the quantized angular momentum is just ½ of the unit $h/2\pi$. For the deuterium nucleus (called the deuteron) the spin is 1 (in the same unit). Now it is easily imagined that the neutron and proton are disposed in the deuteron with spins parallel giving the observed one unit of spin; but no such result is possible with the proton–electron model, where the combined particles (two protons and one electron) would have resultant spin ½ or ³⁄₂. The same argument makes very questionable an earlier view that the neutron itself was composed of one proton and one electron. This argument may be extended to any nucleus whose spin has been determined, with the same result; all nuclei with even mass number show integral spins, and all with odd mass number show half-integral spins. For many nuclei the spin value is not known, but often the system of statistics obeyed is known. Particles with Fermi statistics are considered to have half-integral spin, and those with Bose statistics, integral spin; all this evidence is compatible with the proton–neutron rather than the proton–electron model.

Some additional qualitative remarks might be made about the structure of the deuteron, since especially in this simple case

[3] This unit is defined on page 36.

progress has recently been made in achieving a working quantum-mechanical description. The evidence is that the ground state consists predominantly of overlying proton and neutron waves, with a region of very strong mutual attraction smaller in extent than the dimensions (wave lengths) of the particles, and with no orbital angular momentum. Thus, in spectroscopic notation this is a $^3S_1$ (triplet S_1) state; triplet because of the three permitted orientations of the nuclear spin of 1. The singlet S state $^1S_0$ is known to be virtual (energetically unstable). The actual ground state must contain with $^3S_1$ an admixture of about 4 per cent $^3D_1$, as evidenced by the electric quadrupole moment and the failure of a simple additivity relation between the proton and neutron magnetic moments and the deuteron moment.

C. NUCLEAR PROPERTIES

Mass and Energy. Masses of atomic nuclei are so small when expressed on ordinary mass scales (less than 10^{-21} g) that they are generally expressed on a different scale. The scale used is the so-called *physical atomic-weight scale* in which the mass of an atom of O^{16} is taken as the standard and assigned a mass of exactly 16.00000 units. It should be noted that this scale is not identical with the *chemical atomic-weight scale* (which is used for expressing atomic weights in chemical calculations); in the chemical scale the atomic weight of the natural isotopic mixture of oxygen (containing small amounts of O^{17} and O^{18}) is assigned the value of exactly 16.00000. The unit used is, therefore, larger in the chemical than in the physical scale, and the numerical value of any atomic weight is smaller when expressed on the chemical scale. The conversion factor between the two scales is 1.000272 ± 0.000005, the uncertainty being due to the uncertainty in the isotopic composition of normal oxygen.

The values of isotopic masses given in this book and in most of the literature on nuclear physics and chemistry are on the physical scale and are not nuclear but atomic masses; that is, they include the masses of the extranuclear electrons in the neutral atoms. This convention turns out to have some advantages in the treatment of nuclear reactions and energy relations.

The experimental determination of exact atomic masses involves the use of a mass spectrograph. A number of different types of

mass spectrographs have been devised. In all of these the charge-to-mass ratio of positive ions is determined from the amount of deflection in a combination of magnetic and electric fields; but they use different arrangements for bringing about either velocity focusing or directional focusing or both, for ions of a given e/M. Instruments which use photographic plates for recording the mass spectra are called mass spectrographs; those which make use of collection and measurement of ion currents are referred to as mass spectrometers.

With modern techniques mass determinations can be made with a precision of 1 part in 10^5 for light atoms (up to about $A = 40$) and with somewhat lower precision (sometimes as low as 1 part in 10^4) for heavier atoms. For precision mass determinations the method generally used is the so-called doublet method. This substitutes the measurement of the difference between two almost identical masses for the direct measurement of absolute masses. All measurements must, of course, eventually be related to the standard O^{16}. But for convenience the masses of H^1, H^2, and C^{12} have been adopted as substandards and for this purpose have been carefully measured by determinations of the fundamental doublets:

$(C^{12}H_4^1)^+$	and $(O^{16})^+$	at mass-to-charge ratio 16,
$(H_3^2)^+$	and $(C^{12})^{++}$	at mass-to-charge ratio 6,
$(H^2)^+$	and $(H_2^1)^+$	at mass-to-charge ratio 2.

On the physical scale the mass of a hydrogen atom (sometimes loosely called the proton mass) is 1.00812, the mass of the neutron 1.00893, and that of an electron 0.0005486 mass units. One mass unit equals 1.661×10^{-24} g.

One of the important consequences of Einstein's special theory of relativity is the equivalence of mass and energy. The total energy content E of a system of mass M is given by the relation

$$E = Mc^2,$$

where c is the velocity of light (2.99776×10^{10} cm per sec). Therefore, the mass of a nucleus is a direct measure of its energy content. The measured mass of a nucleus is always smaller than the combined masses of its constituent nucleons, and the difference between the two is called the binding energy of the nucleus.

To find the energy equivalent to 1 mass unit we merely have to put $M = 1.661 \times 10^{-24}$ g and $c = 2.998 \times 10^{10}$ cm sec^{-1}, and we find $E = Mc^2 = 1.493 \times 10^{-3}$ erg. However, energy units much more useful in nuclear work than the erg are the electron volt (ev), the kiloelectron volt (kev; 1 kev = 1000 ev) and the million electron volt (Mev; 1 Mev = 10^6 ev). The electron volt is defined as the energy necessary to raise one electron through a potential difference of 1 volt.

$$1 \text{ ev} = 1.602 \times 10^{-12} \text{ erg}; \qquad 1 \text{ Mev} = 1.602 \times 10^{-6} \text{ erg}.$$

Using these new units, we find

$$1 \text{ mass unit} = 931 \text{ Mev},$$

and

$$1 \text{ electron mass} = 0.5107 \text{ Mev}.$$

As an example we shall calculate the binding energy of He^4. The mass of He^4 is 4.00390; the combined mass of two hydrogen atoms [4] and two neutrons is $2 \times 1.00812 + 2 \times 1.00893 = 4.03410$. Thus the binding energy of He^4 is $4.03410 - 4.00390 = 0.03020$ mass unit or $0.03020 \times 931 = 28.12$ Mev. The binding energy per nucleon in He^4 is, therefore, approximately 7.0 Mev.

The binding energy of the deuteron calculated by the same method is found to be 2.18 Mev. Actually this value was determined experimentally from the threshold for the photodisintegration of the deuteron and combined with the mass-spectrographically-measured masses of proton and deuteron to calculate the neutron mass. No accurate method for a direct measurement of the neutron mass is known.

The average binding energy per nucleon is about 6 to 8 Mev throughout the table of elements except in a few of the lightest nuclei. The binding energy per nucleon is found to have a maximum (near 8.7 Mev) for nuclei in the neighborhood of iron (mass ~ 55). Toward the heavy elements the dropping off is very gradual, and the average binding energy per nucleon reaches values near 7.5 Mev for the heaviest nuclei. On the light side of iron the values drop much faster, and among the lightest nuclei a number of irregularities occur; in particular the binding energies

[4] Since the mass of He^4 includes the mass of two electrons it is clear that it is the *atomic* mass of H^1 which must be used.

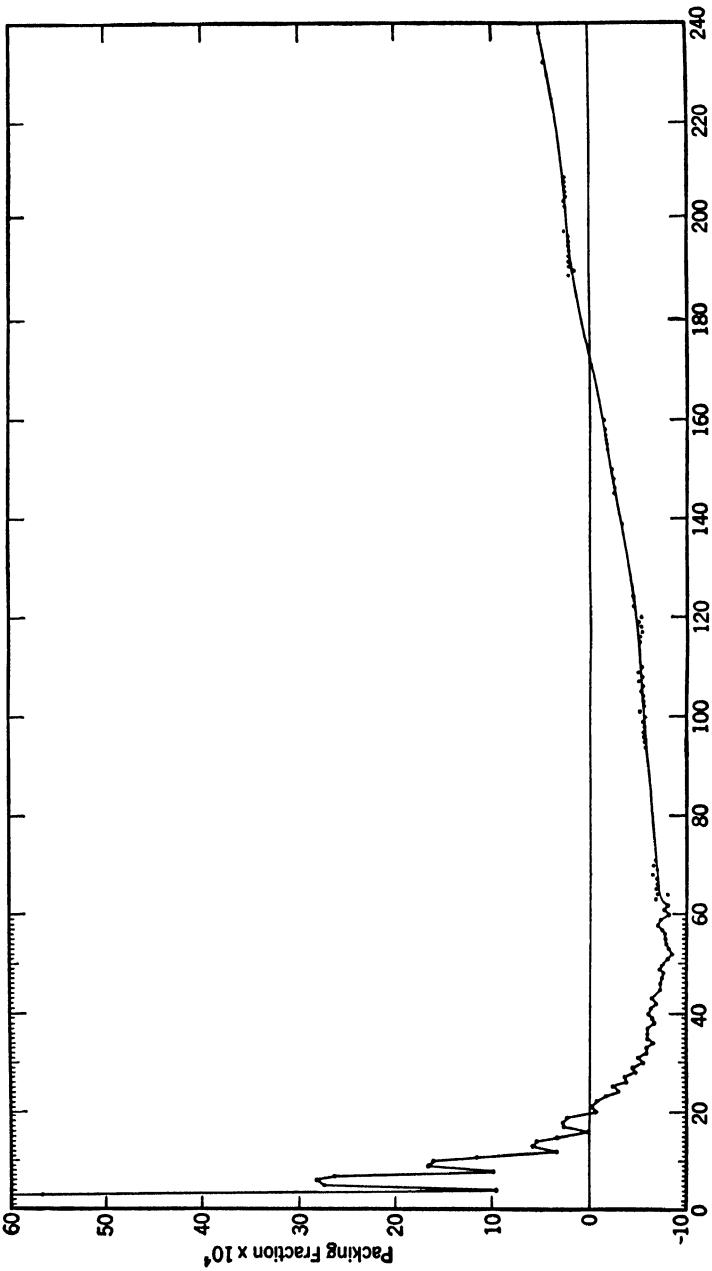


FIGURE II–1. Packing-fraction curve.

of $_2He^4$, $_6C^{12}$, and $_8O^{16}$ are abnormally high. These trends have some important consequences. The sun's radiant energy is believed to result from a series of nuclear transformations whose net effect is the building up of helium atoms from hydrogen atoms which is a very exoergic process. Fission of the heaviest nuclei is energetically possible because nuclei near the middle of the periodic table have higher binding energies per nucleon. There is some evidence that the earth's core consists largely of iron and nickel, and this may well be connected with the maximum in the nuclear stability curve in the region of these elements.

Quantities related to the binding energy are the mass defect and the packing fraction. These are, in fact, more frequently tabulated than the binding energies. The mass defect Δ is the difference between the atomic mass M and the mass number A: $\Delta = M - A$. (Some authors call this the mass excess.) The packing fraction f is the mass defect divided by the mass number: $f = \Delta/A$. (Sometimes f is defined as Δ/M; the difference is negligible.) The packing fraction goes through a minimum in the region of iron. Since the atomic masses fall below the corresponding mass numbers between $A \approx 20$ and $A \approx 180$ the packing fractions are negative in that region. For convenience in tabulation packing fractions are often multiplied by 10^4. A packing fraction curve is reproduced in figure II–1.

Before leaving the subject of binding energies we should mention the binding energy of a nucleus for an additional nucleon, in particular an additional neutron; this is the energy that would be liberated if another neutron were added to the nucleus. This quantity is known for quite a number of nuclei, and values are shown in table II–2.

The masses of some radioactive nuclei can be determined from an accurate knowledge of the energy balance in nuclear reactions involving these nuclei and from their disintegration energies. This subject is discussed in chapter III.

Charge and Radius. Nuclear charges were first determined in the α-particle-scattering experiments mentioned in section A. The most reliable way of determining nuclear charge is by Moseley's relation between Z and the energy characteristic of the K X rays emitted when the element is used as a target in an X-ray tube:

$$\text{Energy} = 10.25(Z - 1)^2 \text{ ev.}$$

TABLE II-2

BINDING ENERGY FOR AN ADDITIONAL NEUTRON

Nucleus	Binding Energy for Added Neutron (in Mev)	Nucleus	Binding Energy for Added Neutron (in Mev)	Nucleus	Binding Energy for Added Neutron (in Mev)
H^1	2.19	Ne^{21}	9.42	K^{39}	7.1
H^2	6.16	Ne^{22}	4.88	Ca^{42}	7.2
He^3	20.51	Na^{23}	7.0	Ti^{46}	9.6
He^4	−0.8	Mg^{24}	7.1	Ti^{47}	9.8
Li^6	7.15	Mg^{25}	12.0	Ti^{48}	6.9
Li^7	1.98	Mg^{26}	5.8	Ti^{49}	10.6
Be^9	6.69	Al^{27}	7.5	Ti^{50}	11.5
B^{10}	11.42	Si^{28}	8.3	Cr^{52}	8.9
B^{11}	3.2	Si^{29}	11.5	Cr^{53}	9.2
C^{12}	4.81	Si^{30}	5.5	Fe^{56}	6.9
C^{13}	8.23	P^{31}	9.9	Fe^{57}	12.0
N^{14}	10.72	S^{32}	9.1	Ni^{60}	4.4
O^{16}	4.12	S^{33}	11.0	Ni^{61}	12.4
O^{17}	7.9	S^{34}	6.7	Zn^{66}	7.0
O^{18}	4.8	Cl^{35}	8.2	Zn^{67}	7.7
Ne^{20}	7.51	A^{40}	7.0		

The data in this table are calculated from the isotopic masses given in table A in the appendix.

A number of different methods have been employed to measure nuclear radii. The results are not all in agreement, and to understand the discrepancies we must consider the shape of the field of force around a nucleus. At distances outside the range of nuclear forces only Coulomb forces act, but closer in towards the center of the nucleus the nuclear attractive forces play the dominant role. This gives rise to a potential-energy curve for a nucleus and a positive particle separated by a distance r (measured between centers) somewhat as sketched in figure II-2.

The first experiments that gave an indication of nuclear dimensions were those on α-particle scattering. In scattering experiments with elements heavier than copper no deviations from the Rutherford scattering formula were observed, because the available α-particle energies did not permit approach to the surface of these nuclei. The minimum separation distances achieved, 1.2×10^{-12} cm for copper and 3.2×10^{-12} cm for gold, merely set upper limits to these nuclear radii. In lighter elements defi-

nite deviations from the Coulomb law were observed. Scattering experiments in aluminum, for example, show marked deviations from inverse-square forces at a distance of about 8×10^{-13} cm. This value presumably corresponds roughly to R in figure II-2.

The radius R of the potential well inside which only the short-range nuclear forces are of importance can be determined in several other ways. The cross-sectional "area" which a nucleus presents to a beam of fast neutrons can be determined from experiments on fast neutron absorption and scattering. This method yields values of R from about 6×10^{-13} for $A \approx 50$ to about 1×10^{-12} for $A \approx 200$. A quantum-mechanical treatment of α-particle decay yields a formula connecting half-life and nuclear radius (see chapter VI); from the known half-lives of the naturally occurring α emitters nuclear radii between 8.4 and 9.8×10^{-13} cm are obtained for these heavy elements. This method involves a calculation of the probability of penetration of the potential barrier around the nucleus by the α particle. By computation of the barrier-penetration probabilities for charged particles from the yields of nuclear reactions between charged particles and nuclei, radii of lighter nonradioactive nuclei can be determined.

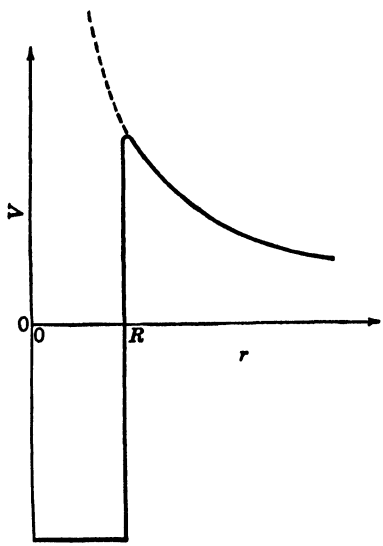


FIGURE II-2. Potential energy in the neighborhood of a nucleus. R is the radius of the potential well.

The nuclear radii determined by the methods described in the last paragraph can be represented approximately by the empirical formula $R = 1.5 \times 10^{-13} A^{1/3}$ cm. We see from this that the nuclear volume is roughly proportional to the nuclear mass and that the volume per nucleon is about constant.

Spin and Magnetic Moment. The intrinsic angular momentum of a nucleus is an integral or half-integral multiple I of $h/2\pi$ (often written as $\hbar$). This spin I is zero or integral for nuclei of

even A and half-integral for nuclei of odd A. Nuclei of even A and even Z seem to have zero spin.

Since the rotation of a charged particle produces a magnetic moment, nuclei with spin also have magnetic moments associated with them. For a particle of mass M, charge Ze, and angular momentum P the magnetic moment is, according to classical theory, $ZeP/2Mc$. For the electron $\left(\text{with } P = \frac{1}{2}\frac{h}{2\pi}\right)$ this formula gives one half of what is called the Bohr magneton (1 Bohr magneton = 0.927×10^{-20} erg per gauss). Since proton and electron have the same magnitude of charge and the same spin the proton magnetic moment calculated from the classical formula is 1/1835 of the electron moment; 1/1835 Bohr magneton is called a nuclear magneton and is used as the unit of nuclear magnetic moments. Actually neither the proton's nor the electron's magnetic moment agrees with this too simple theory. The electron has a magnetic moment of one (instead of one-half) Bohr magneton, and the proton has 2.79 (instead of one-half) nuclear magnetons. The magnetic moments of other nuclei also differ from the classical value (I nuclear magnetons). These moments are sometimes expressed in terms of nuclear g factors; the magnetic moment is then $g \cdot I$ nuclear magnetons. If the magnetic moment is in the direction of the spin, g is taken as positive; if magnetic moment and spin are opposed, g is negative. A negative g factor results from the presence of some neutrons arranged with unpaired spins. The negative magnetic moment of the neutron presumably results from a charge distribution with some negative charge (perhaps due to negative mesons) concentrated near the periphery and overbalancing the effect of an equal positive charge nearer the center.

Nuclear spins and magnetic moments can sometimes be determined from hyperfine structure in atomic spectra. Hyperfine structure arises from the fact that the energy of an atom is slightly different for different (quantized) orientations between nuclear spin and angular momentum of the electrons because of the interaction between the nuclear magnetic moment and the magnetic field of the electrons. From the number of lines in a spectroscopic "hypermultiplet" the nuclear spin I can be determined under suitable conditions. By this method many nuclear spins such as those of Bi^{209} ($I = 9/2$) and Pr^{141} ($I = 5/2$) have been measured.

Although the number of components in a hypermultiplet is determined by the nuclear spin, the amount of the splitting (which is usually of the order of 1 angstrom or less) depends on the value of the nuclear magnetic moment. This dependence is well enough understood to allow a calculation of the magnetic moment from the magnitude of the splitting provided the nuclear spin is known. Thus the nuclear magnetic moments of Bi^{209} and Na^{23} have been determined by this method to be $+3.8$ and $+2.215$ nuclear magnetons, respectively.

Another method of determining nuclear spins is based on the alternating intensities found in rotational spectra of homonuclear diatomic molecules. Molecules in which the two nuclear spins are parallel and antiparallel, respectively, give rise to two sets of alternate lines in the rotational spectrum. The intensity ratio between successive lines is a measure of the abundance ratio for the two types of molecules, which can be shown to be $\frac{I+1}{I}$ at equilibrium except at very low temperatures. For the case of hydrogen the intensity ratio is 3:1, thus confirming the assignment of spin $I = \frac{1}{2}$ to the proton. The two kinds of hydrogen, orthohydrogen (with spins parallel) and parahydrogen (with spins antiparallel) can actually be isolated. In normal hydrogen at moderate and high temperatures they are present in the ratio 3:1.

A second method for the determination of nuclear magnetic moments (and for the measurement of nuclear spin) is the atomic beam method of I. I. Rabi and coworkers. In this method (which is an extension of the Stern–Gerlach experiment for the determination of magnetic moments of atoms) a beam of atoms is sent through an inhomogeneous magnetic field. The nuclear spin I, uncoupled from the electron angular momentum J by the external field, orients itself with respect to the field. This orientation is governed by the usual quantum conditions and the beam is, therefore, split into $2I + 1$ components whose separations are dependent on the nuclear magnetic moment. The energies of these splittings may be found in terms of characteristic alternating magnetic-field frequencies which induce transitions between components. Various modifications of this method, notably the addition of focusing devices and the adaptation to molecular rather than atomic beams, have greatly improved the accuracy of the

results obtainable. The magnetic moment of the neutron has been directly determined by a suitable (and rather drastic) modification of this principle; it was found to be -1.91 nuclear magnetons.

A recent technique for the study of nuclear spins, and especially of magnetic moments, uses the so-called nuclear-induction method. The magnetic dipoles of nuclei of spin I can align themselves with a strong external magnetic field in $2I + 1$ different orientations. The energy differences between the resulting $2I + 1$ energy states correspond to the radio-frequency region, and their magnitude depends on the so-called gyromagnetic ratio, that is, the ratio of magnetic moment to spin. Resonance absorption of radio-frequency radiation will, therefore, take place at a frequency corresponding to these transitions, and the resonance frequency is a measure of the gyromagnetic ratio and, if I is known, of the magnetic moment. In some cases I can be determined separately because the intensity of absorption is a function of I (and of other factors).

Statistics and Other Properties. All nuclei and elementary particles are known to obey one of two kinds of statistics: Bose statistics or Fermi statistics. If the coordinates of two identical particles in a system can be interchanged without change in the sign of the wave function representing the system, Bose statistics applies. If the sign of the wave function does change with the interchange of the coordinates, the particles obey Fermi statistics. In Fermi statistics each completely specified quantum state can be occupied by only one particle; that is, the Pauli exclusion principle applies to all particles obeying Fermi statistics. For particles obeying Bose statistics no such restriction exists. Protons, neutrons, electrons (and some other "elementary" particles such as positrons, neutrinos, and perhaps some types of mesons) all obey Fermi statistics. A nucleus will obey Bose or Fermi statistics, depending on whether it contains an even or odd number of nucleons.

The statistics of nuclei can be deduced from the alternating intensities in rotational bands of the spectra of diatomic homonuclear molecules. With Bose statistics the even-rotational states and with Fermi statistics the odd-rotational states are more populated. This can be illustrated by the cases of hydrogen and deuterium molecules. In normal hydrogen H_2, the ratio of the

populations in the states of odd- and of even-rotational quantum numbers is 3:1 corresponding to spin ½ and Fermi statistics; in deuterium D_2, the ratio is 1:2 corresponding to spin 1 and Bose statistics.

Another nuclear property connected with symmetry properties of wave functions is parity. According to whether or not the wave function of a nucleus changes sign with an inversion of signs of all space coordinates, the nucleus is said to have odd or even parity. States of even and odd parity do not resonate, and it is for this reason that the deuteron ground state, a mixture of $^3S_1$ and $^3D_1$, cannot contain a P-state component.

Finally mention should be made of the electric quadrupole moment of nuclei. This property may be thought of as arising from an elliptic charge distribution in the nucleus. The quadrupole moment q is given by the equation $q = \frac{2}{5}Z(a^2 - b^2)$, where a is the semiaxis of rotation of the ellipsoid, and b is the semiaxis perpendicular to a; q has the dimensions of an area. For the deuteron $q = +2.73 \times 10^{-27}$ cm^2, and the charge distribu-

TABLE II-3

PROPERTIES OF SOME ELEMENTARY AND OTHER SIMPLE PARTICLES

Particle or Nucleus			Rest		Magnetic
Symbol	Name	Charge *	Mass †	Spin ‡	Moment §
e^- or β^-	electron	−1	0.0005486	½	−1835
e^+ or β^+	positron	+1	0.0005486	½	+1835
γ	photon	0	0	1	0
υ	neutrino	0	<0.00002	½	<0.3
n	neutron	0	1.00893	½	−1.913
μ	μ meson	+1, −1	0.118	0 or ½?	
π	π meson	+1, −1, (0)	0.156	0 or 1	
H^1 or p	proton	+1	1.008123	½	+2.793
H^2, d, or D	deuteron	+1	2.01471	1	+0.857
H^3, t or T	triton	+1	3.01702	½	+2.976
He^3		+2	3.01700	(½)	(−)2.13
He^4 or α	α particle	+2	4.00390	0	(0)

* In units of $e = 4.8025 \times 10^{-10}$ esu.

† For the proton and other nuclei the mass of the neutral atom is listed. The unit is the physical atomic weight unit.

‡ In units of $h/2\pi$.

§ In units of the nuclear magneton ($eh/4\pi Mc$), where M is the proton mass. Positive values indicate moment orientations with respect to spin orientations that would result from spinning positive charges.

tion is cigar-shaped. Quadrupole moments, including both positive and negative values, are known for a number of other nuclei with $I > \frac{1}{2}$. They may be determined from abnormal hyperfine splittings in atomic spectra. Some attempts have been made to obtain information on quadrupole moments from a study of transitions between hyperfine components by means of microwave spectroscopy.

D. ISOTOPY AND ISOTOPE SEPARATIONS

Occurrence of Isotopes in Nature. The phenomenon of isotopy was discovered when different radioactive bodies in the natural decay series, for example, RaB, AcB, and ThB, were found to exhibit identical chemical properties (in the case mentioned the properties are those of lead). This led to a search for the existence of isotopes in nonradioactive elements. In early experiments with ion deflections in magnetic and electric fields J. J. Thomson showed in 1913 that neon consisted of two isotopes of mass numbers 20 and 22 (now a third isotope Ne^{21} is known). Since that time the development of improved instruments, notably F. W. Aston's mass spectrograph and its various modifications, has made very careful searches for isotopes possible in all the elements existing in stable form. As a result of these mass-spectrographic investigations we now know that the elements with atomic numbers between 1 and 83 have on the average more than three stable isotopes each. Some elements such as beryllium, phosphorus, arsenic, and bismuth have a single stable nuclear species each, whereas tin, for example, has as many as 10 stable isotopes. In addition each element from $Z = 1$ to $Z = 96$ has at least one known radioactive isotope, and in some cases there are as many as 12 or 15. The total number of radioactive species now known is about 700.

The stable isotopes of a given element generally occur together in constant proportions. This accounts for the fact that atomic weight determinations on samples of a given element from widely different sources generally agree within experimental errors. However, there are some notable exceptions to this rule of constant isotopic composition. One is the variation in the abundances of lead isotopes, especially in ores containing uranium and thorium, which has already been discussed in chapter I in connection with

the determination of the age of the earth. Similarly the isotope Sr^{87} has been found to have an abnormally high abundance in rocks which contain rubidium; this is explained by the fact that Rb^{87} is a naturally occurring β emitter and decays to Sr^{87}. Helium from gas wells probably has its origin in radioactive processes (α disintegrations) and contains a much smaller proportion of the rare isotope He^3 than does atmospheric helium. Water from various sources shows slight variations in the H^1/H^2 ratio. This is in some cases due to the fact that heavy water has a slightly lower vapor pressure than ordinary water and is, therefore, concentrated by evaporation. The enrichment of H^2 in the water of the Dead Sea and in certain vegetables is ascribed to this cause. The waters which show abnormally high H^2 concentrations usually also have slightly higher O^{18}/O^{16} ratios than normal. Another cause for small variations in isotopic composition is the fact that chemical equilibria are slightly dependent on the molecular weights of the reactants, and this may lead to isotopic enrichments in the course of reactions occurring in nature. For example, the slight enrichment of C^{13} in limestones relative to some other sources of carbon is explained by the fact that the equilibrium in the reaction between CO_2 and water to form carbonic acid lies somewhat further towards the side of carbonic acid for $C^{13}O_2$ than for $C^{12}O_2$.

Isotope Separations. Some of the principles involved in isotope fractionations in nature have been exploited for the artificial concentration and separation of isotopes. We stated earlier that chemical properties are determined by the nuclear charge, and it would follow from this that isotopes of a given element are completely identical in their chemical behavior. However, the isotopic mass does have a very slight effect on chemical equilibria. In fact, for light elements such as carbon and nitrogen multistage exchange reactions have been used to produce separated isotopes on a commercial scale. These chemical effects become vanishingly small for isotopes of heavier elements because they depend essentially on percentage differences in mass.

Other methods for isotope separations should be mentioned briefly. Diffusion of gases or liquids through porous membranes results in separation, the lighter isotopes diffusing more rapidly. This method has been successfully applied to the large-scale separation of the uranium isotopes. The thermal diffusion technique of K. Clusius and G. Dickel makes use of the fact that in a

thermal gradient the heavy isotopic component concentrates at the cold end and, by means of vertical adjacent hot and cold walls, provides a very ingenious arrangement for obtaining multistage separations in simple apparatus through the combined actions of convection and thermal diffusion. Separations have also been effected by use of high-speed centrifugation. Differences in vapor pressure between compounds containing different isotopes lead to concentration of the heavy constituents in the residues from slow evaporations; this method has been used for the concentration of heavy water (H_2^2O or D_2O). Most heavy water is now produced by electrolysis, which also enriches the heavy component in the residues. Finally the electromagnetic methods of separation must be mentioned. Mass spectrographs have long been used to separate small quantities of isotopes, but during World War II the electromagnetic method was developed from a microgram to a kilogram scale for the purpose of separating U^{235} in quantity. The large electromagnetic separators (calutrons) at Oak Ridge, Tenn., are now used also for the separation of isotopes of a large number of elements throughout the periodic table.

E. NUCLEAR SYSTEMATICS

Binding Energies. Numerous attempts have been made to account for the shape of the packing-fraction or binding-energy curve. On a semiempirical basis an expression for the binding energy of a nucleus as a function of its proton and neutron composition, that is as a function of Z and $N = A - Z$, can be obtained. Several different expressions are given in the literature; one that gives rather good agreement with measured binding energies at least for $A > 80$ is

$$E_B = 14.1\,A - 13.1\,A^{\frac{2}{3}} - 0.585\,Z(Z-1)\,A^{-\frac{1}{3}} - 18.1(N-Z)^2\,A^{-1} + \delta\,A^{-1}, \quad \text{(II–4)}$$

where $\delta = 132$ for Z even and N even, $\delta = -132$ for Z odd and N odd, and $\delta = 0$ for A odd. The binding energy E_B is here expressed in million electron volts (Mev).

The first term in equation II–4, and this is the most important term, is proportional to the number of nucleons A. This observation can be interpreted to mean that the nuclear forces have short

ranges and act between a small number of nucleons only. The saturation of these forces is apparently almost (but certainly not entirely) complete when four particles, two protons and two neutrons, interact, as is indicated by the large observed binding energies of He^4, C^{12}, O^{16}.

Those nucleons at the surface of a nucleus can be expected to have unsaturated forces, and, consequently, a reduction in the binding energy proportional to the nuclear surface should be taken into account. This is the second term, containing $A^{2/3}$ which is a measure of the surface since A is proportional to the volume.

The coulombic repulsive force between protons is, of course, not of the saturation type and is of sufficient range to be effective for all the protons in a nucleus. Therefore, each of the Z protons interacts with the other $Z - 1$ protons to reduce the binding energy as shown in the third term. The factor $A^{-1/3}$ enters this term because it measures the average separation distance for protons distributed in a volume proportional to A.

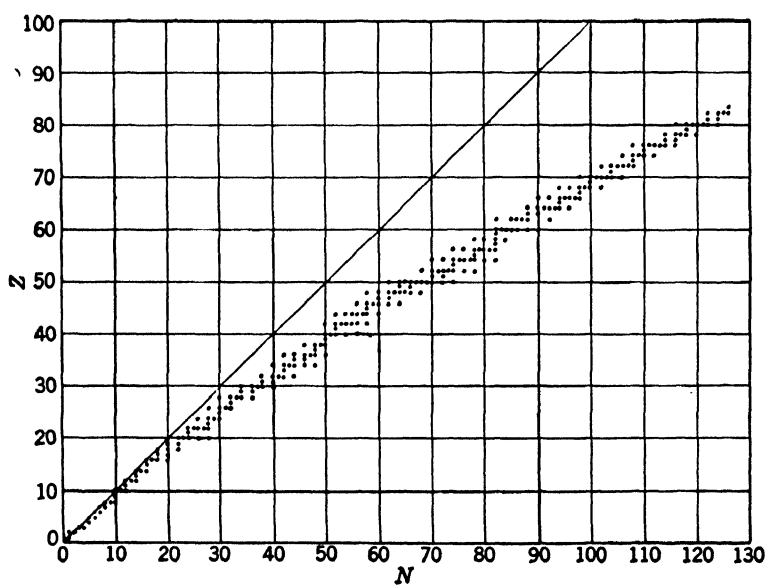


FIGURE II-3. The known stable nuclei on a plot of Z versus N. Note the gradual increase in the neutron-proton ratio; the 45° line indicates a neutron-proton ratio of 1.

Other factors being equal, maximum stability, that is, minimum binding energy per nucleon, is found for nuclei with equal numbers of protons and neutrons. This would indicate that (except for the coulombic energy effect) the protons and neutrons have similar energy-level spacings in nuclei. In this case the levels of lowest energy are occupied when $Z = N$; additional nucleons, most often neutrons, are by the Pauli exclusion principle required to go into levels of greater energy. This effect is expressed in quantitative (empirical) fashion by the fourth term.

Closed Shells in Nuclear Structure. There is some evidence in binding energies and radii of nuclei that there is a shell structure somewhat analogous to the electron-shell structure in atoms, although less pronounced and much less understood. Attempts have been made to derive the quantum numbers for the successive shells from assumptions about the nuclear forces, and the order of the first few levels for protons and neutrons appears likely to be $1s$, $2p$, $2s$, $3d$. Remembering that s, p, d stand for angular-momentum quantum numbers $l = 0$, 1, 2, and that for each l there are $2l + 1$ different states (corresponding to m values from $+l$ to $-l$ including 0) each of which may now be occupied by two protons and two neutrons, we get for the numbers of nucleons in nuclei with these shells successively completed:

$$A = 4; \qquad A = 4 + 12 = 16;$$

$$A = 4 + 12 + 4 = 20; \qquad A = 4 + 12 + 4 + 20 = 40.$$

These completed shell structures correspond to ${}_2\mathrm{He}^4$, ${}_8\mathrm{O}^{16}$, ${}_{10}\mathrm{Ne}^{20}$, and ${}_{20}\mathrm{Ca}^{40}$. Experimental observations on the stabilities (binding energies) of these nuclei are not in disagreement with this conclusion. Above ${}_{20}\mathrm{Ca}^{40}$ there are no stable nuclei with $Z = N$ as a consequence of the coulombic repulsion, but there is evidence that energy shells fill with protons or neutrons to produce particularly stable structures when Z or N equals 20 (as in Ca^{40}), 50, 82, or 126.

Types of Nuclei. Nuclei can be classified according to whether they contain even or odd numbers of protons and neutrons. There are then four types of nuclei as shown in the following tabulation. The distribution of the known stable nuclear species among these four types and the corresponding values of δ in equation II–4 are as follows:

162 even–even [5]	(Z even, N even)	$\delta = +132$ [6]
56 even–odd	(Z even, N odd)	$\delta = 0$
52 odd–even	(Z odd, N even)	$\delta = 0$
4 odd–odd	(Z odd, N odd)	$\delta = -132$ [6]

A relation between the frequency of occurrence and the stability as measured by the binding energy is apparent. The striking preponderance of even–even nuclei and complete absence of odd–odd nuclei outside the region of the lightest elements (the four odd–odd nuclei are ${}_1H^2$, ${}_3Li^6$, ${}_5B^{10}$, and ${}_7N^{14}$) can be explained in terms of a tendency of two like particles to complete an energy level by pairing opposite spins.

The greater stability of nuclei with filled energy states is apparent not only in the larger number of even–even nuclei, but also in their greater abundance relative to the other types of nuclei. On the average, elements of even Z are much more abundant than those of odd Z (by a factor of about 10). For elements of even Z the isotopes of even mass (even N) account in general for about 70 to 100 per cent of the element (beryllium, xenon, and dysprosium being exceptions).

Stability Rules. Related to the foregoing is the observation that for any odd Z there are never more than two stable isotopes, and if there are two their mass numbers differ by two units. Analogously, for any odd N there are no more than two stable isotones, and if there are two their mass numbers differ by two units. The only exceptions to these two rules are the four light odd–odd nuclei. Beyond $Z = 7$, the only odd values of N for which two stable isotones exist are 55, 65, and 85.

Another empirical rule about the stable isotopes of elements of odd Z is that their mass numbers do not have values other than $A - 3$, $A - 1$, or $A + 1$, where A is the mass number of the heaviest stable isotope of the preceding element. This rule applies without exception above oxygen.

For any even Z there exist no more than two stable isotopes of odd mass number. The one exception to this rule is ${}_{50}Sn$ with

[5] Sometimes these are called g–g, g–u, etc., from the German gerade = even and ungerade = odd.

[6] The quantity δ is actually not a constant but varies rather irregularly with A. The value given is an average for $A > 80$. For $A < 60$ a lower value, perhaps $\delta \sim \pm 65$, should be used, but in that region equation II–4 does not give reliable results anyway.

three odd isotopes; but there is reason to suspect that Sn^{115} is actually not stable (see below). The analogous rule can again be stated for isotones with even N; in fact, even N with two stable isotones of odd mass number occurs only at $N = 20$ and $N = 82$.

Between oxygen and bismuth the following two rules also apply: (1) for every stable nuclide with odd Z there are two stable isotones, one with $Z - 1$ and one with $Z + 1$ protons; (2) for every stable nuclide with odd N there are two stable isotopes, one with $N - 1$ and one with $N + 1$ neutrons.

Finally, the following important rule is stated here, although its discussion must be deferred to chapter VI, section B. No pairs of stable isobars exist with Z differing by one unit. Apparent exceptions to this rule are ${}_{48}Cd^{113}$–${}_{49}In^{113}$, ${}_{49}In^{115}$–${}_{50}Sn^{115}$, ${}_{51}Sb^{123}$–${}_{52}Te^{123}$, and ${}_{56}Ba^{138}$–${}_{57}La^{138}$–${}_{58}Ce^{138}$.

EXERCISES

1. Show that h ($= 6.624 \times 10^{-27}$ erg sec) has the dimensions of angular momentum.

2. Derive the expressions for the kinetic and potential energies of an electron in a Bohr orbit.

3. Show that the circumference of a stable Bohr orbit is always an integral multiple of the de Broglie wave length of the electron in the orbit.

4. From the definitions of binding energy and packing fraction of a nucleus, derive a relation between the two quantities.

5. Calculate the binding energy per nucleon in Li^6, P^{31}, Ti^{50}, Ni^{58}, Ag^{107}, La^{139}, Gd^{158}, Au^{197}, U^{238}.

Answers for first three: 5.30, 8.42, 8.69 Mev.

6. Calculate from the masses given in table A in the appendix the binding energy for an additional neutron in O^{16}, P^{31}, Ti^{50}.

7. Using the natural abundances of the oxygen isotopes from table A in the appendix, calculate the atomic weight of ordinary oxygen on the physical scale and the conversion factor between the physical and chemical scales of atomic weights.

8. What is the energy of an electron whose de Broglie wave length is 1.5×10^{-13} cm? *Answer:* 830 Mev.

9. Without reference to tables, calculate the approximate charge and mass of the proton in familiar units (coulombs and grams, respectively).

10. The three fundamental mass doublets have been found to have the following separations:

$$(C^{12}H_4)^+ - (O^{16})^+ = 36.30 \text{ millimass units}$$

$$H_2^+ - D^+ = 1.538 \text{ millimass units}$$

$$D_3^+ - (C^{12})^{++} = 42.22 \text{ millimass units}$$

Calculate the atomic masses of H, D, and C^{12}.

11. The radiation from the sun at normal incidence at the earth (93 million miles away) amounts to 0.135 joule per cm^2 per sec. The source of this energy is believed to be the conversion of hydrogen into helium. At what rate is hydrogen being consumed, in gram atoms per second?

Answer: 5.9×10^{14}.

12. Using what information you have on nuclear radii, estimate the minimum α-particle energy necessary to observe deviations from Rutherford scattering in silver. *Answer:* ~14 Mev.

13. With the aid of equation II–4 estimate: (*a*) the energy liberated when one additional neutron is added to U^{235}, (*b*) the energy liberated when one additional neutron is added to U^{238}, (*c*) the amount of energy by which I^{129} is unstable with respect to β decay to Xe^{129}. *Answer:* (*a*) 7.3 Mev.

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III. NUCLEAR REACTIONS

A. THE NATURE OF NUCLEAR REACTIONS

A nuclear reaction is a process in which a nucleus reacts with another nucleus, an elementary particle, or a photon, to produce in a time of the order of 10^{-12} sec or less one or more other nuclei (and possibly neutrons or photons). Most of the nuclear reactions studied to date are of the type in which a nucleus reacts with a light particle (neutron, proton, deuteron, alpha particle, electron, photon) and the products are a nucleus of a different species and again one or more light particles. The chief exception to this description is the fission reaction.

Notation. As an example of a nuclear reaction we may cite the first such process discovered (in 1919), the disintegration of nitrogen by α particles. When Rutherford bombarded nitrogen with α particles from RaC′ he could observe scintillations on a zinc sulfide screen even when enough material was interposed between the nitrogen and the screen to absorb all the α particles. Further experiments proved the long-range particles causing the scintillations to be protons, and the results were interpreted in terms of a nuclear reaction between nitrogen and α particles to give oxygen and protons, or, in the usual notation:

$$_7N^{14} + {}_2He^4 \rightarrow {}_8O^{17} + {}_1H^1.$$

Since 1919 well over 1000 different nuclear reactions have been studied. The recognition of reaction products is greatly facilitated when these are unstable because characteristic radioactive radiations can then be observed. Artificial radioactivity was discovered in 1933 when I. Curie and F. Joliot were studying the emission of positrons by light elements under α-particle bombardment and found that some of the elements such as B, Mg, and Al continued to emit positrons after the removal of the α-particle source. For example, in the reaction between aluminum and α particles the product is P^{30} which decays by positron emission to Si^{30} with a

2.5-min half-life. The reaction is

$$_{13}Al^{27} + {}_2He^4 \rightarrow {}_{15}P^{30} + {}_0n^1.$$

The notation used for nuclear reactions is analogous to that in chemical reactions, with the reactants on the left- and the reaction products on the right-hand side of the equation. In all reactions so far observed the total number of protons and the total number of neutrons (or total Z and total A) are conserved, just as in chemical reactions the number of atoms of each element is conserved. In addition other properties such as energy, momentum, angular momentum, statistics, and parity are conserved in nuclear reactions.

A short-hand notation is often used for the representation of nuclear reactions. The light bombarding particle and the light fragments (in that order) are written in parentheses between the initial and final nucleus; in this notation the two afore-mentioned reactions would read

$$N^{14}\ (\alpha, p)\ O^{17} \quad \text{and} \quad Al^{27}\ (\alpha, n)\ P^{30}.$$

The symbols n, p, d, α, e, γ, x are used in this notation to represent neutron, proton, deuteron, alpha particle, electron, gamma ray and X ray.

Comparison of Nuclear and Chemical Reactions. Nuclear reactions, like chemical reactions, are always accompanied by a release or absorption of energy, and this is expressed by adding the term Q to the right-hand side of the equation. Thus a more complete statement of Rutherford's first transmutation reaction reads

$$_7N^{14} + {}_2He^4 \rightarrow {}_8O^{17} + {}_1H^1 + Q.$$

The quantity Q is called the energy of the reaction or more frequently just "the Q of the reaction." Positive Q corresponds to energy release (exoergic reaction); negative Q to energy absorption (endoergic reaction).

Here an important difference between chemical and nuclear reactions must be pointed out. In treating chemical reactions we always consider macroscopic amounts of material undergoing reactions, and, consequently, heats of reaction are usually given per mole or occasionally per gram of one of the reactants. In the case of nuclear reactions we usually consider single processes, and the Q values are therefore given per nucleus transformed. If

the two are calculated on the same basis, the energy release in a representative nuclear reaction is found to be many orders of magnitude larger than that in any chemical reaction. For example, the reaction N^{14} (α, p) O^{17} has a Q value of -1.13 Mev or $-1.13 \times 1.602 \times 10^{-6}$ erg or $-1.13 \times 1.602 \times 10^{-6} \times 2.390 \times 10^{-11}$ kg cal $= -4.33 \times 10^{-17}$ kg cal for a single process. Thus to convert 1 g atom of N^{14} to O^{17}, the energy required would be $6.02 \times 10^{23} \times 4.33 \times 10^{-17}$ kg cal $= 2.61 \times 10^{7}$ kg cal. This is about 10^5 times as large as the largest values observed for heats of chemical reactions. On the other hand, one must keep in mind the fact that nuclear reactions are very rare events compared with chemical reactions; one reason is that the small sizes of nuclei make effective nuclear collisions quite improbable.

B. ENERGETICS OF NUCLEAR REACTIONS

The Q of a Reaction. It is clear from the foregoing discussion that the energy changes involved in nuclear reactions are of such magnitude that the corresponding mass changes in nuclear particles must be observable. (The mass changes accompanying chemical reactions are too small to be observable with the most sensitive balances available.) If the masses of all the particles participating in a nuclear reaction are known from mass spectrographic data, as is the case for the N^{14} (α, p) O^{17} reaction, the Q of the reaction can be calculated. The sum of the N^{14} and He^4 masses is 18.01141 mass units, and the sum of the O^{17} and H^1 masses is 18.01262 mass units; thus an amount of energy equivalent to 0.00121 mass unit has to be supplied to make the reaction energetically possible, or $Q = -0.00121 \times 931$ Mev $= -1.13$ Mev. In cases where the Q value is known experimentally (from the kinetic energies of the bombarding particle and the reaction products), it is sometimes possible to compute the unknown mass of one of the participating nuclei. By this method the masses of a number of radioactive nuclei have been determined (see exercise 4).

It is often possible to calculate the Q value of a reaction even if the masses of the nuclei involved are not known if the product nucleus is radioactive and decays back to the initial nucleus with known decay energy. Consider, for example, the reaction Pd^{106} (n, p) Rh^{106}. The product Rh^{106} decays with a 30-sec

half-life and the emission of 3.55-Mev β particles to the ground state of Pd^{106}. We can write this sequence of events as follows:

$$_{46}Pd^{106} + {}_0n^1 \rightarrow {}_{45}Rh^{106} + {}_1H^1 + Q;$$

$$_{45}Rh^{106} \rightarrow {}_{46}Pd^{106} + \beta^- + 3.55 \text{ Mev}.$$

Adding the two equations we see that the net change is just the transformation of a neutron into a proton and an electron with accompanying energy change; or, symbolically,

$${}_0n^1 \rightarrow {}_1H^1 + \beta^- + Q + 3.55 \text{ Mev}.$$

Note that the symbol $_1H^1$ must here stand for a bare proton (evident from the charge conservation) whereas the listed "proton mass" includes the mass of one orbital electron.[1] For energy balance we therefore write

$$M_n = M_{H^1} + Q + 3.55 \text{ Mev}$$

where $M_n = 1.00893$ and $M_{H^1} = 1.00812$ mass units. Then

$$Q = (1.00893 - 1.00812) \times 931 - 3.55$$

$$= 0.75 - 3.55 = -2.80 \text{ Mev}.$$

In the first example calculated we found the Q value of the reaction N^{14} (α, p) O^{17} to be -1.13 Mev. Does that mean that this reaction can actually be produced by α particles whose kinetic energies are just over 1.13 Mev? The answer is no, for two reasons. First, in the collision between the α particle and the N^{14} nucleus conservation of momentum requires that at least 4/18 of the kinetic energy of the α particle must be retained by the products as kinetic energy; thus, only 14/18 of the α particle's kinetic energy is available for the reaction. The threshold energy of α particles for the N^{14} (α, p) O^{17} reaction, that is, the kinetic energy of α particles just capable of making the reaction energetically possible, is $18/14 \times 1.13$ Mev $= 1.45$ Mev. The fraction of the bombarding particle's kinetic energy which is retained

[1] In general, for negative β-particle emission and electron-capture processes, the masses of electrons never have to be included in calculations when atomic masses are used. However, whenever a positron is involved in a reaction, two electron masses have to be taken into account: one for the positron and one for the extra electron that has to leave the electron shells to preserve electrical neutrality.

as kinetic energy of the products becomes smaller with increasing mass of the target nucleus (see exercise 5).

Barriers for Charged Particles. The second reason why the α particles must have higher energies than is evident from the Q value to produce the reaction N^{14} (α, p) O^{17} in good yield is the Coulomb repulsion between the α particle and the N^{14} nucleus. The repulsion increases with decreasing distance of separation until the α particle comes within the range of the nuclear forces of the N^{14} nucleus. This Coulomb repulsion gives rise to the potential barrier already mentioned in chapter II. The height V of the potential barrier around a nucleus of charge Z_1e and radius R_1 for a particle of positive charge Z_2e and radius R_2 may be estimated as the energy of Coulomb repulsion when the two particles are just in contact: $V = \dfrac{Z_1Z_2e^2}{R_1 + R_2}$. Obtaining the nuclear radii from the formula [2] $R = 1.5 \times 10^{-13} A^{1/3}$, we get for the barrier height between N^{14} and an α particle a value of about 3.4 Mev. Thus, at least according to the classical theory an α particle of energy less than 3.4 Mev cannot enter the N^{14} nucleus. Rutherford actually used α particles of over 7 Mev in his experiments. In the quantum mechanical treatment of the problem there exists a finite probability for "tunnelling through the barrier" by lower-energy particles, but this probability drops rapidly as the energy of the particle decreases. The penetration of potential barriers is discussed in connection with α decay (chapter VI, section A).

The potential barrier around a given nucleus for protons and for deuterons is about half as high as for α particles. The height of the potential barrier increases with increasing Z of the target nucleus; it is roughly proportional to $Z^{2/3}$ (not to Z, because the nuclear radius R increases approximately as $Z^{1/3}$). For the heaviest elements the potential barriers are about 15 Mev for protons and deuterons and about 30 Mev for α particles. In order to study nuclear reactions induced by charged particles, especially reactions involving heavy elements, it was therefore necessary to develop machines capable of accelerating charged particles to energies of many millions of electron volts.

[2] For the lightest nuclei this formula for nuclear radii is actually a poor approximation; but for an estimate of barrier heights it is adequate.

It must be emphasized that potential barriers have an effect not only for particles entering, but also for particles leaving nuclei. For this reason a charged particle has to be excited to a rather high energy inside the nucleus before it can either go over the top of the barrier or, according to the quantum mechanical picture, leak through the barrier with appreciable probability. Therefore, charged particles are usually emitted from nuclei with considerable energies (more than 1 Mev).

Slow Neutrons. It is evident from the foregoing discussion that, in general, it should be much easier for neutrons to enter and leave nuclei than it is for charged particles. This is indeed the case; even neutrons of very low energy can enter most nuclei with comparative ease. In fact, the so-called thermal neutrons, that is, neutrons whose energy distribution is approximately that of gas molecules in thermal agitation at ordinary temperatures, have particularly high probabilities for entering nuclei. The fact that a neutron of thermal energy (about 0.035 ev at 0°C) has a de Broglie wave length of 1.5×10^{-8} cm may be considered as responsible for the unusually high reaction probabilities of slow neutrons with some nuclei. This subject is discussed further in section D.

Our only sources of neutrons are nuclear reactions, in which neutrons are emitted from highly excited nuclei and, therefore, usually have initially rather high kinetic energies. Because of the great importance of slow or thermal neutrons in bringing about nuclear reactions, processes for slowing down fast neutrons to thermal energies have received much attention, both theoretically and experimentally. Although fast neutrons may lose energy in inelastic collisions with nuclei, most slowing down is accomplished through a process of many successive elastic collisions with nuclei. The lighter the nucleus with which a neutron collides, the greater is the fraction of the neutron's kinetic energy that can be transferred in the elastic collision. For this reason hydrogen-containing substances such as paraffin or water are the best slowing-down media for neutrons. Heavy water, helium, and carbon are also used. The mean free path between collisions in water or paraffin is several centimeters for a neutron of a few million electron volts energy, and a few millimeters for a thermal neutron. In each collision between a neutron and a proton the neutron's energy is on the average distributed equally between the two particles.

The average percentage energy loss of the neutrons is constant in each collision, and, as a result of this logarithmic averaging, the average neutron energy after n collisions with protons is e^{-n} times the initial neutron energy. Approximately 20 collisions are therefore necessary to reduce neutrons from a few million electron volts to thermal energies. Eight or ten inches of paraffin surrounding a neutron source are adequate for reducing most neutrons to the thermal energy distribution. The whole slowing-down process requires less than 0.001 sec. The probable eventual fate of a thermal neutron in a hydrogenous medium like water or paraffin is capture by a proton to form a deuteron; but, since the probability of this reaction is quite small compared with the probability for scattering, a neutron after reaching thermal energies makes about 150 further collisions before being captured. Paraffin and water are good substances to use for the slowing down of neutrons because the capture probabilities in oxygen and carbon are even much smaller than in hydrogen. Heavy water is better than ordinary water because of the low probability of neutron capture by deuterium. Carbon (graphite) is also useful as a slowing-down medium because of its extremely low capture probability for neutrons; many more collisions are, of course, necessary to reduce neutrons to thermal energies in carbon than in hydrogen, but after reaching thermal energies the neutrons can exist longer in carbon. In either substance the lifetime of a neutron is only a fraction of a second. Even if neutrons could be kept in a medium where they would not eventually be captured, they might not be able to exist very long; they are believed to be unstable with respect to decay into protons and electrons, and theory predicts a half-life of about 20 min for this process. This decay has not yet been definitely observed.

It should be evident from the foregoing discussion that thermal neutrons do not all have the same energy. After neutrons are slowed to energies comparable to thermal agitation energies (about 0.035 ev) they may either lose or gain energy in collisions, and the result is a Maxwellian distribution of velocities, in which the fraction [3] of the total number of neutrons with velocity

[3] The complete expression for this fraction, denoted by $N(v)dv$, is

$$N(v)dv = 4\pi^{-\frac{1}{2}} \left(\frac{M}{2kT}\right)^{\frac{3}{2}} v^2 e^{-\frac{Mv^2}{2kT}} dv.$$

between v and $v + dv$ at any given temperature is proportional to $v^2 e^{-\frac{Mv^2}{2kT}} dv$, where M is the neutron mass, and k is the Boltzmann constant. The average energy of the neutrons can be varied by varying the temperature of the slowing-down medium. At very low temperatures the Maxwellian distribution function becomes a poor approximation because of the discrete energy levels of the bound atoms of the slowing-down medium.

C. MECHANISMS AND TYPES OF NUCLEAR REACTIONS

The Bohr Picture of Nuclear Reactions. We have talked about nuclear reactions without considering in any detail the mechanisms by which such reactions may take place. In 1936 Bohr developed a theory of nuclear reactions which has been very successful in explaining many features of reactions induced by particles of moderate energies (at least up to 30 or 40 Mev). This theory was based on a concept of the nucleus as a densely packed system with distances between nucleons of the same order of magnitude as the range of the nuclear forces and interaction energies between nucleons of the same order of magnitude as the kinetic energies of the incident particles. Bohr argued that an incident particle hitting such a system would lose much of its kinetic energy in the first few collisions with the nucleons and would then be held by the nuclear forces. Thus he postulated as the first step in any nuclear reaction the amalgamation of target nucleus and incident particle into a compound nucleus. In this compound nucleus the kinetic energy of the incident particle and the additional binding energy contributed by it are rapidly distributed among all the nucleons. The second step of the reaction, the breaking up of the compound nucleus into the reaction products, can take place only after a relatively long time because a large number of collisions is required before enough energy is likely to be "accidentally" concentrated on one nucleon to allow it to escape from the nuclear binding forces. The lifetimes of compound nuclei are of the order of 10^{-12} to 10^{-14} sec [4] which

[4] These times are too short to have been measured directly. By use of the uncertainty relation between time and energy ($\Delta E \cdot \Delta t \approx h/2\pi$) the lifetime of a compound nucleus in a particular energy state can be deduced from the width in energy of this state. This in turn can be determined experimentally from the spread in the energies of the incident particle which can be used to reach the state in question.

is very long compared to the time required for a fast particle to traverse a distance equal to a nuclear diameter. For example, a 0.5-Mev neutron (velocity 10^9 cm per sec) would traverse a medium heavy nucleus (diameter 10^{-12} cm) in $10^{-12}/10^9 = 10^{-21}$ sec.

An essential feature of the Bohr picture is that the two steps of a nuclear reaction, the formation and the breaking up of a compound nucleus, are independent of each other. A given compound nucleus may be formed in different ways and may also be able to disintegrate in different ways (for example, by emission of a proton, a neutron, or an α particle). According to this model each mode of disintegration has a certain probability which is independent of the mode of formation of the compound nucleus.

The Bohr picture is definitely statistical in nature and can therefore be expected to be valid only for nuclei containing a large number of nucleons and for excitation to high energies where the nuclear energy levels are closely spaced. Although there is at present very little detailed knowledge of nuclear energy levels we can say that, in general, the level density increases with increasing mass number (that is, with increasing complexity of the nuclear system) and with increasing excitation energy. Average level spacings (in electron volts) appear to be about as follows:

	Near Ground State	At 8 Mev	At 15 Mev
Light nuclei ($A \sim 10$)	$\sim 10^6$	10^4–10^5	10^3
Heavy nuclei ($A \sim 150$)	$\sim 10^5$	10–100	10^{-2}–1

There is evidence that the individual level widths increase with increasing excitation energy, so that there is a great deal of overlapping of levels in medium and heavy nuclei at excitation energies of 12 or 15 Mev. The experimental evidence on level spacings and level widths comes from γ-ray spectra for the region near the ground state and from resonance capture processes (see section D below) for excitation energies in the neighborhood of 8 Mev.

Competition among Different Reactions. The Bohr theory suggests that a given compound nucleus may break up in several different ways. This is in agreement with the experimental observation that the bombardment of a given nuclide with one type of nuclear particle of one energy usually leads to a variety of

products. For example, the bombardment of Al^{27} with fast neutrons (say 10 Mev) produces the radioactive products $_{13}Al^{26}$, $_{13}Al^{28}$, $_{12}Mg^{27}$, and $_{11}Na^{24}$; according to Bohr's picture this means that the compound nucleus $_{13}Al^{28*}$ is formed and that it may break up in any one of the following ways (the asterisk indicates a high state of excitation of the compound nucleus):

$$_{13}Al^{27} + {}_0n^1 \longrightarrow {}_{13}Al^{28*} \begin{cases} \longrightarrow {}_{13}Al^{26} + 2\,{}_0n^1 \\ \longrightarrow {}_{13}Al^{28} + \gamma \\ \longrightarrow {}_{12}Mg^{27} + {}_1H^1 \\ \longrightarrow {}_{11}Na^{24} + {}_2He^4 \end{cases}$$

The Bohr picture permits some tentative predictions about the relative probability of various competing reaction types and their energy dependence, and, within the limitations to be discussed later, these predictions are in accord with experimental evidence. For example, the theory predicts that for a compound nucleus which has been formed by the entry of a thermal neutron into the target nucleus the probability of emission of any nuclear particle should be extremely low; for neutron emission the entire binding energy contributed by the incident neutron would have to be concentrated on one neutron again, and proton emission would require the concentration on one proton of a comparable binding energy plus the barrier energy. The compound nucleus from thermal-neutron capture generally gives up its excess energy in electromagnetic radiation (γ rays). Indeed almost the only reaction observed with slow neutrons is the n, γ process, often referred to as radiative capture. A few exceptions are found among reactions with the light nuclei in cases where the binding energy of a proton or α particle is appreciably lower than that of a neutron: the reactions B^{10} (n, p) Be^{10}, N^{14} (n, p) C^{14}, Cl^{35} (n, p) S^{35}, B^{10} (n, α) Li^7, and Li^6 (n, α) H^3 occur with thermal neutrons.

As the excitation energy of the compound nucleus increases above the binding energy of the most loosely bound particle the probability for γ-ray emission becomes small relative to the probability for heavy-particle emission. For this reason radiative capture is important only for neutrons (which can enter a nucleus

without kinetic energy) and is rarely observed for charged particles. The type of heavy particle emitted depends not only on the binding energy but also on the height of the potential barrier relative to the excitation energy. For moderate excitation energies the emission of neutrons is favored over the emission of protons, and that in turn is more probable than α-particle emission. For example, at moderate bombarding energies α, n reactions have much larger probabilities than α, p reactions. With increasing excitation energy the effect of the barrier becomes less pronounced.

At excitation energies above about 15 Mev the competition among different reactions becomes even more complex because the emission of a single particle (say a neutron) may leave the nucleus still in a sufficiently high state of excitation to "boil off" a second particle. Thus reactions of the types $(n, 2n)$, $(d, 2n)$, $(\alpha, 2n)$, (n, np), (d, np) will appear and compete with the simpler reactions (d, n), (α, n), (n, p), etc. At sufficiently high excitation energies one would expect the probability of single-particle emission to drop owing to the competition of the two-particle emissions, and this again agrees with experimental results.

Excitation Functions. A plot of reaction yield versus energy of the incident particle is called an excitation function. To illustrate the effect of competition, figure III–1 shows the excitation functions of some reactions produced by α-particle bombardment of silver.[6] The reaction yield determined from the amount of radioactive product formed is plotted (in arbitrary units) against the α-particle energy in million electron volts. The production of In^{110} has an observed threshold at about 11 Mev, presumably due to the reaction Ag^{107} (α, n) In^{110}. The In^{110} yield goes through a maximum at 17.5 Mev and drops off rapidly after that; this decrease in the α, n yield coincides with a rise in the yield of the competing reaction Ag^{107} $(\alpha, 2n)$ In^{109} which has its observed threshold at 13.5 Mev. However, after going through a minimum at about 24 Mev, the In^{110} yield increases again, and this second rise can be interpreted as caused by the $\alpha, 3n$ reaction on the other silver isotope Ag^{109}. The excitation function for the Ag^{109} $(\alpha, 2n)In^{111}$ reaction is almost identical with that for Ag^{107} $(\alpha, 2n)$ In^{109} until it reaches a peak at 27 Mev; it drops off when the

[6] S. N. Ghoshal, *Phys. Rev.* **73**, 417 (1948). We are grateful to Mr. Ghoshal for permission to reproduce the figure.

reaction Ag^{109} $(\alpha, 3n)$ In^{110} begins to compete. The fact that the In^{109} yield does not follow this pattern beyond about 25 Mev has been interpreted to mean that the "In^{109} activity" actually includes In^{108} activity which is thought to have a very similar half-life; thus the "In^{109} curve" may really be the sum of the excitation functions for Ag^{107} $(\alpha, 2n)$ In^{109} and Ag^{107} $(\alpha, 3n)$ In^{108}.

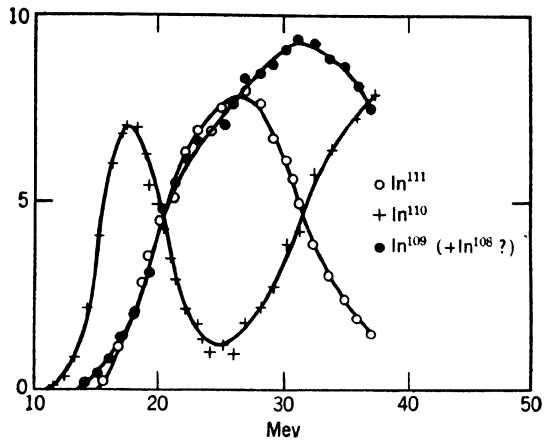


FIGURE III–1. Excitation functions for (α, n), $(\alpha, 2n)$, and $(\alpha, 3n)$ reactions in silver. Energy of the incident α particles in million electron volts is plotted on the abscissa; yield in arbitrary units on the ordinate. The three curves represent the yields of the various indium isotopes as indicated.

Deviations from the Bohr Mechanism with High-energy Projectiles. The Bohr picture leads one to believe that with higher and higher bombarding energies more and more complex reactions may be expected to replace the simpler ones; this expectation is again borne out by experiment, at least with heavy-particle projectiles up to 40 or 50 Mev. Reactions such as $(d, 4n)$, $(d, p3n)$ and $(\alpha, 6n)$, $(\alpha, p5n)$ have been obtained in the heaviest elements with the 20-Mev deuterons and 40-Mev α particles available from the Berkeley 60-inch cyclotron; similarly complex reactions have been produced by high-energy photons from the Schenectady 100-Mev betatron. More recently the 190-Mev deuterons and 380-Mev α particles from the new 184-inch cyclotron in Berkeley have been used for transmutation experiments, and it appears that with the excitation energies thus available the mechanism of the reactions is no longer entirely the one described by the Bohr

theory. Actually reactions of the type predicted by the Bohr theory, with the emission of a large number of light fragments because of the high excitation energies, are observed. (These have been termed "spallation reactions." As produced with 190-Mev deuterons these processes have been reported to result in decreases of as many as 30 units of mass and 14 units of charge.) However, at the same time the whole spectrum of simpler reactions also occurs, with probabilities very much larger than the Bohr theory would permit. In fact the excitation function [6] of the reaction C^{12} (p, pn) C^{11} shows a rise up to a proton energy of about 60 Mev and then stays constant up to 140 Mev. Similar curves were found [7] for the reactions C^{12} (d, dn) C^{11} with deuterons up to 190 Mev and C^{12} $(\alpha, \alpha n)$ C^{11} with α particles up to 380 Mev.

The following explanation for the observed phenomena in the high-energy region under discussion has been given by R. Serber. A nucleon with an energy of about 100 Mev or more will have a mean free path in nuclei comparable to nuclear dimensions, and, therefore, nuclei will not be entirely opaque for such particles, and the compound nucleus picture becomes inapplicable. Furthermore, the impinging nucleon will transfer only a small part of its kinetic energy to a nucleon struck by it and will therefore, in general, have more than enough energy left to escape from the nucleus, even after one or two collisions. Thus the particular mechanism depends on whether the impinging particle strikes the nucleus near the periphery (in which case it may escape after a single collision and transfer only about 25 Mev to the nucleus) or near the center (in which case it may be stopped in the nuclear matter by several collisions and transfer its entire kinetic energy to the nucleus). Clearly there are intermediate possibilities, and according to this picture it is reasonable to expect a variety of modes of disintegration corresponding to the different possible excitation energies of the compound nucleus, all of which may have comparable probabilities. The reactions (p, pn), (d, dn), and $(\alpha, \alpha n)$ previously mentioned are then thought to be brought about by collisions in which the incident particle takes away most of its energy, leaving the nucleus sufficiently excited to boil off a neutron. The probability of this type of reaction depends on

[6] W. W. Chupp and E. M. McMillan, *Phys. Rev.* **72**, 873 (1947).

[7] L. R. Thornton and R. W. Senseman, *Phys. Rev.* **72**, 872 (1947).

the mean free path of the incident particle in the nucleus, which varies only slowly with energy at the high energies under discussion; this accounts for the observed flatness of the excitation functions. Deuterons and α particles may be considered in terms of their individual nucleons because their kinetic energies are large compared with their binding energies.

An additional observation made in connection with the high-energy reactions is that the incident nucleon may undergo an exchange collision: a proton may emerge as a neutron and vice versa. This result is apparent in the angular distribution of emitted particles and represents one of the most direct pieces of evidence for a charge exchange nature of nuclear forces. Other recent experiments in Berkeley have demonstrated the production of mesons in nuclear reactions at these very high energies.

Types of Reactions. Returning now to reactions in the more usual energy range [8] we shall discuss a few particular types of reactions because of their special interest. Bombarding particles which have been used to effect nuclear reactions are neutrons, protons, deuterons, H^3 nuclei, α particles, γ rays (and X rays), and electrons, and the same particles except electrons and deuterons are commonly observed among the fragments produced in nuclear reactions.

The n, γ reaction has already been mentioned as the only type commonly occurring with slow neutrons. The reaction is always exoergic and occurs with very nearly every target. In about 150 cases it is known to lead to radioactive products. This reaction type is particularly important for the production of radioactive isotopes, because of the relatively high reaction yields and because of the enormous neutron fluxes now available in the chain-reacting piles.

Inelastic Scattering. Reactions in which the incident and emitted particles are of the same type—(n, n), (p, p), (γ, γ), etc.—

[8] The overwhelming majority of the artificially produced radioactive species now known (and listed in table A in the appendix) have been produced by relatively simple reactions involving the emission of one or two or at most three light fragments. Furthermore, it seems almost certain that for the production of practical amounts of radioactive tracers these simple reactions will continue to be used. However, the number of known radioactive species will undoubtedly increase considerably with the use of bombarding particles of higher and higher energies. In general, these will be further away from the region of stable nuclides and will tend to have short half-lives.

lead to an excited state of the initial nucleus which usually reverts to the ground state by γ emission. The outgoing particle has less energy than the incident one, and, therefore, such a process is referred to as inelastic scattering. In general, this type of reaction can be detected only by a measurement of the energy of the emitted (inelastically scattered) particle, but occasionally the residual nucleus is left in a metastable state of measurable lifetime; that is, an isomer of a stable nucleus is formed. More than 20 isomers of stable nuclei [9] are now known, among them Sr^{87*} ($t_{1/2}$ = 2.7 hr), In^{115*} (4.5 hr), Ta^{181*} (22×10^{-6} sec), Pb^{204*} (68 min). The n, n reaction is the most probable process with neutrons between a few hundred kev and a few Mev because in this region the n, γ reaction is no longer so important, and the n, p reaction cannot compete favorably because of the potential barrier. The p, p reaction is not very prevalent at any energy because of competition from the p, n reaction. For the same reason d, d and α, α reactions are rarely observed. Excitation to isomeric levels by γ, γ reactions and even by e^-, e^- reactions has been observed; in the latter case the excitation is not caused by an amalgamation of the electron with the nucleus, but by interaction between the nucleus and the electromagnetic field of the high-speed electron. The e^-, ne^- reactions recently observed [10] in exceedingly small yields in Cu^{63}, Ag^{107}, and Ag^{109} probably occur by a similar mechanism.

Oppenheimer–Phillips Process. The d, p reaction deserves special mention because it occurs very commonly and sets in at much lower energies than one would expect from the Bohr theory. In fact, the observed d, p thresholds are usually even lower than the corresponding d, n thresholds. This apparent anomaly has been explained by Oppenheimer and Phillips [11] as being due to the polarization of the deuteron by the Coulomb field of the nucleus. As the deuteron approaches the nucleus, its "neutron end" is thought to be turned toward the nucleus, the "proton end" being repelled by the Coulomb force. Because of the relatively large neutron–proton distance in the deuteron (several times 10^{-13} cm),

[9] An asterisk after the mass number is often used to indicate an excited isomeric state.

[10] L. S. Skaggs, J. S. Laughlin, A. O. Hanson, and J. J. Orlin, *Phys. Rev.* **73**, 420 (1948).

[11] J. R. Oppenheimer and M. Phillips, *Phys. Rev.* **48**, 500 (1935).

the neutron reaches the surface of the nucleus while the proton is still outside most of the potential barrier. Since the binding energy of the deuteron is only 2.18 Mev, the action of the nuclear forces on the neutron tends to break up the deuteron, leaving the proton outside the potential barrier. The process just described is now generally called an Oppenheimer–Phillips (or O–P) process. An analogous mechanism appears to be responsible for the H^3, p reaction recently reported.[12] An interesting feature of the O–P process is that the emergent protons have a spread of energies which includes values in excess of the incident deuteron energy, so that in a small fraction of the cases the excitation of the compound nucleus is that which would result from the capture of a neutron of negative kinetic energy.

Fission. Finally we mention a reaction of special practical significance, the fission process. By fission is meant the breakup of a heavy nucleus into two or more medium-heavy fragments. The process is usually accompanied by the emission of neutrons and much more rarely by the emission of α particles and possibly other light fragments. Fission has been produced in some nuclides (notably U^{235}, U^{238}, and Th^{232}) by neutrons, protons, deuterons, α particles, and γ rays, and more recently such elements as tantalum, platinum, thallium, lead, and bismuth have been found to undergo fission with 200-Mev deuterons and 400-Mev α particles. By far the most important of these reactions is neutron-produced fission. The species ${}_{92}U^{235}$ and ${}_{94}Pu^{239}$ undergo fission with thermal or with fast neutrons, whereas fission of ${}_{90}Th^{232}$, ${}_{91}Pa^{231}$, and ${}_{92}U^{238}$ requires fast neutrons. The fission process may occur in many different modes, and a very large number of fission products are known, ranging from $Z = 30$ (zinc) to $Z = 63$ (europium), and from $A = 72$ to $A = 158$ for the case of U^{235} neutron fission. Because of the fact that the neutron excess required for stability is much greater in the region of the heaviest elements than in the fission-product region, the primary fission products have neutron excesses far greater than the stable isotopes of the same elements. These primary fission products achieve stability through successive β^- decays; some chains with as many as six successive β^- decays are known. The yields of fission

[12] D. N. Kundu and M. L. Pool, *Phys. Rev.* **73**, 22 (1948).

products plotted against mass number (figure III-2) show two peaks separated by a very pronounced minimum; for slow-neutron fission of U^{235} the maxima in the yield curve occur at $A = 95$

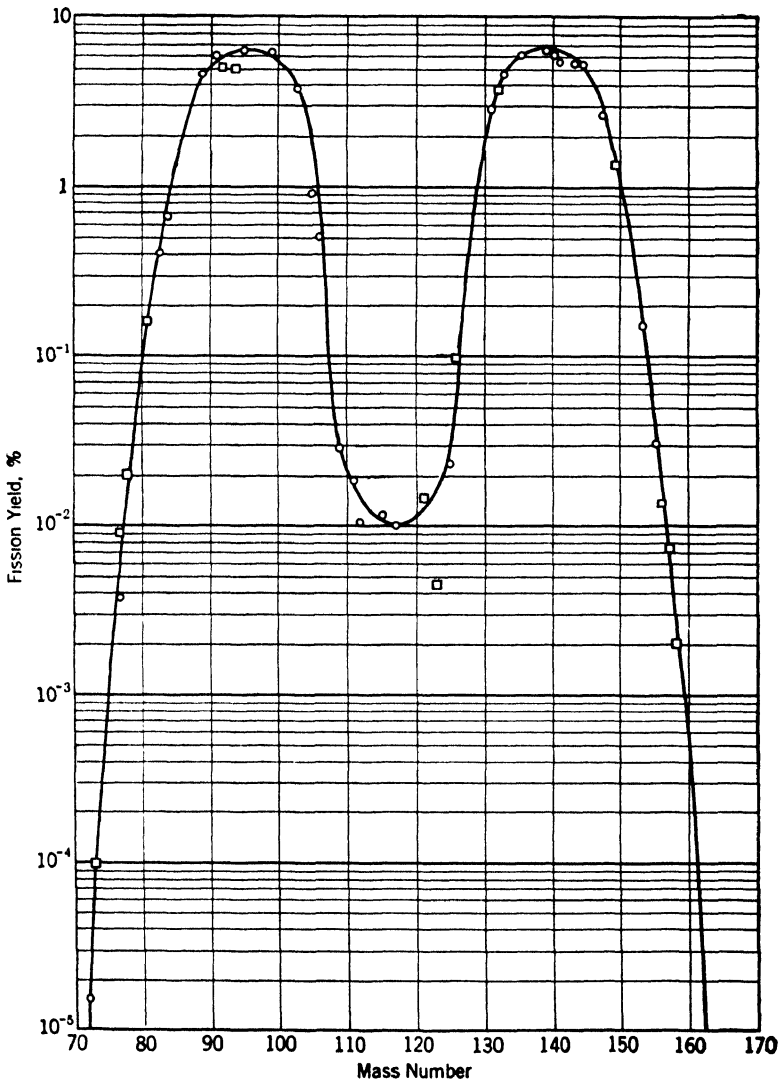


FIGURE III-2. Yields of fission-product chains as a function of mass number (for the slow-neutron fission of U^{235}). ○ Mass assignment certain. □ Mass assignment uncertain.

and $A = 139$, and the fission yield at mass 117 (at the minimum) is about 600 times smaller than at the peaks. In other words, fission is preferably asymmetric. The asymmetry appears to become less pronounced with higher excitation energy. The fission process is accompanied by the very large energy release of about 200 Mev. The unique importance of the fission reaction is mainly due to the release of more than one neutron in each neutron-produced process, which makes a divergent chain reaction possible.

The analogy between a nucleus and a liquid drop which Bohr had used when he proposed the idea of the compound nucleus can be extended to explain fission at least in a qualitative way. A heavy nucleus is held together by nuclear forces analogous to the cohesive forces holding together a liquid drop. Just as a liquid drop tends to assume spherical shape under the action of surface tension, the unsaturation of the nuclear forces at the surface makes a sphere (which has the smallest surface for a given volume) the most stable configuration for a heavy nucleus. Bohr and J. A. Wheeler have shown that there will be a certain critical size for nuclei, depending on Z^2/A, above which the force of electrostatic repulsion will be greater than the surface forces holding the nucleus together. This critical size has been calculated to occur for Z somewhere near 100, and it is therefore reasonable that for a nucleus only slightly below this limit of stability a small excitation should be sufficient to induce breakup into two fragments. Bohr and Wheeler have calculated the energetic conditions for fission of various heavy nuclear species on the basis of this model, and their theory is in fair agreement with the facts. They were able to predict the fission of Pa^{231} and to estimate its threshold energy before the reaction had been discovered.

Fission is not the only reaction that can occur with the heaviest elements. Rather, it competes with other reactions such as radiative capture and inelastic scattering, and the compound nucleus picture appears to account adequately for the relative probabilities of the competing processes at moderate excitation energies.

D. CROSS SECTIONS

Definitions. We shall now turn to a more quantitative consideration of reaction probabilities. The probability of a nuclear process is generally expressed in terms of a cross section σ which has the dimensions of an area. This originates from the simple

picture that the probability for the reaction between a nucleus and an impinging particle is proportional to the cross-sectional target area presented by the nucleus. Although this picture certainly does not hold for reactions with charged particles which have to overcome Coulomb barriers or for slow neutrons (it does hold fairly well for the total probability of a fast neutron interacting with a nucleus), the cross section is a very useful measure of the probability for any nuclear reaction. For a beam of particles striking a thin target, that is, a target in which the beam is attenuated only infinitesimally, the cross section for a particular process is defined by the equation,

$$N = \mathrm{I}n\sigma x, \tag{III-1}$$

where N is the number of processes of the type under consideration occuring in the target,

I is the number of incident particles,

n is the number of target nuclei per cubic centimeter of target,

σ is the cross section for the specified process, expressed in square centimeters, and

x is the target thickness in centimeters.

The total cross section for collision with a fast particle is never greater than the geometrical cross-sectional area of the nucleus, and therefore fast-particle cross sections are rarely as large as 10^{-24} cm^2 (radii of the heaviest nuclei are about 10^{-12} cm). Therefore, a cross section of 10^{-24} cm^2 is considered "as big as a barn" and 10^{-24} cm^2 has been named the barn, a unit often used in expressing cross sections.

If instead of a thin target we consider a thick target, that is, one in which the intensity of the incident particle beam is attenuated, then the attenuation $-d\mathrm{I}$ in the infinitesimal thickness dx is given by the equation,

$$-d\mathrm{I} = \mathrm{I}n\sigma\, dx,$$

where σ must be the total cross section. If we are able to neglect the variation in σ as the incident particles traverse the target, which is often the case for neutron reactions, we may obtain, by integration,

$$\mathrm{I} = \mathrm{I}_0 e^{-n\sigma x},$$

$$\mathrm{I}_0 - \mathrm{I} = \mathrm{I}_0(1 - e^{-n\sigma x}), \tag{III-2}$$

where **I** is the intensity of the beam after traversing a target thickness x, $\mathbf{I}_0$ is the incident intensity, and $\mathbf{I}_0 - \mathbf{I}$ is the number of reactions occurring.

As an illustration we shall calculate the number of radioactive Au^{198} nuclei produced per second in a sheet of gold 0.3 mm thick and 5 cm^2 in area exposed to a thermal neutron flux of 10^7 neutrons per cm^2 per sec. The capture cross section of Au^{197} for thermal neutrons is 95 barns, and we neglect any other reactions of neutrons with gold. The density of gold is 19.3 g per cm^3, and its atomic weight is 197.2; therefore,

$$n = \frac{19.3}{197.2} \times 6.02 \times 10^{23} = 5.89 \times 10^{22} \text{ } Au^{197} \text{ nuclei per } cm^3;$$

$$x = 0.03 \text{ cm};$$

$$\mathbf{I}_0 = 5 \times 10^7 \text{ incident neutrons per sec.}$$

Therefore, according to equation III–2:

$$\mathbf{I}_0 - \mathbf{I} = 5 \times 10^7(1 - e^{-5.89\times10^{22}\times95\times10^{-24}\times0.03})$$

$$= 5 \times 10^7(1 - e^{-0.168})$$

$$= 7.8 \times 10^6 \text{ } Au^{198} \text{ nuclei formed per sec.}$$

Partial and Total Cross Sections. A cross section may be given for any particular nuclear process. For example, the total cross section for absorption of 10-Mev neutrons by a particular nuclear species may be measured. This corresponds to the cross section for formation of the compound nucleus; but that compound nucleus may break up in various ways, and one may therefore wish to define partial cross sections for particular processes such as (n, n), (n, γ), (n, p), and (n, α) reactions. The sum of all the partial cross sections equals the total cross section. We have already indicated that in equation III–2 total cross sections should be used, and only the total number of reactions may be obtained directly. This may be multiplied by the ratio of a partial cross section to the total cross section to obtain the number of reactions of a particular kind. The partial cross section might be for a single isotope, but the total cross section must be that for the target substance.

The total cross section for fast-neutron reactions is approximately equal to the geometrical cross section of the nucleus, πR^2, and nuclear radii R have been determined by measurements of total fast-neutron cross sections. For charged particles the total cross section is generally less than πR^2 because of the potential barrier. Charged particles with energies near the barrier height often have σ's in the neighborhood of 10^{-1} barn. Gamma-ray cross sections are commonly observed in the range of 10^{-4} to 10^{-1} barn.

Slow-neutron Cross Sections. The cross section for capture of slow neutrons is in many cases much greater than the nuclear geometrical cross section πR^2. This is explained by the fact that the de Broglie wave length λ of a thermal neutron is much larger than nuclear dimensions. One would expect the theoretical upper limit for slow-neutron cross sections to be something less than λ^2 or about 10^{-16} cm^2 (10^8 barns). Some of the largest thermal-neutron cross sections observed are those of gadolinium (30,000 barns), samarium (6500 barns), and cadmium (2900 barns). These values are average cross sections for the natural mixtures of isotopes; since in each case one or two particular isotopes seem to be responsible for the large cross section the isotopic cross sections are even bigger (for $Cd^{113}\sigma_{\text{thermal}} \approx 23{,}000$ barns). Cadmium is commonly used as an absorber of slow neutrons. The slow-neutron cross section varies in a quite irregular way from element to element. Data on neutron cross sections of most elements are now available in the literature. A number of thermal-neutron activation cross sections are listed in table B in the appendix.

One-over-v Law and Resonance Processes. For neutron energies in the thermal region, and somewhat above, the cross sections for many substances are found to obey a $1/v$ law; that is, they vary inversely with the neutron velocity. For some light elements the $1/v$ law is valid over a wide energy range. Neutron capture in B^{10}, for example, shows a $1/v$ dependence up to about 50 kev; neutron energies below that limit can therefore be measured by absorption in boron.

In practically all elements there are deviations from the $1/v$ law in one or more energy regions due to the existence of resonance levels. If the neutron entering a nucleus has an energy which

will just bring the compound nucleus into one of its energy levels, the cross section will be particularly high. Reactions occurring in this way are called resonance processes. If a cross section is measured as a function of neutron energy a curve such as the one in figure III–3 may be obtained, with resonance peaks superimposed on the $1/v$ curve. It is from the spacings and widths of these resonance peaks that conclusions can be drawn about the

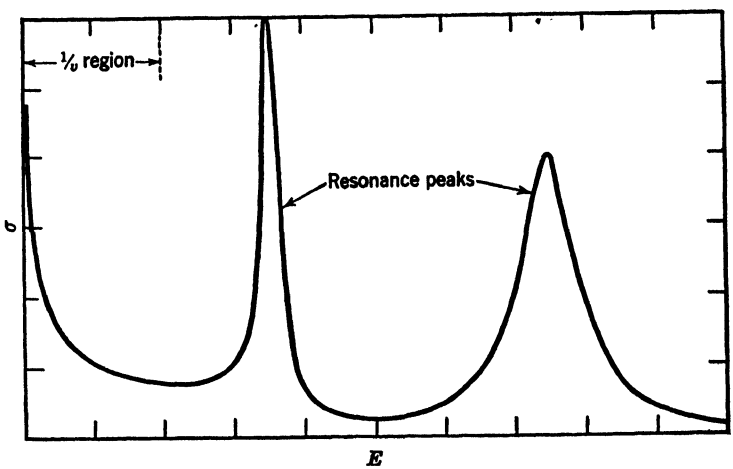


FIGURE III–3. Typical curve of neutron cross section versus neutron energy.

level densities and level widths in nuclei with excitation energies in the neighborhood of 6 to 8 Mev (the binding energy of a neutron). For light elements resonances have also been observed with protons and α particles. Some typical neutron resonances are at 1.44 ev in indium ($\sigma_{res} \approx 26{,}000$ barns), at 5.5 ev in silver ($\sigma_{res} \approx 7200$ barns) and at about 100 kev in lithium; the large thermal-neutron cross section of cadmium is due to a resonance at 0.18 ev ($\sigma_{res} \approx 7250$ barns). The energy widths of resonance levels vary over wide limits; in general, the levels are narrower at low than at high energies. In many elements (including indium and rhodium) resonance capture leads to the formation of a radioactive isotope, and such substances are useful as detectors for neutrons of particular energies. Resonances occur not only for capture but also for scattering processes.

EXERCISES

1. Compute, from masses in table A in the appendix, the Q values for the following reactions:

(a) Mg^{24} (d, p) Mg^{25}

(b) B^{10} (n, α) Li^7

2. Would the Ca^{43} (n, α) reaction go with thermal neutrons? Justify your answer. *Answer:* No.

3. By what nuclear reactions may I^{129} be made? What are some important considerations that would determine the best practical choice?

4. In the reaction N^{14} (n, p) C^{14} 0.60 Mev is liberated. What is the mass of the nucleus C^{14}?

5. Show from conservation of momentum that in any nuclear reaction $A(a, b)B$ the fraction of the kinetic energy of the bombarding particle a which goes into kinetic energy of the products is $M_a/(M_a + M_A)$, where M_a and M_A are the masses of a and A, respectively.

6. Calculate the approximate heights of the potential barriers around $_{13}Al^{27}$, $_{26}Fe^{56}$, $_{47}Ag^{107}$, $_{73}Ta^{181}$, and $_{92}U^{238}$ for protons.
Answers to first two: 3.1, 5.2 Mev.

7. Calculate the de Broglie wave length of a neutron of (a) 1 ev, (b) 1 kev, (c) 1 Mev energy. *Answer:* (b) 0.9×10^{-10} cm.

8. What are the average energies (in electron volts) of thermal neutrons at 25°C and at −196°C (liquid nitrogen temperature)?

9. Estimate (from Coulomb barrier considerations) the minimum total kinetic energy (in Mev) of the two fission fragments obtained when a uranium nucleus splits into (a) krypton and barium (b) rhodium and silver.
Answer: (a) 198 Mev.

10. Using the table of nuclear species in the appendix, decide what reactions would be practical for the production of (a) Mn^{53}, (b) Gd^{159}, (c) Tc^{100}, if you had at your disposal 12-Mev deuterons, 6-Mev protons, 24-Mev α particles, and neutrons up to 16 Mev.

11. Approximately what thickness of cadmium (σ_{th} = 2900 barns) is necessary to reduce a beam of thermal neutrons to 0.1 per cent of its intensity?

12. At room temperature (say 27°C) a beam of neutrons is brought to thermal equilibrium in graphite. This beam then falls on a thin boron absorber, σ_{th} for boron = 703 barns. The beam is reduced to 90 per cent

of its original intensity by the absorber. What would have been the intensity reduction if the entire experiment had been performed at 327°C?

Answer: To 93 per cent.

13. The nuclide Cl^{33} can be produced by the reaction S^{33} (p, n) Cl^{33}. The Cl^{33} emits positrons with an upper energy limit of 4.13 Mev. What is the Q value of the reaction? What is the height of the potential barrier around the S^{33} nucleus for the proton? Estimate the minimum proton energy required to produce the reaction.

Answer: Minimum proton energy = 6.1 Mev.

14. With the very crude assumption that in the highly excited compound nucleus the energy partition between nucleons is the same as in an ideal gas, estimate the temperature of a heavy nucleus (say $A = 150$) after the capture of a 10-Mev neutron. *Answer:* About 10^9 °K.

15. ${}_{18}A^{35}$ decays to the ground state of ${}_{17}Cl^{35}$ with the emission of positrons of 4.4 Mev maximum energy. Without using the masses of A^{35} and Cl^{35}, calculate the Q value of the reaction Cl^{35} $(d, 2n)$ A^{35}.

Answer: −8.37 Mev.

16. (*a*) The measured cross section for absorption of thermal neutrons in B^{10} is 3500 barns at 300°K. Assuming the $1/v$ law to hold, find the discrete velocity v' at which $\sigma = 3500$ barns.

(*b*) Using the answer to (*a*) estimate the mean life of a thermal neutron in 0.1 M borax ($Na_2B_4O_7 \cdot 10H_2O$) solution. You may assume B^{10} to be the only capturing material.

Answer: (*a*) 1.97×10^5 cm per sec; (*b*) 3.2×10^{-5} sec.

17. With deuteron energies up to 100 Mev available, how would you propose to make each of the following nuclides with deuteron-induced reactions: Ti^{43}, Gd^{150}, Pb^{200}, Eu^{158}, La^{143}? Give specific answers, in terms of targets and deuteron energies. State your reasoning.

18. Ordinary microbalances have a capacity of 20 g and a sensitivity of about 1 μg. How much better would the sensitivity have to be to permit detection of the mass change accompanying the reaction $Na + \frac{1}{2}Cl_2 = NaCl$?

19. The energy liberated in the sun is known to be the result of a cycle of nuclear reactions. We may think of the first step as the radiative capture of a high-speed proton (a thermal proton in the very hot interior of the sun) by a C^{12} nucleus. The resultant N^{13} is a positron emitter, half-life 10 min, and so transforms into C^{13}, which in turn undergoes a p, γ reaction. With the help of table A in the appendix, can you complete this mechanism to obtain the net reaction, that is, the transformation of four hydrogen atoms into one helium atom? You may want to refer to H. A. Bethe, "Energy Production in Stars," *Phys. Rev.* **55,** 103 and 434 (1939).

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IV. SOURCES OF BOMBARDING PARTICLES

A. HEAVY CHARGED PARTICLES

Natural Sources of α Particles. From the discovery of nuclear transmutations in 1919 until 1932 the only known sources of particles which would induce nuclear reactions were the natural α emitters. In fact the only type of nuclear reaction known during that period of 13 years was the α, p reaction. The natural α-particle sources most frequently used in transmutation experiments were Po^{210} (5.30 Mev, $t_{1/2}$ = 140 days) and RaC′ (7.68 Mev, $t_{1/2} = 1.5 \times 10^{-4}$ sec) used in equilibrium with its β-emitting parent RaC. Today natural α-particle sources for nuclear reactions are chiefly of historical interest because of the much higher intensities and higher energies now available from man-made accelerators for charged heavy particles.

Voltage Multiplier. The first transmutation by artificially accelerated particles was achieved in 1932 when J. D. Cockroft and E. T. Walton bombarded lithium with protons from their voltage-multiplying rectifier set at the Cavendish Laboratory. The reactions were Li^6 (p, α) He^3 and Li^7 (p, α) He^4; the protons used in the initial studies had energies of 100 to 500 kev. The Cockroft–Walton type of machine consists essentially of a moderately high-voltage a-c transformer with an arrangement of vacuum-tube rectifiers and filter condensers so that the d-c output voltage is several times greater than the peak alternating voltage. With this type of equipment to supply the high voltage, protons have been accelerated up to about 1 Mev, and currents as high as 200 μa have been obtained.

Cascade Transformer. C. C. Lauritsen and his coworkers at the California Institute of Technology adapted multistage transformers to the acceleration of positive ions. In this arrangement a number of high-voltage transformers are placed in cascade (to reduce transformer-winding insulation requirements) by having each primary winding excited by a portion of the preceding secondary winding. Units with four or five stages and with 200 or 250 kv

per stage have been built, giving a maximum potential difference of about 1 million volts.

The cascade transformer is entirely an a-c device, and, therefore, positive ions can be accelerated only during half a cycle; in fact only during a small fraction of the cycle can they be accelerated to the maximum energy. To obtain nearly monoenergetic beams an arrangement is used whereby the ions are admitted to the accelerating tube during the appropriate fraction of the positive half-cycle. Ion currents of about 30 μa have been obtained with cascade transformers.

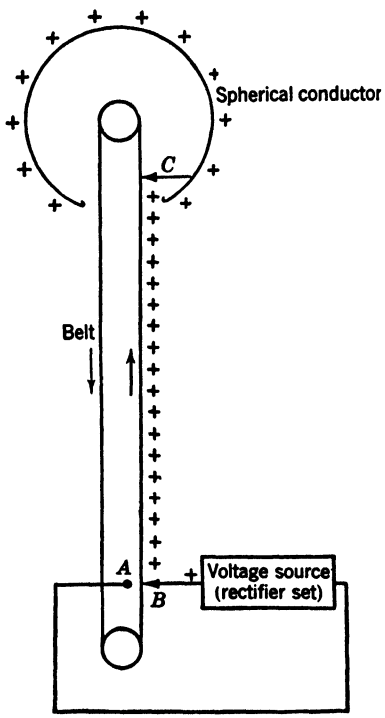


FIGURE IV–1. Schematic representation of the charging mechanism of a Van de Graaff generator.

Electrostatic (Van de Graaff) Generator. The adaptation of the electrostatic machine to the production of high potentials for the acceleration of positive ions was pioneered by R. J. Van de Graaff of the Massachusetts Institute of Technology, beginning in 1931. In the Van de Graaff machine a high potential is built up and maintained on a conducting sphere by the continuous transfer of static charges from a moving belt to the sphere. This is illustrated in figure IV–1. The belt, made of silk, rubber, paper, or some other suitable insulator, is driven by a motor and pulley system. It passes through the gap AB which is connected to a high-voltage source (10,000 to 30,000 volts d-c, usually from a vacuum-tube rectifier arrangement) and adjusted so that a continuous discharge is maintained from the sharp point B. Thus positive (or negative) charges are sprayed from B onto the belt which carries them to the interior of the insulated metal sphere; there another sharp point or sharp-toothed comb C connected to the sphere

takes off the charges and distributes them to the outside surface of the sphere. (Because of their coulombic repulsion the charges always go to the outside of a spherical conductor.) The sphere will continue to charge up until the loss of charge from the surface by corona discharge and by leakage along its insulating support balances the rate of charge transfer from the belt. The continuous current that can be maintained with an electrostatic generator depends on the rate with which charge can be supplied to the sphere. The use of wide belts (2 to 4 feet) driven at high speeds is therefore indicated.

Van de Graaff's first installation consisted of two spheres, each 2 feet in diameter; one charged to a positive potential of about 750,000 volts above ground and the other to an equal negative potential. All recent electrostatic generators use a single electrode, with acceleration of the ions between that electrode and ground potential, because considerable practical advantages are gained if much of the auxiliary equipment can be operated at ground potential.

Since the voltage of an electrostatic generator is limited by the breakdown potential of the gas surrounding the charged electrode it is desirable to use conditions under which this breakdown potential is as high as possible. The breakdown potential is a function of pressure and goes through a minimum at a rather low pressure (small fraction of an atmosphere). It is therefore advantageous to operate an electrostatic generator either in a high vacuum, which presents formidable difficulties, or in a high-pressure atmosphere. R. G. Herb and his associates at the University of Wisconsin have developed electrostatic generators completely enclosed in steel tanks in which pressures of several atmospheres are maintained. This has considerably increased the potentials attainable. A further improvement is the use of gases which have higher breakdown potentials than air. Admixtures of carbon tetrachloride and Freon to air or nitrogen, and more recently the use of sulfur hexafluoride, have been successful in increasing voltages of existing generators. A number of pressure-type electrostatic generators capable of accelerating protons (or other positive ions) to 2 to 5 Mev are in operation, and it appears likely that energies of 10 to 12 Mev may be reached with larger machines of similar design. Proton currents of 5 to 100 μa are common. The chief application of machines of this type is in nuclear physics work

requiring high precision because unlike other machines such as cyclotrons they supply ions of precisely controllable energies (constant to about 0.1 per cent).

Accelerating Tubes. We have so far discussed methods of obtaining high voltages for the acceleration of positive ions without going into any detail about the accelerating tube across which the potential is applied. The need for such a tube is common to all the types of machines described so far. A source of ions near

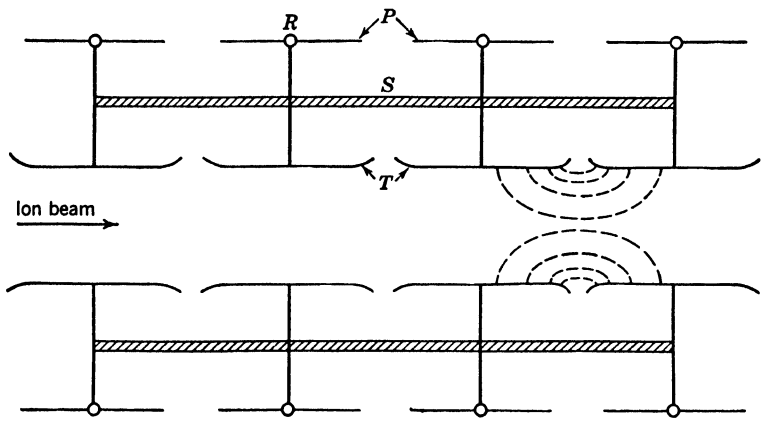


FIGURE IV–2. Schematic diagram of a portion of an accelerating tube.

the high-voltage end, a system of accelerating electrodes, and a target at the low-voltage end must be provided and enclosed in a vacuum tube connected to the necessary pumping system. The ion source is essentially an arrangement for ionizing the proper gas (hydrogen, deuterium, helium) in an arc or electron beam; the ions are drawn through an opening into the accelerating system. A typical accelerating tube (figure IV–2) is built of glass or porcelain sections S. Inside this tube, sections of metal tube T define the path of the ion beam. Each metal section is supported on a disk which passes between two sections of insulator out into the gas-filled space to a corona ring R which is equipped with corona points P. The purpose of the corona rings and points is to carry the corona discharge from the high- to the low-voltage end of the tube and to distribute the voltage drop uniformly along the tube. Depending on the number of sections used a potential difference somewhere between ten and several hundred kilovolts

exists between successive sections. Each gap between successive sections has both a focusing and a defocusing action on the ions traveling down the tube. The ions tend to travel along the electric lines of force (see figure IV–2 for the pattern of these lines between a pair of sections). In entering the gap the ions are therefore focused, and in leaving it they are defocused; but because the ions move more slowly on entering the gap than on leaving it the focusing effect is stronger than the subsequent defocusing. Well-focused beams (cross-sectional area less than 0.1 cm^2) can be obtained. It should be mentioned that from hydrogen gas in an ion source not only protons but also hydrogen molecule ions (H_2^+), and H_3^+ ions are obtained; these are also accelerated in the tube but can be separated from the protons before striking the target by means of a magnetic analyzer.

In a pressure electrostatic generator the charging system, high-voltage electrode, and accelerating tube are all enclosed in the steel tank containing the high-pressure gas. The high-potential electrode is often more like a cylinder than a sphere.

Linear Accelerator. In all the devices for accelerating ions discussed so far the full high potential corresponding to the final energy of the ions must be provided, and the limitations of this type of device are introduced by the insulation problems. These problems are very much reduced in machines which employ repeated acceleration of ions through relatively small potential differences. The most successful scheme of this type is the cyclotron. But the first device proposed for achieving high-voltage acceleration of ions by multiple application of a much lower voltage was the linear accelerator suggested by R. Wideröe in 1929 and built in the United States by D. H. Sloan and E. O. Lawrence (1931).

In this machine a beam of ions from an ion source is injected into an accelerating tube containing a number of coaxial cylindrical sections (see figure IV–3 for a schematic diagram). Alternate sections are connected together, and a high-frequency alternating voltage from an oscillator is applied between the two groups of electrodes. An ion traveling down the tube will be accelerated at a gap between electrodes if the voltage is in the proper phase. By choosing the frequency and the lengths of successive sections correctly one can arrange the system so that the ions arrive at each gap at the proper phase for acceleration across

the gap. The successive electrode lengths have to be such that the ions spend just one half-cycle in each electrode. Acceleration takes place at each gap, and the focusing action is the same as in the accelerating tubes of high-voltage sets.

In early models of the linear accelerator, mercury ions were accelerated to 2.85 Mev with an input voltage of about 80 kv. However, because the cyclotron was developed almost simultaneously and had obviously great advantages, the linear accelera-

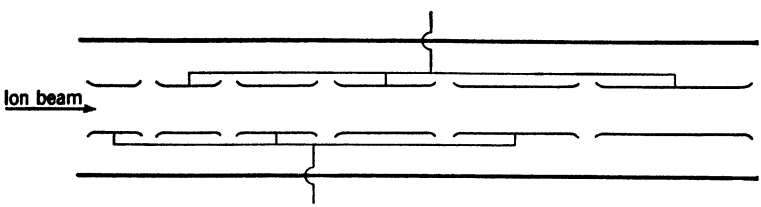


FIGURE IV-3. Schematic diagram of the accelerating tube of a linear accelerator.

tor did not receive much further attention after about 1934. Very recently, interest in modified linear accelerators has been renewed because the development of high-power microwave oscillators apparently offers the possibility of extending the energy range of linear accelerators to the billion-electron-volt (Bev) region. Preliminary work in this field indicates that energy increments of about 1 Mev per linear foot can be achieved. But it appears that for the acceleration, at least of positive ions, to ultrahigh energies other devices are more practical than the linear accelerator.

Cyclotron. By far the most important and successful device to date for accelerating positive ions to millions of electron volts is the cyclotron proposed by Lawrence in 1929. A remarkable development has taken place from the first working model which produced 80-kev protons in 1930 to the giant machine recently put in operation in Berkeley which accelerates α particles to 380 Mev and deuterons to 190 Mev.

In the cyclotron, as in the linear accelerator, multiple acceleration by a radio-frequency potential is used. But the ions, instead of traveling along a straight tube, are constrained by a magnetic field to move in a spiral path consisting of a series of semicircles with increasing radii. The principle of operation is illustrated in

figure IV–4. Ions are produced in an arc ion source *I* near the center of the gap between two hollow semicircular electrode boxes called "dees" (figure IV–5). The dees are enclosed in a vacuum tank, which is located between the circular pole faces of an electromagnet and is connected to the necessary vacuum pumping system. A high-frequency potential supplied by an oscillator is applied between the dees. A positive ion starting from the ion source is accelerated toward the dee which is at negative potential at the

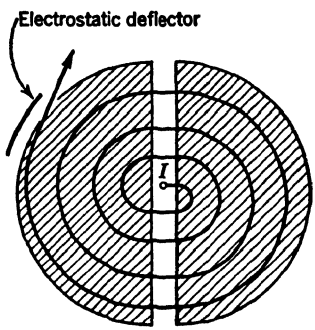


FIGURE IV–4. Ion path in the cyclotron. The magnetic field is perpendicular to the plane of the paper. Shaded area represents the "dees."

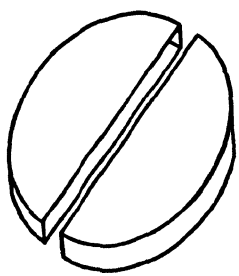


FIGURE IV–5. Perspective view of the dees.

time. As soon as it reaches the field-free interior of the dee, the ion is no longer acted on by electric forces, but the magnetic field perpendicular to the plane of the dees constrains the ion to a semicircular path. If the frequency of the alternating potential is such that the field has reversed its direction just at the time the ion again reaches the gap between dees, the ion again is accelerated, this time toward the other dee. Now its velocity is greater than before, and it therefore describes a semicircle of larger radius; however, as we shall see from the equations of motion, the time of transit for each semicircle is independent of radius. Therefore, although the ion describes larger and larger semicircles it continues to arrive at the gap when the oscillating voltage is at the right phase for acceleration. At each crossing of the gap the ion acquires an amount of kinetic energy equal to the product of the voltage difference between the dees and the ion charge. Finally, as the

ion reaches the periphery of the dee system it is removed from its circular path by a negatively charged deflector plate and is allowed to strike a target.

The equation of motion of an ion of mass M, charge e, and velocity v in a magnetic field H is given by the necessary equality of the centripetal magnetic force Hev and the centrifugal force Mv^2/r where r is the radius of the ion's orbit:

$$Hev = \frac{Mv^2}{r},$$

and, therefore,

$$r = \frac{Mv}{He}. \tag{IV-1}$$

Remembering that the angular velocity $\omega = v/r$, we see that

$$\omega = \frac{He}{M}. \tag{IV-2}$$

From this equation it is evident that the angular velocity is independent of radius and ion velocity and that the time required for half a revolution is constant for ions of the same e/M provided the magnetic-field strength is constant. In practice, the magnetic field is kept constant, e/M is a characteristic of the type of ion used, and therefore ω is constant; the radio frequency has to be chosen such that its period equals the time it takes for the ions to make one revolution. For $H = 15{,}000$ gauss,[1] and e/M for a proton, the revolution frequency $\omega/2\pi$, and therefore the necessary oscillator frequency turns out (from equation IV–2) to be about 23×10^6 cycles per sec. For deuterons or helium ions (He^{++}) at the same H the frequency is half that value. Most cyclotrons are operated as deuteron sources, and they often use r-f oscillators tuned to about 11 or 12 megacycles.

It is clear from equation IV–2 that on a given cyclotron both the magnetic field and the oscillator frequency can be left unchanged when different ions of the same e/M, such as deuterons and α particles, are accelerated. Equation IV–1 shows that the velocity reached at a given radius is the same for ions of the same

[1] Actually to make the equations dimensionally correct, not the field strength H, but the magnetic induction B (maxwells per square centimeter in emu) should be used.

e/M; therefore, α particles are accelerated to the same velocity and, hence, twice the energy as deuterons. To accelerate protons in a cyclotron designed for deuterons either the frequency must be approximately doubled (which is very impractical) or H must be about halved. Although the latter makes inefficient use of the magnet, it is occasionally done, and the final velocity is again the same as for deuterons (see equation IV–1); therefore, protons are accelerated to half the energy available for deuterons.

By squaring equation IV–1 one gets

$$r^2 = \frac{M^2v^2}{H^2e^2} \quad \text{or} \quad \frac{1}{2}Mv^2 = \frac{H^2e^2}{2M}r^2.$$

Thus the final energy attainable for a given ion varies with the square of the radius of the cyclotron. With $H = 15{,}000$ gauss the deuteron energy $E = 0.035r^2$ for E in million electron volts and r in inches.

From the equations of motion it is clear that an ion can reach the dee gap at any phase of the dee potential and still be in resonance with the radio frequency. As we have just derived, the final energy acquired by an ion is entirely independent of the energy increment the ion receives at each crossing of the dee gap. However, in practice it is found that ions making too many revolutions generally do not reach the exit slit because they may be deflected in collisions with residual gas molecules and may strike the surface of the dees. Therefore, only ions which enter the first gap in a favorable phase of the radio frequency (perhaps during about one third of the cycle) contribute to the beam current. To avoid difficulties due to excessively long paths for the ions, rather high dee voltages (20,000 to 50,000 volts) are generally used.

A very important feature of the cyclotron is the automatic focusing action it provides for the ion beam. The electrostatic focusing at the dee gap is entirely analogous to that in the high-voltage accelerating tubes. However, as the energy of the ions increases, this effect becomes almost negligible. Fortunately a magnetic focusing effect becomes more and more pronounced as the ions travel towards the periphery. This can be seen from the shape of the magnetic field as shown in figure IV–6. Near the edge of the pole faces the magnetic lines of force are curved, and, therefore, the field has a horizontal component which provides a

restoring force toward the median plane to an ion either below or above that plane. The focusing is so good that a cyclotron beam generally covers less than 1 cm^2 at the target.

One difficulty we have so far neglected is presented by the relativistic mass increase of the ions as they reach high energies. This increase is about ½ per cent for a 10-Mev deuteron and about 5 per cent for a 100-Mev deuteron. It is clear from equation IV–2 that if the revolution frequency is to be kept constant the increase in mass must be compensated by a proportional increase in field

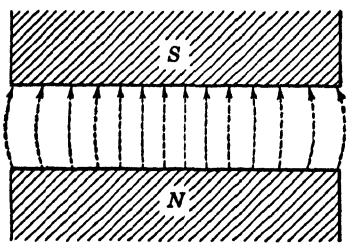


FIGURE IV–6. Shape of magnetic field in the gap of a cyclotron magnet. Curvature of lines of force near the edge of the magnet gives rise to focusing action.

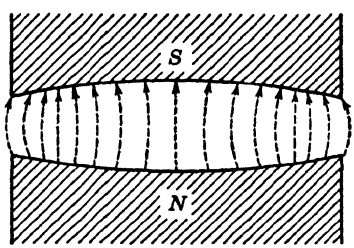


FIGURE IV–7. Exaggerated representation of defocusing effect of cyclotron magnet shimming.

strength. When the relativity effects are small this increase of the magnetic field toward the periphery can be readily achieved by slight radial shaping or shimming of the pole faces.[2] Notice, however, that this shaping of the field creates regions of magnetic defocusing (exaggerated in figure IV–7). For moderate relativistic mass increases this difficulty has been overcome, mainly by the use of higher dee voltages and correspondingly shorter ion paths. The 60-inch cyclotron in Berkeley operates excellently for 20-Mev deuterons or 40-Mev α particles, producing currents of about 100 μa and several microamperes, respectively. But it is clear that the energies available with the conventional cyclotron are limited by the relativity effects.

[2] In a given cyclotron the field should be shaped slightly differently for protons and for deuterons because of the different relativity effects. For this reason deuteron cyclotrons do not give very good proton beams without major readjustments.

FM Cyclotron. The relativity limitation can be overcome if the oscillator frequency can be modulated corresponding to the mass increase of the ions. The 184-inch cyclotron in Berkeley actually operates as a frequency-modulated cyclotron (sometimes also

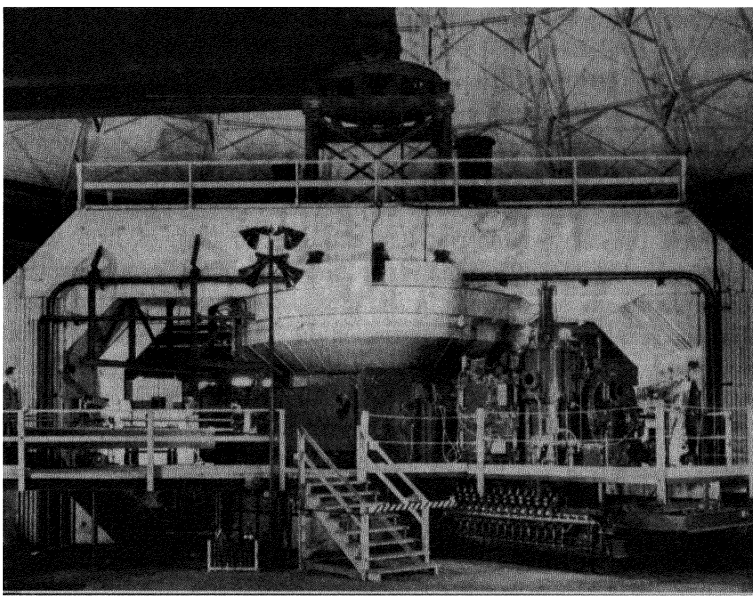


FIGURE IV-8. A general view of the giant (184-inch) Berkeley cyclotron, now producing 200-Mev deuterons and 400-Mev α particles operating as a synchrocyclotron, taken before the installation of the concrete shielding. In the right foreground, on the truck which moves on rails in the floor, are mounted the round vacuum housing for the rotating condenser, its associated vacuum pump, and behind these the oscillator housing. At the left can be seen the target probe, a shaft entering a port in the main vacuum chamber. (Photograph courtesy of Radiation Laboratory, University of California)

called synchrocyclotron). Many other large accelerators of this type are now under construction or design. In the FM cyclotron the decrease in rotation frequency due to the increasing mass is compensated by a decrease in the frequency of the r-f voltage applied to the dees; this frequency modulation may be brought about by means of a rotating condenser in the oscillator circuit. Obviously, for successful acceleration ions have to start their spiral path at or near the time of maximum frequency. Because

ions are accepted into stable orbits only during about 1 per cent of the FM cycle the ion currents are appreciably lower than in ordinary cyclotrons. On the other hand, the magnetic field can actually be shaped so as to increase the magnetic focusing effects. Frequency-modulated cyclotrons can be operated with relatively low dee voltages. The operating dee voltage for the 184-inch Berkeley cyclotron is 15 kv, which means that the deuterons make about 10^4 revolutions for a total path length of the order of 10^5 meters, and take about 1 millisec to reach the target.

Proton Synchrotron. Even with frequency modulation, cyclotrons probably become impractical for still higher energy ranges (above 1 Bev), mainly because of prohibitive magnet costs. (The 184-inch Berkeley cyclotron has about 4000 tons of iron in its magnet.) On the other hand, it appears that the synchrotron principle which is discussed in the next section can be extended to the acceleration of heavy particles in the billion-electron-volt range. A machine of this type for the acceleration of protons to 1.3 Bev is under construction in England, and work has begun on 3-Bev and 6-Bev proton synchrotrons at the Brookhaven National Laboratory and at the Berkeley Radiation Laboratory, respectively.

B. ELECTRONS

Very little work has been done with electron-induced nuclear reactions. There are several reasons for this: the cross sections for such reactions appear to be extremely low (10^{-5} barn and less), natural and artificially produced β emitters do not give out electrons of sufficiently high energy to be useful as electron sources for nuclear reactions, and the cyclotron cannot be used for the acceleration of electrons because of the high frequencies that would be required and because of the enormous relativistic mass increase of electrons even at moderate energies (at 1 Mev the total mass is three times the rest mass). The direct high-voltage machines discussed in section A can be applied to the acceleration of electrons as well as of positive ions, and especially the electrostatic generator has been extensively used for this purpose; e^-, e^- reactions and the Be^9 (e^-, ne^-) reaction have been observed this way. However, in recent years, methods for accelerating electrons to much higher energies have been developed, and these will be briefly described.

Betatron. The use of a varying magnetic field for the acceleration of electrons had been suggested by a number of investigators, but the first successful device using this idea was the induction accelerator or betatron developed by D. W. Kerst in 1940.

The betatron may be thought of as a transformer in which the secondary winding is replaced by a stream of electrons in a vacuum "doughnut." To understand the principle of operation consider the circular orbit described by an electron in a magnetic field. Suppose that the magnetic flux φ perpendicular to the plane of the orbit and inside it increases at the rate $d\varphi/dt$. As long as this increase in central flux continues, an induced emf will persist at the position of the electron orbit, steadily increasing the momentum of the electrons. In order for the electrons to continue to move in a fixed orbit it is necessary that the field at the orbit change proportionally with the momentum of the electrons. This can be seen from the condition of equality of centripetal magnetic force Hev and centrifugal force mv^2/r, which can be written $mv = Her$. Now to keep r constant, the rate of change of momentum $d(mv)/dt$ must be proportional to the rate of change of field strength dH/dt:

$$\frac{d(mv)}{dt} = er\frac{dH}{dt}. \tag{IV-3}$$

On the other hand, the rate of change of the momentum of an electron in an orbit of radius r due to a rate of change of flux $d\varphi/dt$ inside the orbit is given by

$$\frac{d(mv)}{dt} = \frac{e}{2\pi r}\frac{d\varphi}{dt}. \tag{IV-4}$$

Combining equations IV–3 and IV–4, we see that the condition

$$\frac{dH}{dt} = \frac{1}{2}\cdot\frac{1}{\pi r^2}\cdot\frac{d\varphi}{dt} \tag{IV-5}$$

must be fulfilled. This means that the field at the orbit must increase at exactly half the rate at which the average flux inside the orbit increases. This condition can be achieved by the proper tapering of the pole faces as indicated schematically in figure

IV–9. The radial variation of the field in the region of the electron orbit is important in the focusing problem. It turns out that, if the field falls off as the inverse nth power of the radius in the region of the orbit and if $\frac{1}{2} < n < 1$, the electrons will describe damped oscillations about the equilibrium orbit. Betatrons are generally designed with $n = \frac{3}{4}$, and the focusing is excellent, permitting the electrons to make hundreds of thousands of revolutions.

Electrons for acceleration are pulse-injected into the doughnut at 20 to 50 kev from an electron gun. The injection takes place

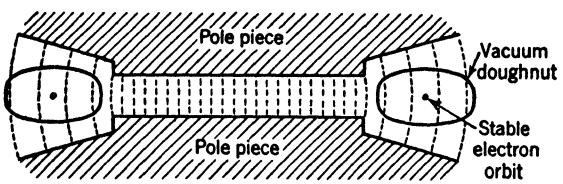


FIGURE IV–9. Cross section through central region of a betatron (schematic), with the magnetic lines of force indicated.

for 1 to 2 μsec when the field at the orbit is zero. The rapid variation of the magnetic field necessary for betatron operation is achieved with a magnet constructed of low-loss laminated iron and energized by an alternating current. The magnetizing coils are resonated with a capacitor bank. Since electrons are injected only once during each field cycle, the time average electron current obtainable is proportional to the frequency. Various frequencies between 60 and 1900 cycles have been used; the cooling of the magnet becomes more difficult with increasing frequency.

The energy obtainable with a given betatron is limited by the saturation of the iron in the pole pieces. As the central flux increases to the point where the iron in the pole pieces begins to saturate, the orbit begins to shrink. The electron beam can then be allowed to strike a target (usually a tungsten wire) mounted in the doughnut inside the equilibrium orbit. The maximum momentum and, therefore, the maximum energy attainable for a given orbit radius can be estimated by integration of equation IV–4, provided the maximum flux that can be reached without saturation is known. By orbit shift pulses from auxiliary magnet coils the orbit can be expanded, raised, lowered, or contracted toward a suitably placed target at any desired phase and, there-

fore, at any desired electron energy. Recently Kerst and his associates at the University of Illinois have succeeded in bringing the electron beam out of the vacuum doughnut into the air for more convenient experimentation.

A number of 20-Mev betatrons are in operation in the United States. Most of these are used as X-ray sources (see section C)

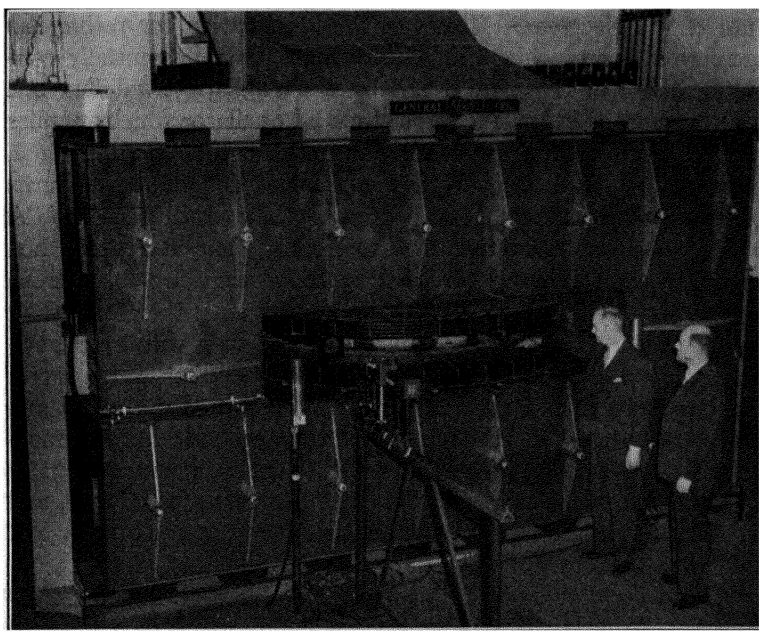


FIGURE IV–10. 100-Mev betatron, Research Laboratory, General Electric Company, Schenectady, N. Y. (The authors are indebted to Dr. E. E. Charlton who kindly supplied this photograph.)

for radiographic work. The largest betatron now in operation is the 100-Mev model at the General Electric Company in Schenectady which has a magnet weighing about 130 tons and an equilibrium orbit of 1 meter radius. A 250-Mev betatron is under design at the University of Illinois. It appears that for energies much in excess of this value the betatron becomes impractical, because with magnets of reasonably small size the energy loss by radiation as the electrons travel in circular paths becomes very serious for electrons of several hundred million electron volts.

Synchrotron. Another device for electron acceleration which has some advantages over the betatron, particularly from the point of view of size and cost, was proposed independently by V. I. Veksler and by E. M. McMillan and has come to be known as the synchrotron. In the synchrotron as in the betatron the radius of the electron orbit is kept approximately constant by a magnetic field which at the orbit increases proportionally with the momentum of the electrons. However, the acceleration (or rather the increase in energy since the velocity must remain essentially constant at $v \approx c$) is provided not by a changing central flux but (more nearly as in the cyclotron) by a r-f oscillator which supplies an energy increment every time the electrons cross a gap in a resonator which forms part of the vacuum doughnut. It turns out that with this arrangement phase stability is obtained at the orbit, because the electrons tend to cross the gap at a time when the field changes from accelerating to decelerating. Electrons arriving at the gap too early gain energy, and, therefore, their mass and period of revolution increase, and they will arrive relatively later at the next revolution. Those electrons which arrive too late have their mass and period of revolution decreased and are more nearly "on time" the next time around. As the magnetic field is steadily increased, a slight phase difference is maintained between the electron orbits and the resonator voltage, and on the average the electrons gain some energy at each passage of the gap.

With a constant radio frequency the stable orbit in a synchrotron has a constant radius only after the electrons have reached constant velocity. Therefore, if the electrons are injected from an electron gun at relatively low energies (say 50 kev, with velocity $= 0.41c$), the synchrotron orbit expands until the velocity of light is practically reached. However, this expansion of the orbit, which requires a large vacuum tube and magnet, can be avoided if the electrons are initially accelerated in the same machine by its operation as a betatron. The changeover from the betatron to the synchrotron principle in each cycle is achieved by saturation of small central flux bars in the magnet when the electrons reach an energy of about 2 Mev (velocity $= 0.98c$). At this time the r-f circuit is turned on, and thereafter the electrons receive their additional energy almost entirely from the synchrotron operation.

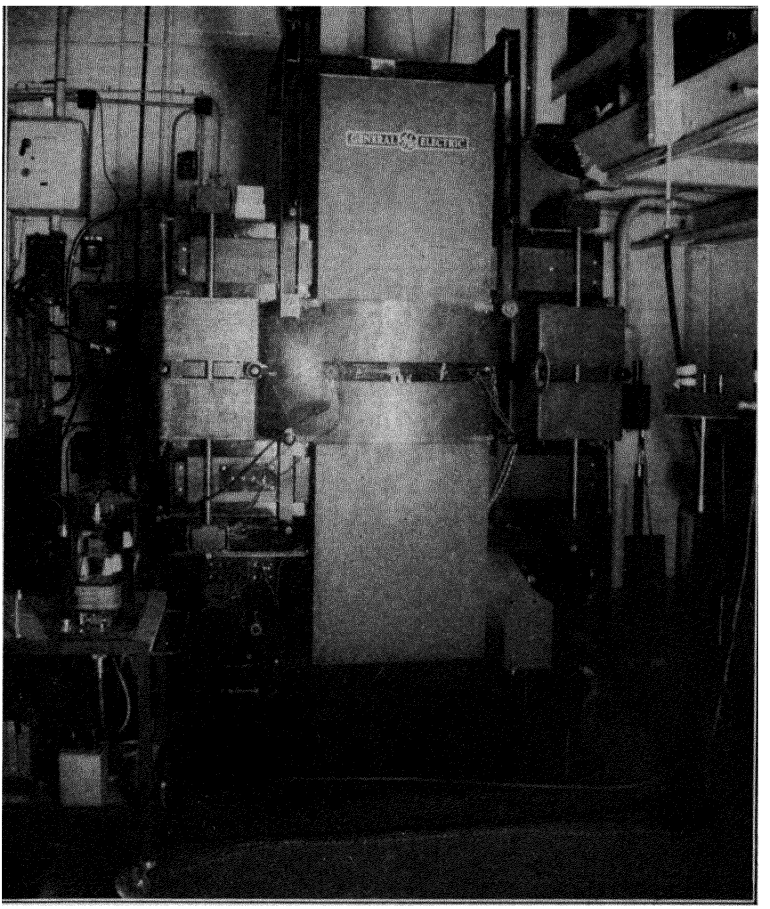


FIGURE IV–11. 70-Mev synchrotron, Research Laboratory, General Electric Company, Schenectady, N. Y. Notice the light spot near the left edge of the magnet gap. This light emerging tangentially from the doughnut is due to the energy loss by the accelerating electrons in their circular path. (The authors are indebted to Dr. E. E. Charlton who kindly supplied this photograph.)

In the synchrotron as in the betatron the rapidly varying magnetic field is provided by an a-c magnet, but because no large central flux is necessary the magnet size and cost are substantially lower. The maximum energy available for an orbit of given radius is determined by the peak value of the magnetic field at

the orbit (see equation IV-3). If radio frequency is turned off before the magnetic guide field has reached its peak, the electron beam spirals inward, and electrons of any desired energy below the maximum can thus be allowed to hit a target. If the r-f field is turned off after the magnetic field has passed its peak, the electrons will spiral out as the field decreases.

A number of synchrotrons designed for electron energies between 30 and 300 Mev are under construction. The largest unit in operation now is the 80-Mev synchrotron at the General Electric Company in Schenectady with an orbit radius of about 12 inches and a magnet weighing about 8 tons.

The synchrotron principle appears to be applicable to positive ions as well as to electrons. In this case, however, the velocity of light is not approached until very high energies are reached (at 200 Mev for a proton $v = 0.57c$). Therefore, in order to achieve a constant orbit radius a wide-band modulation of the radio frequency is proposed (much wider than in the FM cyclotron which uses spiraling orbits). This presents a serious but probably not insurmountable problem. An advantage of a synchrotron over an FM cyclotron is the fact that the synchrotron requires a magnetic field only at the circular orbit; thus a ring-shaped magnet can be used, which represents an enormous saving. Proton synchrotrons for the 1-to-10-Bev range are in various stages of design and construction.

Linear Accelerator. The principle of the linear accelerator is applicable to electrons as well as to positive ions, and a number of linear electron accelerators, using series of cavity resonators excited by microwave oscillators, are under construction. Some of these units are designed to accelerate electrons to energies of several hundred million electron volts.

C. GAMMA RAYS AND X RAYS

Radioactive Sources. Since nucleons are bound in most nuclei with binding energies of 6 to 8 Mev, photons having energies less than 6 Mev cannot be expected to induce many nuclear reactions. No γ rays known to be emitted in radioactive processes (from either natural or artificially produced radioactive substances) have energies that high; nor do the X rays from conventional X-ray

tubes. The only γ-ray- or X-ray-induced [3] nuclear reactions produced with such sources are, therefore, excitations of nuclei to isomeric levels and the "photodisintegrations" of the deuteron (threshold 2.18 Mev) and of Be^9 (threshold 1.63 Mev). Some of the radioactive γ-ray sources that have been used are listed in table IV–1.

TABLE IV–1

SOME TYPICAL RADIOACTIVE γ-RAY SOURCES

Source	Half-life	Energy (Mev)	Number of Quanta per Disintegration
ThC″	3.1m	2.62	1
Na^{24}	14.8h	2.76	1
		1.38	1
Y^{88}	105d	0.9	1
		1.87	1
Sb^{124}	60d	1.70	0.5
		0.60	1

Nuclear Reactions as Sources. In a number of nuclear reactions, especially with the lightest elements (which have large nuclear level spacings), very energetic γ rays are emitted. These in turn have been used to induce other nuclear reactions. Such sources are of particular value if the γ rays are monoenergetic. The most important sources of this type are listed in table IV–2. With these reactions relatively high intensities of γ rays (of the order of 10^6 quanta per sec) can be obtained from rather moderate high-voltage sets operating at 500 to 1000 kev.

TABLE IV–2

HIGH-ENERGY γ RAYS FROM NUCLEAR REACTIONS

Reaction	γ-Ray Energy (Mev)	Remarks
Li^7 (p, γ) Be^8	17.2	Resonance at 440 kev proton energy. Other components, at ~15 Mev and at lower energies, also found.
B^{11} (p, γ) C^{12}	11.8, 16.6	11.8 Mev has 7 times intensity of 16.6 Mev; also low-energy component (~ 4 Mev) found.

[3] For convenience we shall speak of γ-ray-induced reactions even when the electromagnetic radiation used is not of nuclear origin but is produced by the deceleration of electrons in a target in the manner of continuous X rays.

Bremsstrahlung. The continuous X rays produced when electrons are decelerated in the Coulomb fields of atomic nuclei are called bremsstrahlung (German for "slowing-down radiation"). This type of radiation is produced whenever fast electrons pass through matter, and the efficiency of the conversion of kinetic energy into bremsstrahlung goes up with increasing electron energy and with increasing atomic number of the material. In tungsten, for example, a 10-Mev electron loses about 50 per cent of its energy by radiation, whereas a 100-Mev electron loses over 90 per cent of its energy by that mechanism (see chapter VII, section B).

The spectrum of bremsstrahlung from a monoenergetic electron source extends from the electron energy down to zero, with approximately equal amounts of energy in equal energy intervals; in other words, the number of quanta in a narrow energy interval is about inversely proportional to the mean energy of the interval.

The stopping of fast electrons in matter thus produces a continuous spectrum of X rays, and any electron accelerator also serves as an X-ray source. Van de Graaff machines, betatrons, and synchrotrons have all been used as sources of X rays for producing nuclear reactions. In fact, unless special devices are used to bring the electron beam out of betatron or synchrotron doughnuts, the X rays are the only radiation available outside the vacuum systems of these machines. The higher the energy of an electron producing bremsstrahlung, the more the X-ray emission is concentrated in the forward direction; with the 100-Mev betatron, for example, about half the intensity of the X-ray beam is contained in a 2° cone. The chief disadvantage of bremsstrahlung sources for nuclear work is their spectral distribution. However, they are capable of producing electromagnetic radiation in energy and intensity ranges not accessible by other means.

D. NEUTRONS

Radioactive Sources. Our only sources of neutrons are nuclear reactions. There are several naturally occurring and several artificially produced α and γ emitters which can be combined with a suitable light element to make useful neutron sources. Because of the short ranges of the α particles, α emitters must be intimately mixed with the light element (usually beryllium, be-

cause it gives the highest yield). A γ emitter may be enclosed in a capsule surrounded by the light-element target. Some of the most commonly used sources are listed in table IV-3.

TABLE IV-3 †

Source	Reaction	Energy of Neutrons (Mev)	Approximate Yield (neutrons sec^{-1} rd^{-1})
Ra + Be (mixed)	Be^9 (α, n) C^{12}	Up to 13	460
Rn + Be (mixed)	Be^9 (α, n) C^{12}	Up to 11	400
Po + Be (mixed)	Be^9 (α, n) C^{12}	Up to ~11 (avg. ~4)	80
Ra + B (mixed)	B^{11} (α, n) N^{14}	Up to 8	180
Ra + Be (separated)	Be^9 (γ, n) Be^8	0.12, 0.51	0.8 *
Na^{24} + D_2O	H^2 (γ, n) H^1	0.27	7.8 *
Sb^{124} + Be	Be^9 (γ, n) Be^8	0.029	5.1 *
Na^{24} + Be	Be^9 (γ, n) Be^8	1.00	3.8 *
La^{140} + Be	Be^9 (γ, n) Be^8	0.75	0.062 *

* The photoneutron yields are given for 1 g of target (D_2O or Be) at 1 cm from the γ-ray source.

† Data for this table were obtained from:

H. L. ANDERSON, "Neutrons from Alpha Emitters," Preliminary Report No. 3 in Nuclear Science Series published by the National Research Council;

B. RUSSELL, D. SACHS, A. WATTENBERG, and R. FIELDS, "Yields of Neutrons from Photo-Neutron Sources," *Phys. Rev.* **73**, 545 (1948);

A. WATTENBERG, "Photo-Neutron Sources and the Energy of the Photo-Neutrons," *Phys. Rev.* **71**, 497 (1947).

Neutron-producing Reactions with Accelerators. Much more copious sources of neutrons than can be obtained with radioactive α and γ emitters are available with ion accelerators. The reaction $H^2(d, n)$ He^3 (often called a D, D reaction) is exoergic ($Q = +3.25$ Mev), and because the potential barrier is very low good neutron yields can be obtained with deuteron energies as low as 100 to 200 kev. With thick targets of solid D_2O, the yields are about 0.7, 3, and 80 neutrons per 10^7 deuterons at 100 kev, 200 kev, and 1 Mev deuteron energy, respectively. High-voltage sets and electrostatic generators are often used to produce the D, D reaction. The neutrons are monoenergetic if monoenergetic deuterons of moderate energies (up to a few million electron volts) fall on a sufficiently thin target.

For a controlled source of monoenergetic neutrons of very low energy (down to about 30 kev) the Li^7 (p, n) Be^7 reaction is suitable, especially when produced with the protons of well-defined energy available from electrostatic generators. The reaction is endoergic ($Q = -1.63$ Mev) and has a threshold of 1.86 Mev. Advantage may be taken of the differences in neutron energy in the forward and backward (and intermediate) directions.

With X rays from electrostatic generators, betatrons, and the like, neutrons can be produced by means of the Be^9 (γ, n) or H^2 (γ, n) reactions.[4] The yields of these reactions go up quite sharply with energy. With an electrostatic generator operating at 2.5 Mev with 100 μa electron current the neutron yield per gram of beryllium is equivalent to that from about 4 g of radium mixed with beryllium; at 3.2 Mev energy the corresponding figure is 26 g of radium.

Where high-energy deuterons are available, the most prolific neutron source is obtained by bombarding beryllium with deuterons. The reaction Be^9 (d, n) B^{10} has a positive Q value of about 4.3 Mev, but the neutrons are far from monoenergetic. When a beryllium target is bombarded with deuterons of E Mev energy, neutrons with a distribution of energies up to about $(E + 4)$ Mev are emitted. The neutron yield goes up rapidly with deuteron energy; it is about 10^8 neutrons per sec per μa for 1-Mev deuterons, about 10^{10} neutrons per sec per μa for 8-Mev deuterons, and about 3×10^{10} neutrons per sec per μa for 14-Mev deuterons.

With a given deuteron source a lithium target gives the highest neutron energies because the Li^7 (d, n) reaction is exoergic by 14.6 Mev. The neutron yield is only about one-third that for the Be^9 (d, n) B^{10} reaction. Neutrons are also obtained in the bombardment of practically any element with fast protons, deuterons, or α particles. The yields and energies vary from reaction to reaction, but if a neutron bombardment is needed for the activation of some substance it is often sufficient to place the sample near a cyclotron target which is being bombarded by deuterons, even if the target is not beryllium or lithium.

In the bombardment of targets with deuterons of very high energy (the 200-Mev deuterons of the 184-inch FM cyclotron in

[4] The product Be^8 is unstable and decomposes into two α particles. The threshold for the $Be^9(\gamma,n)Be^8$ reaction has been measured as 1.63 Mev, so that $Q = -1.63$ Mev.

Berkeley) it has been found [5] that high-energy neutrons are emitted in a rather narrow cone in the forward direction; the energy distribution of these neutrons is approximately Gaussian, with the maximum at half the deuteron energy. On striking the edge of a nucleus the high-energy deuteron may have its proton or neutron stripped off; thus the cone of neutrons and a similar cone of protons are produced. (See chapter III, section C.)

Neutrons from nuclear reactions initially are fast neutrons. The slowing down of neutrons and some properties of thermal neutrons have already been discussed in chapter III, section D.

Nuclear Chain Reactors (Piles). By far the most prolific sources of neutrons known are the nuclear chain reactors or piles. A nuclear reactor is an assembly of fissionable material (such as uranium, enriched U^{235}, Pu^{239}, or U^{233}) arranged in such a way that a self-sustaining chain reaction is maintained. In each fission process a number of neutrons (somewhere between one and three) are emitted. The requirement common to all reactors is that at least one of these neutrons must be available to produce another fission rather than escape from the assembly or be used up in some other type of nuclear reaction. Therefore, for a given type of reactor there is a minimum (or critical) size, below which the chain reaction cannot be self-sustaining. It is also necessary to avoid as much as possible the presence in the reactor of materials which consume neutrons in processes other than the fission reaction. This imposes a severe restriction on structural materials, coolants, and moderators.

If a reactor has exactly the critical size, the neutrons produced in each fission will, on the average, give rise to exactly one new fission, and the neutron density will remain constant. The ratio of the number of neutrons in one generation to that in the preceding generation is called the multiplication constant or reproduction factor k, and, for an exactly critical assembly, $k = 1$. In practice, reactors are designed in such a way that k can be made slightly larger than one. The reactor can then be made supercritical ($k > 1$), just critical ($k = 1$), or subcritical ($k < 1$) by means of so-called control rods. These are rods made of material with large neutron-absorption cross section (in the case of slow-neutron piles usually cadmium or boron steel) which can be intro-

[5] R. Serber, *Phys. Rev.* **72**, 1008 (1947). A. C. Helmholz, E. M. McMillan, and D. C. Sewell, *Phys. Rev.* **72**, 1003 (1947).

duced into the pile to accurately adjustable depths. The farther the control rods are introduced, the smaller becomes k. In order to increase the power level and the neutron density of a reactor it is necessary to allow k to become greater than 1. It is possible to do this without letting the reactor get out of control because a certain fraction (about 0.6 per cent) of the neutrons emitted in fission are not emitted instantaneously, but with half-lives ranging from about 0.4 sec to about 1 min (see chapter VI, section D). Therefore, as long as $k - 1$ is smaller than this fraction, the exponential increase in neutron density is relatively slow, and reactors can then be controlled very easily by means of the control rods. Since no elements with very high capture cross sections for fast neutrons are known, control rods seem hardly feasible for reactors operating on fast neutrons; in such reactors one might be able to control the reproduction factor by adjusting the position of some of the fissionable material itself or of a surrounding neutron reflector. In addition to the accurately adjustable operating controls, reactors are equipped with safety devices which allow rapid shutdown. These may consist of neutron-absorbing rods which can be rapidly introduced into the reactor and whose introduction insures a decrease in k to well below 1. Such safety devices may, of course, be triggered automatically, for example, when a certain neutron density is reached.

In most of the nuclear reactors now in operation thermal neutrons are used for the propagation of the chain reaction. Since the neutrons produced in fission are emitted with kinetic energies of the order of 1 Mev, such reactors contain so-called moderators, that is, materials whose function it is to slow down the neutrons to thermal energies. Moderators should, of course, be of low mass number and have low cross sections for neutron absorption. Graphite, heavy water, and ordinary water have been used as moderators. Moderators may be either homogeneously mixed with the fissionable material or separated from it in a heterogeneous arrangement. Actually all the reactors which have been constructed with ordinary uranium as the "fuel" are of the heterogeneous type. The reason is that the resonance absorption in U^{238} which accounts for much of the loss of neutrons [6] in thermal

[6] This is a "loss" in terms of the chain-reaction propagation; but the neutron capture in U^{238} is the process responsible for the production of plutonium in piles by the set of reactions: U^{238} (n, γ) U^{239}; $U^{239} \xrightarrow{23m} Np^{239} + \beta^-$; $Np^{239} \xrightarrow{2.3d} Pu^{239} + \beta^-$.

uranium reactors is reduced considerably when the uranium is arranged in aggregates such as lumps or rods. In a homogeneous mixture of uranium and moderator the probability that a neutron during the slowing-down process is absorbed by U^{238} in the resonance region is quite large. If, however, the uranium is arranged in aggregates, the probability for resonance capture will be large only in a relatively thin layer at the surface of the aggregate, whereas this layer effectively shields the interior from neutrons of the resonance energy. In most of the existing reactors uranium cylinders (canned in aluminum jackets for corrosion protection) are imbedded in a graphite lattice. Such reactors are now in operation at Oak Ridge, Tenn.; Hanford, Wash.; and Harwell, England; and one is under construction at Brookhaven National Laboratory. Heavy-water moderators are used in heterogeneous uranium reactors at Argonne National Laboratory, at Chalk River, Canada, and in a pile recently put into operation at Fort de Chatillon, France.

A homogeneous thermal reactor (called the "water boiler") using uranium enriched in U^{235} has been in operation at Los Alamos, N. Mex. The enriched uranium is in the form of a uranyl salt, and this is in solution in ordinary water which serves as the moderator. The solution is in a spherical container surrounded by a neutron reflector (in this case beryllium oxide). The critical size of a reactor may often be significantly reduced by a neutron reflector surrounding the reactor; to be a good reflector a substance must have a low capture cross section and a high scattering cross section for neutrons.

Although many schemes for reactors operating in the resonance (or epithermal) and in the high-energy regions have been discussed, only one such reactor seems to have been put into operation so far: the so-called "Fast Reactor" at Los Alamos. In this device plutonium metal is used as the nuclear fuel, and no moderator is present. A circulating liquid coolant is employed.

The total energy release in each fission process is about 200 Mev or 3.2×10^{-4} erg or 3.2×10^{-11} watt-sec. (Over 80 per cent of this energy appears as kinetic energy of the fission fragments; most of the remainder is released in subsequent radioactive processes.) Therefore, in a reactor operating at a power of 1 watt, about 3×10^{10} fissions take place every second. The power ratings of most of the operating piles are measured in kilowatts or even megawatts. For example, the air-cooled graphite–uranium

piles at Oak Ridge and Harwell, England (BEPO, the larger of the two reactors there), have been reported to operate at greater than 2000 kw and about 6000 kw, respectively. The power rating of the water-cooled Hanford piles (whose chief function is plutonium production) has not been released but is certainly much larger than that of the Oak Ridge pile. The first nuclear reactor built in 1942 in Chicago and later reconstructed at the Argonne Laboratory near Chicago consists of uranium and uranium oxide lumps in a graphite lattice with no cooling provisions and has been reported to operate at a few kilowatts. The water-cooled Los Alamos water boiler has operated at about 10 kw. The heavy-water reactor at Argonne (in which the heavy water also serves as a coolant) has been reported to run at a power level of 300 kw.

Since we are here considering nuclear reactors chiefly as neutron sources, not as plutonium production plants or as power sources, we are concerned more with available neutron fluxes than with power ratings. Although for a given reactor the two quantities are proportional to each other, the proportionality constant depends on the design details of the reactor. Neutron fluxes of about 10^{12} neutrons per cm^2 per sec have been reported both for the Oak Ridge graphite pile and the Los Alamos Fast Reactor. A maximum flux of about 5×10^{12} neutrons per cm^2 per sec is expected for the graphite–uranium pile under construction at Brookhaven National Laboratory. The neutron flux generally falls off from the center towards the outside of a reactor. In most reactors, especially in those designed primarily for research purposes (such as the Brookhaven and Harwell reactors), facilities are available for exposure of samples in holes or channels in the interior. The neutron energy distribution in these spots depends, of course, on the type of reactor; in graphite–uranium piles thermal neutrons predominate, but the flux of neutrons even up to several million electron volts energy is not entirely negligible. To provide pure thermal-neutron sources so-called thermal columns are often attached to reactors. A thermal column is a column of graphite (or some other moderator) of sufficient length to insure a thermal-energy distribution for the neutrons which have passed through it. The neutron flux at the end of a thermal column is, of course, several orders of magnitude smaller than that available inside the associated reactor.

EXERCISES

1. Show that the magnetic field near the edge of a cyclotron magnet provides a focusing action for positive ions (see figure IV–6).

2. See p. 100. Explain the difference between the Q value and the threshold of the Li^7 (p, n) Be^7 reaction.

3. Estimate (a) the percentage frequency modulation and (b) the pole diameter required for an FM cyclotron designed to accelerate protons to 350 Mev. Assume $H = 16{,}000$ gauss.

4. What would be the minimum n, γ cross section detectable by means of the product activity in a sample of 10 cm^2 area containing 1 mg-equivalent of target isotope, with a mixed Ra–Be source containing 1 g radium? Assume that the bombardment is continued to saturation and that 1 per cent of the neutrons emitted by the source strike each square centimeter of the target sample as slow neutrons. Consider 30 disintegrations per min as the minimum detectable activity.

5. Suppose you want to prepare some 5.3-year Co^{60} with a cyclotron and have the choice of bombarding a cobalt sample directly with 14-Mev deuterons for 2 hr or of surrounding it with paraffin and placing it near a beryllium target bombarded with 14-Mev deuterons for a total of 100 hr. Which is more advantageous from the point of view of total activity obtained? Use data in tables B and C in the appendix, and make reasonable assumptions about the solid angle subtended by the neutron-irradiated sample.

6. (a) What would be the approximate neutron energy from a beryllium target bombarded with As^{76} γ rays? (b) What would be the energy difference between neutrons in the forward and in the reverse directions?
Answer: (a) 107 kev; (b) 5.8 kev.

7. Estimate (a) the equilibrium quantity of Ba^{140} present in a uranium–graphite reactor operating at 1000 kw, and (b) the total amount of Ce^{140} accumulated in the same reactor after 1 year's operation followed by 2 months' shutdown. *Answer:* (a) 0.70 g.

8. What time would be required to convert 2 per cent of the Cd^{113} in a thin cadmium foil to Cd^{114} if the foil were placed in a reactor with a thermal neutron flux of 5×10^{11} neutrons per cm^2 per sec?

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V. QUANTITATIVE TREATMENT OF RADIOACTIVE PROCESSES

A. EXPONENTIAL DECAY

Half-life. We have seen (in chapter I) that a given radioactive species decays according to an exponential law: $N = N_0 e^{-\lambda t}$ or $\mathbf{A} = \mathbf{A}_0 e^{-\lambda t}$, where N and $\mathbf{A}$ represent the number of atoms and the measured activity, respectively, at time t, and N_0 and $\mathbf{A}_0$ the corresponding quantities when $t = 0$, and λ is the characteristic decay constant for the species. The half-life $t_{1/2}$ is the time interval required for N or $\mathbf{A}$ to fall from any particular value to one-half that value. The half-life is conveniently determined from a plot of log $\mathbf{A}$ vs. t when the necessary data are available and is related to the decay constant: $t_{1/2} = \dfrac{\ln 2}{\lambda} = \dfrac{0.69315}{\lambda}$.

Average Life. We may determine the average life expectancy of the atoms of a radioactive species. This average life is found from the sum of the times of existence of all the atoms divided by the initial number; if we consider N to be a very large number we may approximate this sum by an equivalent integral, finding for the average life τ:

$$\tau = -\frac{1}{N_0}\int_{t=0}^{t=\infty} t\,dN = \frac{1}{N_0}\int_0^\infty t\lambda N\,dt$$

$$= \lambda\int_0^\infty te^{-\lambda t}\,dt = -\left[\frac{\lambda t + 1}{\lambda}e^{-\lambda t}\right]_0^\infty = \frac{1}{\lambda}.$$

We see that the average life is greater than the half-life by the factor 1/0.693; the difference arises because of the weight given in the averaging process to the fraction of atoms that by chance survive for a long time. It may be seen that during the time $1/\lambda$ an activity will be reduced to just $1/e$ of its initial value.

Mixtures of Independently Decaying Activities. If two radioactive species, denoted by subscripts 1 and 2, are mixed together,

then the observed total activity is the sum of the two separate activities: $\mathbf{A} = \mathbf{A}_1 + \mathbf{A}_2 = c_1\lambda_1 N_1 + c_2\lambda_2 N_2$. The detection coefficients c_1 and c_2 are by no means necessarily the same and often

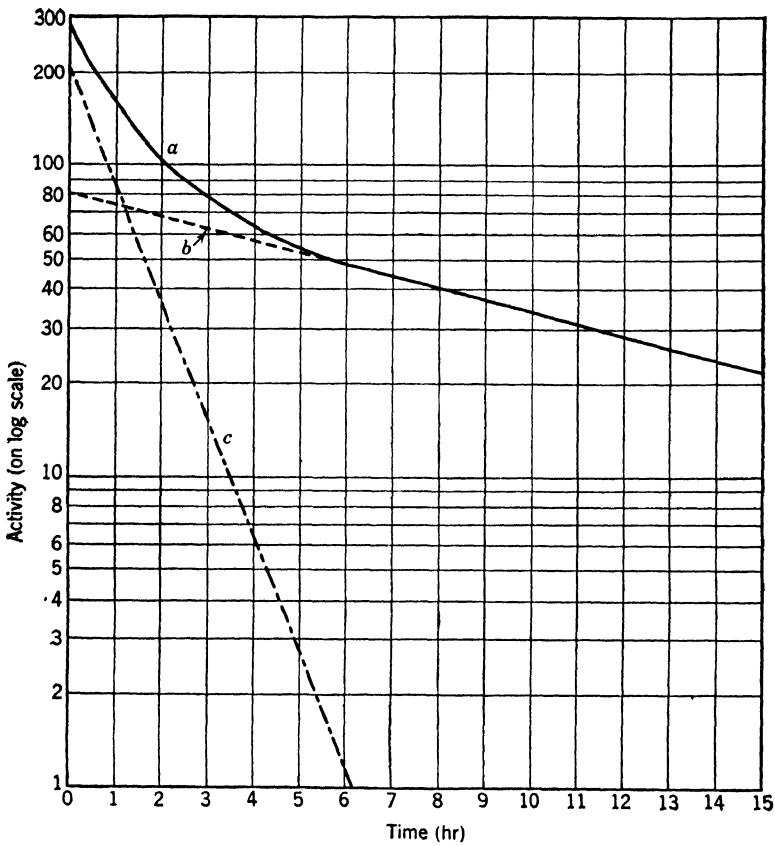


FIGURE V–1. Analysis of composite decay curve.
(*a*) Composite decay curve.
(*b*) Longer-lived component ($t_{1/2}$ = 8.0 hr).
(*c*) Shorter-lived component ($t_{1/2}$ = 0.8 hr).

are very different in magnitude. In general, $\mathbf{A} = \mathbf{A}_1 + \mathbf{A}_2 + \cdots \mathbf{A}_n$ for mixtures of n species.

For a mixture of several *independent* activities the result of plotting log **A** vs. t is always a curve concave upward (convex toward the origin). This curvature results because the shorter-lived components become relatively less significant as time passes.

In fact, after sufficient time the longest-lived activity will entirely predominate, and its half-life may be read from this late portion of the decay curve. Now, if this last portion, which is a straight line, is extrapolated back to $t = 0$ and the extrapolated line subtracted from the original curve, the residual curve represents the decay of all components except the longest-lived. This curve may be treated again in the same way, and in principle any complex decay curve may be analyzed into its components. In actual practice experimental uncertainties in the observed data may be expected to make it most difficult to handle systems of more than three components, and even two-component curves may not be satisfactorily resolved if the two half-lives differ by less than about a factor of two. The curve shown in figure V–1 is for two components with half-lives differing by a factor of 10.

B. GROWTH OF A RADIOACTIVE PRODUCT

General Equation. In chapter I we considered briefly a special case in which a radioactive daughter substance was formed in the decay of the parent. Let us take up the general case for the decay of a radioactive species, denoted by subscript 1, to produce another radioactive species, denoted by subscript 2. The behavior of N_1 is just as has been derived; that is, $-dN_1/dt = \lambda_1 N_1$, and $N_1 = N_1{}^0 e^{-\lambda_1 t}$, where we use the symbol $N_1{}^0$ to represent the value of N_1 at $t = 0$. Now the second species is formed at the rate at which the first decays, $\lambda_1 N_1$, and itself decays at the rate $\lambda_2 N_2$. Thus,

$$\frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2$$

or

$$\frac{dN_2}{dt} + \lambda_2 N_2 - \lambda_1 N_1{}^0 e^{-\lambda_1 t} = 0. \qquad \text{(V-1)}$$

For this linear differential equation of the first order we assume a solution of the form $N_2 = uv$, where u and v are functions of t. Differentiating, we obtain

$$\frac{dN_2}{dt} = u\frac{dv}{dt} + v\frac{du}{dt}.$$

Substituting in equation V–1, we have

$$u\frac{dv}{dt} + v\frac{du}{dt} + \lambda_2 uv - \lambda_1 N_1{}^0 e^{-\lambda_1 t} = 0,$$

which may be rearranged to give

$$u\left(\frac{dv}{dt} + \lambda_2 v\right) + v\frac{du}{dt} - \lambda_1 N_1{}^0 e^{-\lambda_1 t} = 0. \qquad \text{(V–2)}$$

We may choose the arbitrary function v so that the term in parenthesis is zero,

$$\frac{dv}{dt} + \lambda_2 v = 0,$$

$$\frac{dv}{dt} = -\lambda_2 v,$$

$$v = e^{-\lambda_2 t}.$$

By substitution of this result in equation V–2 a differential equation in u is obtained:

$$e^{-\lambda_2 t}\frac{du}{dt} - \lambda_1 N_1{}^0 e^{-\lambda_1 t} = 0,$$

$$du = \lambda_1 N_1{}^0 e^{(\lambda_2 - \lambda_1)t}\, dt,$$

$$u = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1{}^0 e^{(\lambda_2 - \lambda_1)t} + C;$$

and

$$N_2 = uv = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1{}^0 e^{-\lambda_1 t} + Ce^{-\lambda_2 t}. \qquad \text{(V–3)}$$

The constant C is evaluated from the condition $N_2 = N_2{}^0$ at $t = 0$:

$$C = N_2{}^0 - \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1{}^0.$$

Substituting in equation V–3 and rearranging we obtain the final solution for N_2 as a function of time:

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1{}^0 (e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_2{}^0 e^{-\lambda_2 t}. \qquad \text{(V–4)}$$

Notice that the first group of terms shows the growth of daughter from the parent and the decay of these daughter atoms; the last term gives the contribution at any time from the daughter atoms present initially.

Transient Equilibrium. In applying equation V-4 to considerations of radioactive (parent and daughter) pairs one can distinguish two general cases, depending on which of the two sub-

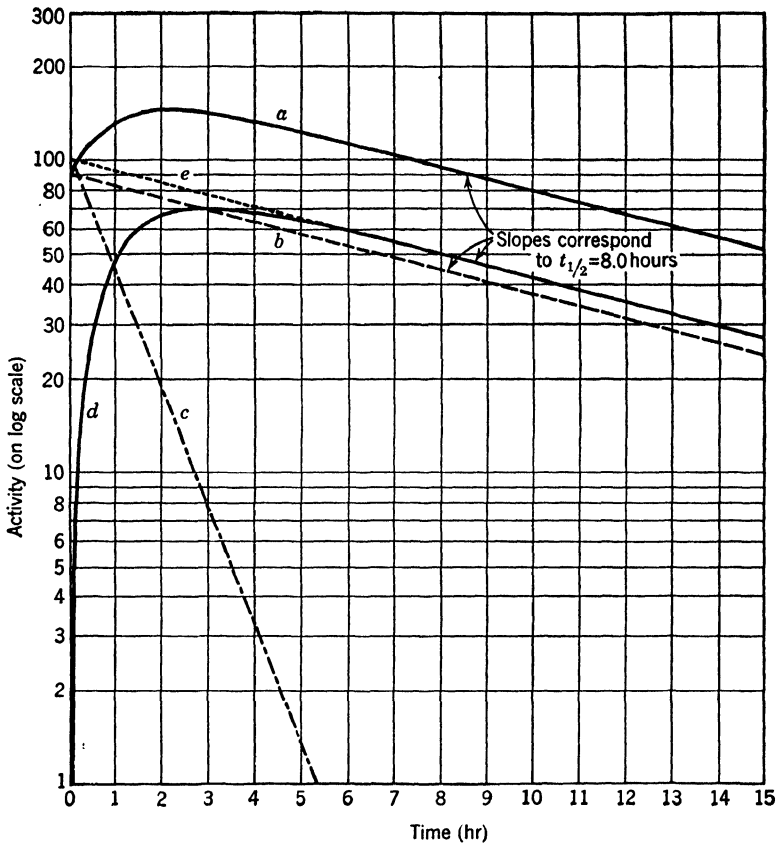


FIGURE V–2. Transient equilibrium.

(*a*) Total activity of an initially pure parent fraction.
(*b*) Activity due to parent ($t_{1/2}$ = 8.0 hr).
(*c*) Decay of freshly isolated daughter fraction ($t_{1/2}$ = 0.80 hr).
(*d*) Daughter activity growing in freshly purified parent fraction.
(*e*) Total daughter activity in parent-plus-daughter fractions.

stances has the longer half-life. If the parent is longer-lived than the daughter ($\lambda_1 < \lambda_2$) a state of so-called radioactive equilibrium is reached; that is, after a certain time the ratio of the numbers of atoms and, consequently, the ratio of the disintegration rates of parent and daughter become constant. This can be readily seen from equation V–4; after t becomes sufficiently large $e^{-\lambda_2 t}$ is negligible compared with $e^{-\lambda_1 t}$, and $N_2{}^0 e^{-\lambda_2 t}$ also becomes negligible; then

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_1{}^0 e^{-\lambda_1 t},$$

and, since $N_1 = N_1{}^0 e^{-\lambda_1 t}$,

$$\frac{N_1}{N_2} = \frac{\lambda_2 - \lambda_1}{\lambda_1}. \tag{V–5}$$

The relation of the two measured activities is found, from $\mathbf{A}_1 = c_1 \lambda_1 N_1$, $\mathbf{A}_2 = c_2 \lambda_2 N_2$, to be

$$\frac{c_2 \mathbf{A}_1}{c_1 \mathbf{A}_2} = \frac{\lambda_2 - \lambda_1}{\lambda_2}. \tag{V–6}$$

Notice that the right-hand sides of equations V–5 and V–6 are not the same. In the special case of equal detection coefficients ($c_1 = c_2$) the ratio of the two activities, $\mathbf{A}_1/\mathbf{A}_2 = 1 - (\lambda_1/\lambda_2)$, may have any value between 0 and 1, depending on the ratio of λ_1 to λ_2; that is, in equilibrium the daughter activity will be greater than that of the parent by the factor $\frac{\lambda_2}{\lambda_2 - \lambda_1}$. In equilibrium both activities decay with the parent's half-life.

Secular Equilibrium. A limiting case of radioactive equilibrium in which $\lambda_1 \ll \lambda_2$ and in which the parent activity does not decrease measurably during many daughter half-lives is known as secular equilibrium. We illustrated this situation in chapter I and now may derive the equation presented there, as a useful approximation of equation V–5:

$$\frac{N_1}{N_2} = \frac{\lambda_2}{\lambda_1}, \quad \text{or} \quad \lambda_1 N_1 = \lambda_2 N_2.$$

In the same way equation V–6 reduces to

$$\frac{c_2 A_1}{c_1 A_2} = 1, \quad \text{or} \quad \frac{A_1}{A_2} = \frac{c_1}{c_2},$$

and the measured activities are equal if $c_1 = c_2$.

The production of a radioactive substance (daughter) by any steady source, for example, steadily operating nuclear chain

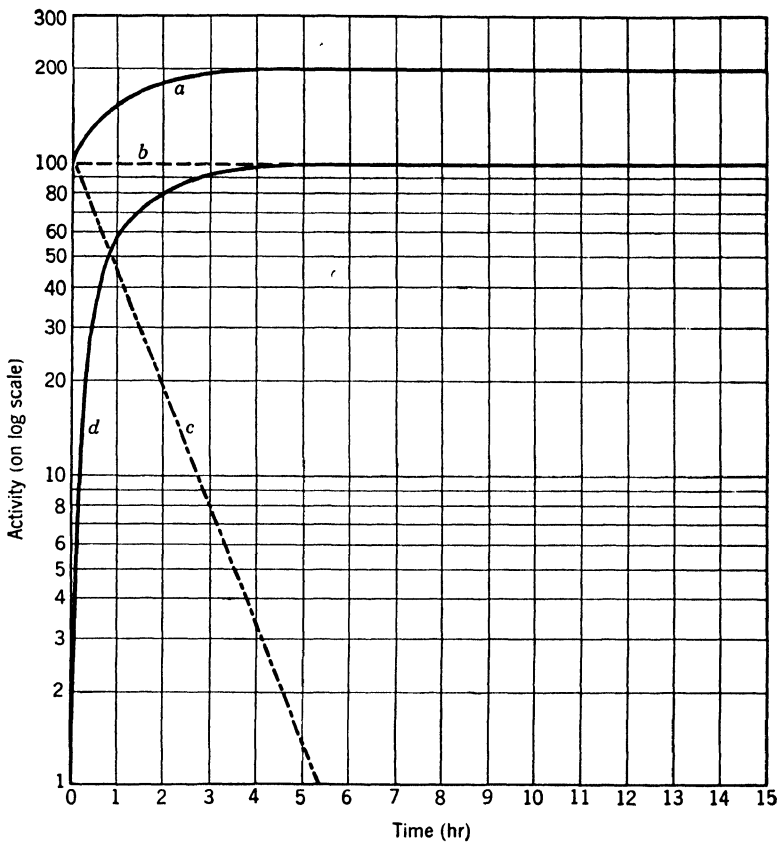


FIGURE V–3. Secular equilibrium.

(*a*) Total activity of an initially pure parent fraction.
(*b*) Activity due to parent ($t_{1/2} = \infty$); this is also the total daughter activity in parent-plus-daughter fractions.
(*c*) Decay of freshly isolated daughter fraction ($t_{1/2} = 0.80$ hr).
(*d*) Daughter activity growing in freshly purified parent fraction.

reactors (piles) or cyclotrons, presents a situation analogous to the approach to secular equilibrium. We may obtain the growth formula for N_2 as a function of time from equation V–4, setting $N_2^0 = 0$ at $t = 0$, using $\lambda_1 \ll \lambda_2$ and $e^{-\lambda_1 t} = 1$, and replacing $\lambda_1 N_1^0$ by the rate **R** of production of the active atoms:

$$N_2 = \frac{\mathbf{R}}{\lambda_2}(1 - e^{-\lambda_2 t}). \qquad \text{(V–7)}$$

As t becomes long compared to the half-life of the activity then N_2 approaches $\mathbf{R}/\lambda_2$ as a maximum limiting value, and we may rewrite equation V–7 in this way:

$$N_2 = (N_2)_{\max}(1 - e^{-\lambda_2 t}). \qquad \text{(V–8)}$$

Thus if an activity with a 20-min half-life (say Ga^{70}) is being steadily produced, one-half the maximum attainable yield is reached after 20 min, three-fourths after 40 min, seven-eighths after 60 min, fifteen-sixteenths after 80 min, and so on. If you must pay by the hour for cyclotron running time, you would want to irradiate for not more than about two half-lives of the desired product.

Figure V–2 presents an example of the transient equilibrium with $\lambda_1 < \lambda_2$ (actually with $\lambda_1/\lambda_2 = 1/10$); the curves represent variations with time of the parent activity and the activity of a freshly isolated daughter fraction, the growth of daughter activity in a freshly purified parent fraction, and other relations; in preparing the figure we have taken $c_1 = c_2$. Figure V–3 is a similar plot for secular equilibrium; it is apparent that as λ_1 becomes smaller compared to λ_2 the curves for transient equilibrium shift to approach more and more closely the limiting case shown in figure V–3.

The Case of no Equilibrium. If the parent is shorter-lived than the daughter ($\lambda_1 > \lambda_2$), it is evident that no equilibrium is attained at any time. If the parent is made initially free of the daughter, then as the parent decays the amount of daughter will rise, pass through a maximum, and eventually decay with the characteristic half-life of the daughter. This is illustrated in figure V–4; for this plot we have taken $\lambda_1/\lambda_2 = 10$, and $c_1 = c_2$. In the figure the final exponential decay of the daughter is extrapolated back to

$t = 0$; this method of analysis is useful if $\lambda_1 \gg \lambda_2$ for then this intercept measures the activity $c_2\lambda_2 N_1{}^0$, the $N_1{}^0$ atoms giving rise to N_2 atoms early enough to take $N_1{}^0$ equal to the extrapolated value of N_2 at $t = 0$. The ratio of the initial activity, $c_1\lambda_1 N_1{}^0$, to this extrapolated activity gives the ratios of the half-lives if the relation between c_1 and c_2 is known:

$$\frac{c_1\lambda_1 N_1{}^0}{c_2\lambda_2 N_1{}^0} = \frac{c_1}{c_2} \cdot \frac{\lambda_1}{\lambda_2} = \frac{c_1}{c_2} \cdot \frac{(t_{1/2})_2}{(t_{1/2})_1}.$$

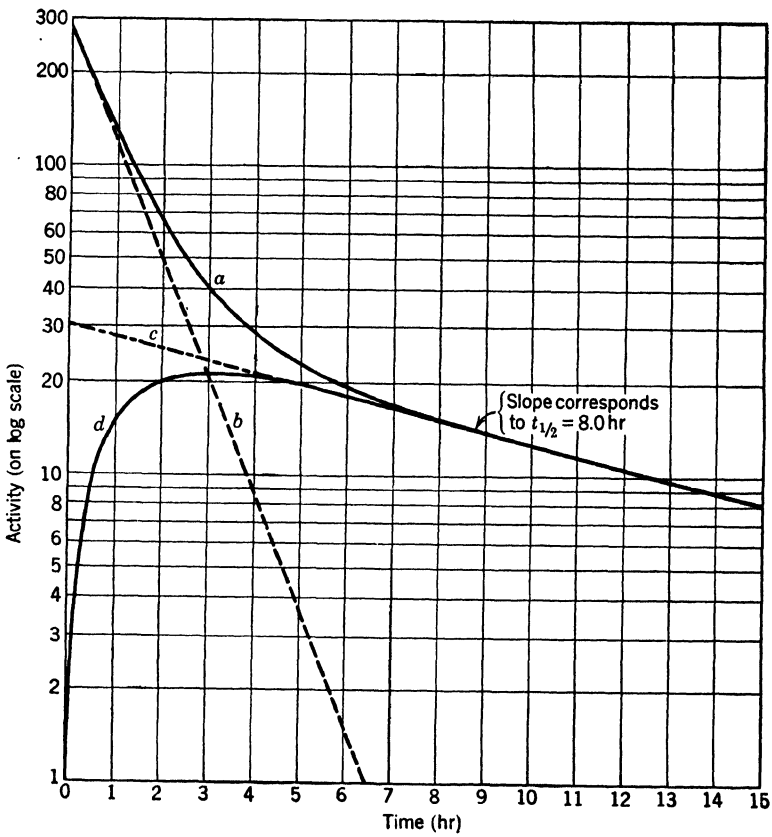


FIGURE V–4. The case of no equilibrium.

(*a*) Total activity.
(*b*) Activity due to parent ($t_{1/2}$ = 0.80 hr).
(*c*) Extrapolation of final decay curve to time zero.
(*d*) Daughter activity in initially pure parent.

If λ_2 is not negligible compared to λ_1, it can be shown that the ratio λ_1/λ_2 in this equation should be replaced by $\frac{\lambda_1 - \lambda_2}{\lambda_2}$, and the expression involving the half-lives changed accordingly.

Both the transient-equilibrium and the no-equilibrium cases are sometimes analyzed in terms of the time t_m for the daughter to reach its maximum activity when growing in a freshly separated parent fraction. This time we find from the general equation V–4 by differentiating,

$$\frac{dN_2}{dt} = -\frac{{\lambda_1}^2}{\lambda_2 - \lambda_1} {N_1}^0 e^{-\lambda_1 t} + \frac{\lambda_1 \lambda_2}{\lambda_2 - \lambda_1} {N_1}^0 e^{-\lambda_2 t},$$

and setting $dN_2/dt = 0$ when $t = t_m$:

$$\frac{\lambda_2}{\lambda_1} = e^{(\lambda_2 - \lambda_1) t_m}, \quad \text{or} \quad t_m = \frac{2.303}{\lambda_2 - \lambda_1} \log \frac{\lambda_2}{\lambda_1}.$$

At this time the daughter decay rate, $\lambda_2 N_2$, is just equal to the rate of formation, $\lambda_1 N_1$ (this is obvious from equation V–1); and in figures V–2, 3, 4, where we assumed $c_1 = c_2$, we have the parent activity $\mathbf{A}_1$ intersecting the daughter growth curve $\mathbf{A}_2$ at the time t_m. (The time t_m is infinite for secular equilibrium.)

C. MORE COMPLICATED CASES

Bateman Solution. If we consider a chain of three or more radioactive products it is clear that the equations already derived for N_1 and N_2 as functions of time are valid, and N_3 may be found by solving the new differential equation:

$$\frac{dN_3}{dt} = \lambda_2 N_2 - \lambda_3 N_3. \qquad \text{(V–9)}$$

This is entirely analogous to the equation for dN_2/dt, but the solution calls for more labor since N_2 is a much more complicated function than N_1. The next solution, for N_4, is still more tedious. H. Bateman has given the solution for a chain of n members with the special assumption that at $t = 0$ only the parent substance is

present, that is, that $N_2^0 = N_3^0 = \cdots N_n^0 = 0$. This solution is

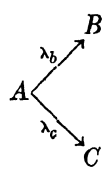
$$N_n = C_1 e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t} + \cdots C_n e^{-\lambda_n t},$$

$$C_1 = \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_2 - \lambda_1)(\lambda_3 - \lambda_1) \cdots (\lambda_n - \lambda_1)} N_1^0,$$

$$C_2 = \frac{\lambda_1 \lambda_2 \cdots \lambda_{n-1}}{(\lambda_1 - \lambda_2)(\lambda_3 - \lambda_2) \cdots (\lambda_n - \lambda_2)} N_1^0, \quad \text{etc.}$$

If we do require a solution to the more general case with N_2^0, $N_3^0 \cdots N_n^0 \neq 0$, we may construct it by adding to the Bateman solution for N_n in an n-membered chain, a Bateman solution for N_n in an $(n-1)$-membered chain with substance 2 as the parent, and, therefore, $N_2 = N_2^0$ at $t = 0$, and a Bateman solution for N_n in an $(n-2)$-membered chain, etc.

Branching Decay. Another variant that is met in general decay schemes is the branching decay, illustrated by



Here both λ_b and λ_c must be considered when the general relations in either branch are studied because, for example, the substance B is formed at the rate $\lambda_b N_A$, but A is consumed at the rate $(\lambda_b + \lambda_c)N_A$. Notice that A can have but one half-life, given in this case by $t_{1/2} = \dfrac{0.693}{\lambda_b + \lambda_c}$. By definition the half-life is related to the total rate of disappearance of a substance regardless of the mechanism by which it disappears.

D. UNITS OF RADIOACTIVITY

A familiar unit of radioactivity is the curie. Originally the term referred to the quantity of radon in equilibrium with one gram of radium. Later it came to be used as a unit of disintegration rate for any radioactive preparation, defined as that quantity of the preparation which undergoes the same number of disintegrations per second as one gram of pure radium. This use for

substances other than members of the radium series has never received the sanction of the appropriate international committees. With this definition the value of the curie should vary with successive refinements in the measurement of the decay constant or atomic weight of radium; the International Radium Standard Commission has recommended the use of the fixed value 3.7×10^{10} disintegrations per sec. The millicurie and the microcurie are practical units also in common use.

A new absolute unit of radioactive disintegration rate has recently been recommended by the National Bureau of Standards. This unit, the rutherford (abbreviated rd), is defined as that amount of a radioactive substance which undergoes 10^6 disintegrations per sec. The abbreviations mrd for millirutherford (10^3 dis per sec) and μrd for microrutherford (1 dis per sec) should be noted. The kilorutherford and the megarutherford are likely to be useful for very active materials.

As an illustration we may calculate the weight in grams W of 1 rd of C^{14} from its half-life of very nearly 5000 years:

$$\lambda = \frac{0.693}{5000}\,\text{year}^{-1} = \frac{0.693}{5000 \times 365 \times 24 \times 60 \times 60}\,\text{sec}^{-1}$$

$$= 4.4 \times 10^{-12}\,\text{sec}^{-1}$$

$$-\frac{dN}{dt} = \lambda N = \lambda \frac{W}{14} \times 6.02 \times 10^{23}$$

$$= 1.9W \times 10^{11}\ \text{disintegrations per sec.}$$

With $-\dfrac{dN}{dt} = 10^6$ disintegrations per sec (1 rd),

$$W = \frac{10^6}{1.9 \times 10^{11}} = 5.3 \times 10^{-6}\ \text{g.}$$

The lack of rigor in previous usage of units of radioactivity is no doubt largely a result of the experimental difficulty associated with determinations of absolute disintegration rates. These difficulties remain, and are discussed with the experimental techniques in chapter X. For cases in which the radioactivity is observed as a γ radiation the problem is particularly awkward. A common practice in the absence of information on the disintegration rate has been the comparison of the γ-radiation intensity with the γ intensity from a unit amount (curie) of radium in equilibrium

with its decay products, with some absorber interposed between sample and detector in each measurement. This procedure is uncertain and arbitrary for a number of obvious reasons. The Bureau of Standards proposes that γ-ray intensities be measured as such, without reference to absolute disintegration rates, and recommends as the unit one roentgen per hour at one meter (abbreviated rhm or r.h.m. and pronounced "rum"). The γ radiations from 1 curie of radium with decay products give a little less than 1 rhm (0.97 rhm bare, and 0.84 rhm through 0.5 mm of platinum absorber; the latter is standard practice). The roentgen as a unit of radiation intensity is defined in chapter VII, section E.

E. DETERMINATION OF HALF-LIVES

From Decay Curves. Half-lives in the range from several seconds to several years are usually determined experimentally by measuring the activity with an appropriate instrument at a number of suitable successive times. Then log **A** is plotted versus time, and the half-life found by inspection, provided that the activity is sufficiently free of other radioactivities that a straight line (exponential decay) is found, preferably extending over several half-life intervals. As we have already discussed, the decay curve resulting from a mixture of independent activities may often be analyzed to yield the half-lives of the various components. When difficulties arise in this analysis it is often adequate to measure separately decay curves through several different thicknesses of absorbing material to obtain curves with some components relatively suppressed. Our treatments of the more general equations have already suggested methods of finding half-lives from more complicated growth and decay curves.

The manipulations necessary for activity measurements become difficult as the time scale to be investigated becomes short. The use of electronic and photographic recording devices can extend the working region to half-lives well below 0.1 sec. With short-lived gaseous products, or products in solution, a method that has been particularly useful for fission products with half-lives of the order of a few seconds is to measure the activity at different points along a tube through which the fluid flows at a measured rate. The ordinary decay curve is then found on a plot of log **A** vs. distance along the tube. A method based on a similar principle

using a rapidly rotating wheel has been employed for solid samples; the half-life (0.022 sec) of B^{12} was determined in this way. In these procedures the limitation usually arises not in the activity measurements but in the rapid preparation, and possibly isolation, of the short-lived sample. The possible use of a modulated source, such as a betatron or synchrotron, or a cyclotron modified to produce periodic pulses of accelerated ions, may be mentioned. Appropriate electric circuits will divide the time between pulses into an arbitrary number of intervals and measure the average resulting activity in each interval.

From Variable-delay Coincidences. When a body of very short half-life results from a radioactive decay with moderate or long half-life the method of variable-delay coincidences can be used. In one form of apparatus the electric pulse produced in a detection instrument by a ray from the parent is electrically delayed by a time d and then recorded in coincidence with any ray from the daughter that may produce a pulse in a detector at that time, or more correctly at that time within the limits of the resolving time τ of the coincidence equipment. Now as d is varied by electrical means the coincidence counting rate—coincidences per unit τ—will vary; the effect is essentially to record disintegrations over the period τ at a time d after formation of the short-lived nucleus. The very short half-life is determined from the typical decay curve with the logarithm of the coincidence rate plotted against d. The Ta^{181} isomer of 22 μsec half-life was found in this way in the β decay of Hf^{181}. In an earlier form of apparatus no provision was made for the introduction of a delay time d, and coincidences C were recorded as τ was varied. Clearly, with τ very short (compared to the half-life sought) no coincidences would result; with τ long (compared to $t_{1/2}$) a maximum number of coincidences C_{max} would be observed. The expected relation, $C = C_{max}\,(1 - e^{-\lambda\tau})$, is analogous to the formula for radioactive growth, equation V–8, and the half-life is obtained from the semilog plot of $(C_{max} - C)$ vs. τ. The half-lives of the C' bodies in the three natural decay series were measured by this method.

From Specific Radioactivity. If the half-life, or disintegration constant, is to be determined for a substance of very long half-life (very small λ), the activity $\mathbf{A} = c\lambda N$ may not change measurably in the time available for observation. In such cases λ may be found from the relation $\lambda N = -dN/dt = \mathbf{A}/c$, provided that

N is known and $-dN/dt$ may be determined in an absolute way (through knowledge of the detection coefficient c). This method is most accurate for α emitters, and the absolute rates of emission of α particles from uranium samples have been investigated with great care to measure the half-life of U_I. In an accurate determination of the half-life of Pu^{239} the value of $-dN/dt$ was established in a calorimetric measurement of the heating effect, with the α-particle energy known from the α-particle range.

In some instances the disintegration rate is better obtained from a measurement of the equal disintegration rate of a daughter in secular equilibrium. Early determinations of the half-life of U^{235} were based on the α-particle counting rate of Pa^{231} obtained in known yield from old uranium ores; the U^{235} α particles were not measurable in a direct way because of the much larger number of α disintegrations occurring in the U^{238} and U^{234}.

From Geiger–Nuttall Rule or Sargent Relation. The half-lives for α emitters in the radioactive families may be estimated from measurements of the range R (or the energy) of the emitted α particles from an empirical relationship known as the Geiger–Nuttall rule: $\log R = A \log \lambda + B$, where A is a general constant, and B is a constant characteristic of the radioactive series. There may be large uncertainties in half-lives determined by this formula, as may be seen in exercise 5 at the end of this chapter. There exists an even less precise relation (the Sargent relation) between the energy (or range) and disintegration constant for β emitters; very roughly, $\lambda = KE^5$, where K is very different for different classes of transitions (allowed, forbidden, doubly forbidden, etc.). This relation is considered in chapter VI, but we may note here that it is sometimes useful in revealing discrepancies in one direction—half-lives too short for the measured energies in β decay.

EXERCISES

1. The following experimental data were obtained when the activity of a certain beta-active sample was measured at frequent intervals:

Time (in hours)	Activity (in counts/min)	Time (in hours)	Activity (in counts/min)
0	7300	4.0	481
0.5	4680	5.0	371
1.0	2982	6.0	317
1.5	1958	7.0	280
2.0	1341	8.0	254
2.5	965	10.0	214
3.0	729	12.0	181
3.5	580	14.0	153

Plot the decay curve on semilog paper and analyze it into its components. What are the half-lives and the initial activities of the component activities?

2. Calculate the weight of (*a*) 1 curie of radon; (*b*) 1 "curie" of P^{32} (see table A in the appendix for the half-life); (*c*) 1 rd of P^{32}; (*d*) 10,000 rd of H^3. *Answer:* (*a*) 6.47 μg; (*c*) 9.43×10^{-11} g.

3. What was the rate of production, in atoms per second, of I^{128} during a 1-hr cyclotron (neutron) irradiation of an iodine sample, if the sample is found to contain 3.5 rd of I^{128} activity at 15 min after the end of the irradiation?

4. From data from table A in the appendix, calculate the total rate of emission of α particles from 1 mg of ordinary uranium. Calculate this answer also for the case of 1 mg of very old uranium, in secular equilibrium with all its decay products. *Answer to first part:* 25.0 per sec.

5. Given the half-lives of Ra and RaA equal 1590 years and 3.05 min, and their α ranges equal 3.39 and 4.72 cm, respectively, estimate the half-lives of U^{238} and RaC′; the ranges for these α emitters are 2.63 and 6.96 cm. Compare the results with the half-lives given in table A of the appendix (obtained for RaC′ from coincidence counting with variable resolving time).

6. A 0.100 mg sample of pure $_{94}Pu^{239}$ (an α-particle emitter) was found to undergo 1.40×10^7 disintegrations per min. Calculate the half-life of this isotope. Pu^{239} is formed by the β decay of Np^{239}. How many rutherfords of Np^{239} would be required to produce a 0.100 mg sample of Pu^{239}?

7. A sample of 1.00×10^{-10} g of RaE is freshly purified at time $t = 0$. (*a*) If this sample is left without further treatment, when will the amount of Po^{210} in it be a maximum? (*b*) At that time of maximum growth, what will be the weight of Po^{210} present, the α activity in disintegrations per second, the beta activity in disintegrations per second, the number of microcuries of Po^{210} present? (*c*) Sketch on semilog paper a graph of α activity and β activity versus time.

8. In the slow-neutron activation of a sample of separated Mo^{100} isotope some 14.6-min Mo^{101} is produced; this decays to 14.0-min Tc^{101}. A sample of Mo^{101} is chemically freed of technetium and then immediately placed under a counter. Sketch the activity as a function of time, assuming the detection coefficient to be the same for the Tc^{101} as for the Mo^{101} radiation.

9. Carry out the solution of the differential equation V–9. Compare your result with the Bateman solution for this case with N_2^0 and N_3^0 not equal to 0.

10. A sample of an activity whose half-life is known to be 30 min was measured from 10:03 to 10:13. The total number of counts recorded in this 10-min interval was 34,650. What was the activity of the sample (in counts per minute) at 10:00? *Answer:* 4159.

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I. TYPES OF RADIOACTIVE DECAY

A. ALPHA DECAY

As we have seen in chapter I, α particles soon after their discovery were identified as doubly charged helium ions, first by the measurement of their charge-to-mass ratio and then by the spectroscopic evidence for the accumulation of helium in a tube surrounding an α emitter. Alpha-particle emission is therefore always accompanied by a decrease of two in atomic number and a decrease of four in mass number.

The α particles from a given isotope either all have the same energy or are distributed among a few monoenergetic groups. Where a single α-particle energy occurs, for example, in the decays of AcA (Po^{215}), Rn^{222}, and probably U^{238}, the transition evidently takes place between a single energy level of the α-emitting nucleus and a single energy level (generally the ground state) of the product nucleus. The emission of α particles of several different energies by one nuclear species may be due to the existence of several energy levels either preceding or following the α emission (or both, although this has not been observed).

Alpha-particle Groups. In the majority of cases, for example, in Ra^{226}, AcX (Ra^{223}), ThC (Bi^{212}), the emission of several α-particle groups of different energies from a given substance is due to the fact that the product nucleus can be left in different states of excitation which subsequently transform to the ground state by γ emission. Each γ-ray energy observed is then as a rule equal to the energy difference between the disintegration energies associated with two of the α-particle groups.(1) From a complete

[1] The total disintegration energy associated with an α-particle emission is larger than the α-particle energy by the recoil energy of the nucleus, which for the heavy α emitters is of the order of magnitude of 0.1 Mev. Since the momentum p and energy E of a particle of mass M are related by the equation $p^2 = 2ME$, it follows from the conservation of momentum that $M_\alpha E_\alpha = M_{\text{nucleus}} E_{\text{nucleus}}$. For example, in the case of the 6.083-Mev α particles of ThC the recoil energy is $6.083 \times 4/208 = 0.117$ Mev.

knowledge of the α and γ energies, an energy-level diagram of the product nucleus can often be constructed (see figure VI–1). Usually not all the possible γ transitions between the levels of

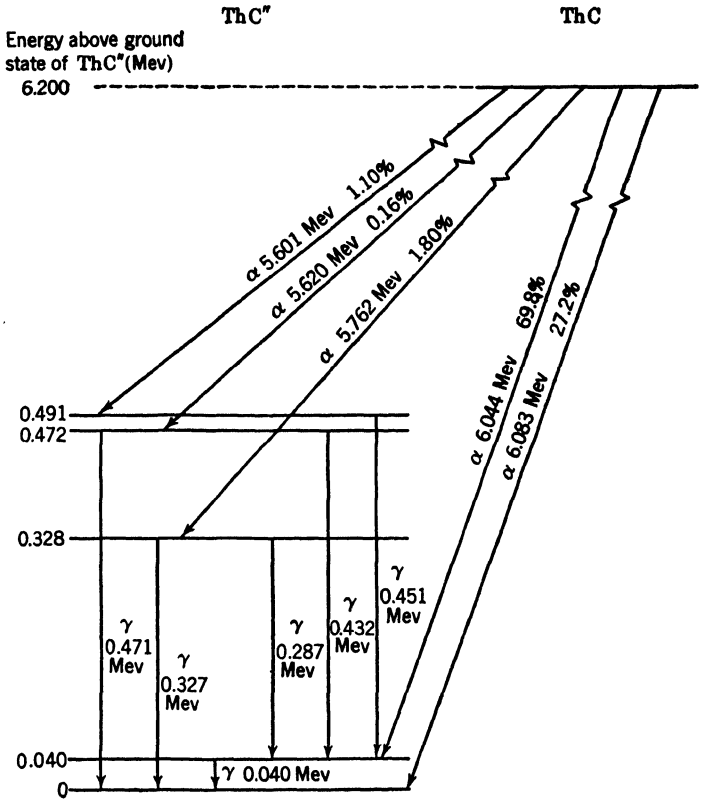


FIGURE VI–1. Energy-level diagram for the α-particle decay of ThC to ThC″. The α-particle energies given are the kinetic energies, not total disintegration energies.

the product nucleus are actually observed; in the ThC $\rightarrow$ ThC″ decay, for example, only six of the ten theoretically possible γ rays are found (figure VI–1). The reason is that in nuclear as in atomic or molecular transitions selection rules are operative (see section C).

In most of the cases where more than one α-particle energy is observed from a single α emitter, the groups of highest energy

also have the largest abundance. This can be understood from the fact that the probability for the penetration of the potential barrier increases with increasing energy. However, there are other factors (such as the angular momenta of the initial and final states) which also play a part in determining the relative probabilities for the emission of α particles of different energies.

Long-range α Particles. The disintegration of each of the short-lived α emitters RaC′ and ThC′ takes place predominantly with the emission of α particles of a single energy (7.683 Mev for RaC′ and 8.778 Mev for ThC′). However, in both cases small numbers of higher-energy α particles have been observed. Twelve higher-energy groups (with energies up to 10.51 Mev) are known in RaC′ and two (with energies up to 10.54 Mev) in ThC′; only 0.003 per cent of all the RaC′ α particles and 0.02 per cent of all the ThC′ α particles belong to these high-energy groups. The high-energy α particles are due to transitions from excited states of RaC′ and ThC′, as can be seen from a comparison with the different RaC and ThC β-particle transitions leading to these various states, The energy-level diagram for the ThC $\rightarrow$ ThC′ $\rightarrow$ ThD transitions is shown in figure VI-2.

Alpha emission from the excited levels in ThC′ and RaC′ must compete with γ transitions from these same levels to the respective ground states. The decay constants for α emission can be calculated approximately for the excited states from the Geiger–Nuttall rule which relates decay constant and α-particle energy (chapter V, section E). If the relative numbers of α particles and γ quanta from a given excited state are known experimentally, the decay constant of that excited state for γ emission can be estimated. This is actually the only experimental method of determining the "lifetimes" for the fast γ-ray transitions; it leads to values in the neighborhood of 10^{-13} sec. Gamma-ray transitions are discussed further in section C.

Penetration of Potential Barriers. The 10.54-Mev α particles of ThC′ are the most energetic α particles known from radioactive sources. At the other extreme are the 2.0-Mev α particles of samarium. Most of the naturally occurring α particles are emitted with energies between 4 and 8 Mev. Before the advent of quantum mechanics, the fact that α particles could be emitted with such low energies from nuclei which were known to have much higher potential barriers was very puzzling. For example, scattering

experiments with 8.78-Mev α particles (from ThC′) on uranium show that around a uranium nucleus no deviation from the Cou-

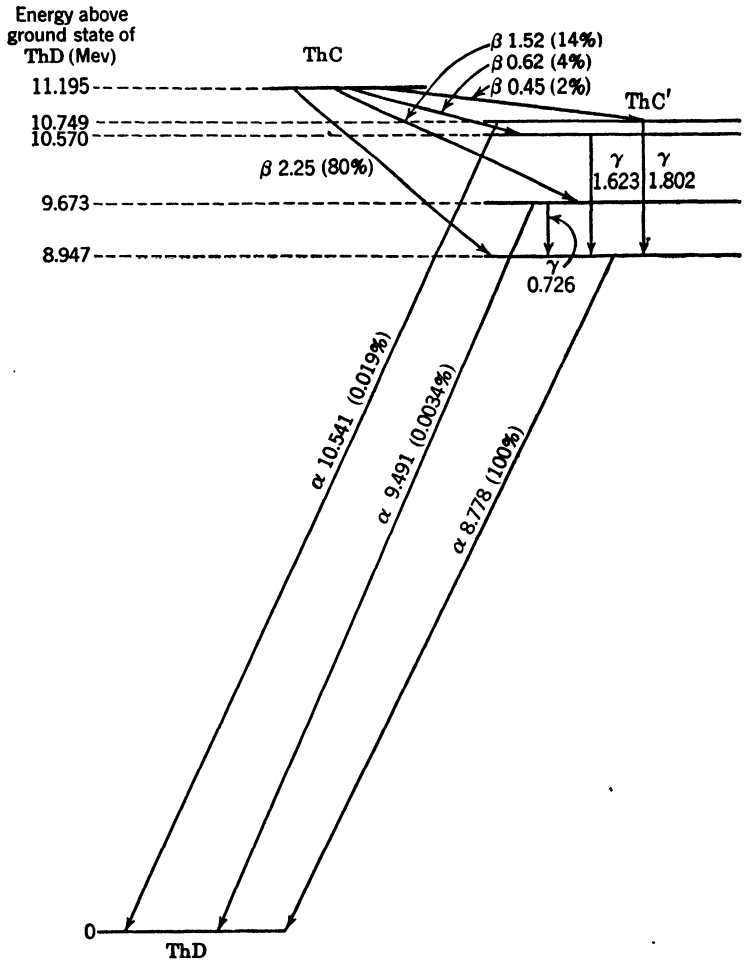


FIGURE VI–2. Energy-level diagram for the ThC–ThC′–ThD decays showing the origin of the long-range α particles of ThC′. The energies given do not include the rest energies of the α and β particles.

lomb law exists at least up to 8.78 Mev; yet U^{238} emits 4.5-Mev α particles. How do these α particles get outside the potential barrier which is at least 8.78 Mev high (and actually quite a bit higher)?

increases again, and, finally, the gap breaks down into a glowing discharge or arc, with a very sharp rise in the current. In the measurement of gas ionization it is obviously of some advantage to measure the saturation current: the current is easily interpreted in terms of the rate of gas ionization, and the measured current does not depend critically on the applied voltage or other like factors. The range of voltage over which the saturation current is obtained depends on the geometry of the electrodes and their spacing, the nature and pressure of the gas, and the general and local density and spatial distribution of the ionization produced in the gas. In air, for many practical cases, this region may be taken to extend from $\sim 10^2$ to $\sim 10^4$ volts per cm of distance between the electrodes.

We may classify detection systems (of the ion-collection type) according to whether saturation collection is employed or whether the multiplicative collection region is used. In the multiplicative region, where V is above the maximum value for saturation collection, the additional current is due to secondary ionization processes which result from the high velocities reached by the ions (particularly electrons) moving in the high field gradient. The use of this current amplification makes multiplicative collection methods inherently sensitive but unfortunately also inherently critical to many experimental variables.

Lauritsen-type Electroscope. We will call the gas-filled electrode systems designed for saturation collection ionization chambers. Saturation current instruments consist of the ionization chamber, in which ions produced are collected with as little recombination or multiplication as possible, and an electric system for measuring the very small currents obtained. The essential differences between the various instruments of this sort are in the nature of the current-measuring systems. In one common and relatively inexpensive instrument, the Lauritsen-type electroscope, a sensitive quartz-fiber electrometer measures the change in voltage produced on the fiber and its support by collection of the ionization charge. An external battery or rectifier is used to provide the initial voltage V (by means of a temporary connection to the fiber support); then, the fiber position is observed through a small telescope to measure ΔV as a function of time. For a collected charge q, the resulting $\Delta V = q/C$, where C is the approximately constant capacitance of the fiber and electrode system.

It is clear from this expression that the probability for barrier penetration decreases with increasing value of the integral in the exponent, that is, with increasing barrier height and width. (The higher the barrier, the larger is the difference $U(r) - E$; and the wider the barrier, the greater is the difference between the limits of integration R_0 and R_1.)

The decay constant λ may be considered as the product of P and the frequency f with which an α particle strikes the potential barrier; the order of magnitude of f may be estimated as follows. The de Broglie wave length h/Mv of the α particle of velocity v and momentum Mv inside the nucleus is taken comparable to R_0, thus

$$\frac{h}{Mv} \approx R_0, \quad \text{or} \quad v \approx \frac{h}{MR_0}.$$

If the α particle is considered as bouncing back and forth between the potential walls,

$$f = \frac{v}{2R_0}, \quad \text{or} \quad f \approx \frac{h}{2MR_0^2}.$$

Therefore, the decay constant

$$\lambda \approx \frac{h}{2MR_0^2} e^{-\frac{4\pi}{h}\sqrt{2M}\int_{R_0}^{R_1}\sqrt{U(r)-E}\,dr}.$$

By a more elaborate treatment more accurate expressions for f and λ are obtained. If a simple shape is assumed for $U(r)$ (for example a Coulomb law up to R_0 as indicated by a dotted line in figure VI–3), it is possible to evaluate the integral in the exponent and thus to obtain an explicit expression for λ in terms of R_0, Z, and E. The values of λ calculated for the known α emitters are in fair agreement with experimental values. Actually the known values of λ, Z, and E were used to calculate nuclear radii from this relation, and these were found to be in good agreement with values obtained by other methods (see chapter II, section C).

The Geiger–Nuttall rule relates λ and E by an empirical equation which may be written $\log \lambda = a \cdot \log E + b$; this is different in form from the Gamow theory, $\log \lambda = f(Z)E^{-\frac{1}{2}} + F(Z, R_0)$ where $f(Z)$ and $F(Z, R_0)$ are complicated functions of the variables indicated. However, owing to some compensating factors and mainly because log–log plots tend to minimize small irregularities, a Geiger–Nuttall plot fits experimental data fairly well.

It may be appropriate to say a few words about the fact that proton decay has not been observed. Protons are emitted by nuclei in less than about 10^{-12} sec or not at all. The explanation of the difference in behavior between α particles and protons is as follows. The proton barrier is half as high as the α-particle barrier and therefore also narrower, and for high excitation energies the life times for proton emission are thus much smaller than for α emission. For lower excitation energies α emission is more probable because the energy balance is much more favorable than for proton emission: the nucleons in the α particle are still about as tightly bound as in the original nucleus, whereas a proton to be emitted must be supplied with an energy of several million electron volts (its binding energy).

B. BETA DECAY

Electrons and Positrons. Electrons had been recognized as the ultimate units of negative electricity, and their properties had begun to be investigated, by the time radioactivity was discovered. Not long after their discovery β rays were characterized as electrons. Much later the existence and properties of positrons had been both predicted theoretically and established in cosmic-ray studies before positron emission from artificially produced radioactive bodies was discovered. The existence of positrons or positive electrons was first postulated by P. A. M. Dirac on the basis of his relativistic quantum theory of the electron. Solutions of the relativistic wave equations reveal possible states of electrons with energies always larger than mc^2 (where m is the electron mass), but with either positive or negative signs. As to the physical meaning of the unobserved negative energy states of electrons Dirac suggested that normally all the negative energy states are filled. The raising of an electron from a negative- to a positive-energy state (by the addition of an amount of energy necessarily greater than $2mc^2$) should then be observable not only through the appearance of an ordinary electron, but also through the simultaneous appearance of a "hole" in the infinite "sea" of electrons of negative energy. This hole would have the properties of a positively charged particle, otherwise identical with an ordinary electron. The subsequent discovery of positrons, first in cosmic rays and then in radioactive disintegrations,

was soon followed by discoveries of the processes of pair production and positron–electron annihilation, which may be regarded as experimental verifications of Dirac's theory.

Pair production is the name for a process which involves the creation of a positron–electron pair by a photon of at least 1.02 Mev ($2\ mc^2$). It can be shown (see exercise 4 at the end of this chapter) that in this process momentum and energy cannot both be conserved in empty space; however, the pair production may take place in the field of a nucleus which can then carry off some momentum and energy. The cross section for pair production goes up with increasing Z and with increasing photon energy. Pair production may be thought of as the lifting of an electron from a negative- to a positive-energy state. The reverse process, the falling of an ordinary electron into a hole in the sea of electrons of negative energy, with the simultaneous emission of the corresponding amount of energy in the form of radiation, is observed in the so-called positron–electron annihilation process. This process accounts for the very short lifetime of positrons: whenever a hole in the sea of electrons is created it is quickly filled again by an electron. The energy corresponding to the annihilation of a positron and electron is released either in the form of two γ quanta emitted in nearly opposite directions (to conserve momentum) or, much more rarely, in the form of a single quantum if the electron involved in the annihilation is strongly bound (say, in an inner shell of an atom) so that a nucleus is available to carry off the excess momentum. (The latter process, although theoretically possible, has not been definitely established experimentally.) The two-quantum annihilation occurs principally with very slow positrons, that is, positrons which have almost come to rest by ionization processes; it is then accompanied by the emission of two γ quanta, each of energy equal to mc^2 (0.51 Mev); this radiation is often referred to as annihilation radiation.

Beta-ray Spectrum and the Neutrino. In contrast to α particles, β particles [2] from a given radioactive species do not belong to a

[2] By β particles we shall mean any electrons, positive or negative, emitted from nuclei. Whenever necessary, negative β particles or negatrons (β^-) and positive β particles or positrons (β^+) will be distinguished. Electrons originating in the extranuclear shells should not be referred to as β particles; they are often represented by the symbol e^-. In the early literature any electrons emitted in radioactive processes are usually called β particles.

limited number of energy groups but are emitted with a continuous energy distribution extending from zero up to a maximum value. The shapes of β-ray spectra have been studied in some detail by magnetic-deflection methods. Some typical shapes of β spectra are shown in figure VI–4. The average energy is usually about one-third the maximum energy. Maximum energies ranging from 15 kev to 15 Mev occur among known β emitters. Ever since its discovery, by J. Chadwick in 1914, the continuous spectrum of β rays has presented a very puzzling problem. Studies

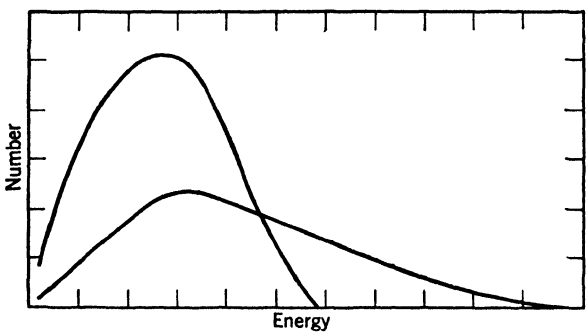


FIGURE VI–4. Typical shapes of β-ray spectra.

of the α- and γ-ray spectra have revealed that nuclei exist in definite energy states. Yet in every known β-decay process the transition from one such definite energy state to another takes place with the emission of β particles of variable kinetic energy. It was proved by calorimetric measurements that when all the β particles are absorbed in a calorimeter the measured energy per β particle is the average and not the maximum energy of the β spectrum. Thus the law of the conservation of energy might appear to be violated in β decay.

Furthermore, the observations show discrepancies with other conservation laws also. As we have seen in chapter II, all nuclei of even mass number have integral spins and obey Bose statistics; all nuclei of odd mass number have half-integral spins and obey Fermi statistics. Since the mass number remains unchanged in β decay the spins of initial and final nuclei should belong to the same class, either integral or half-integral, and the statistics should

remain the same. Yet electrons (and positrons) have one-half unit of spin and obey Fermi statistics. Thus angular momentum and statistics appear not to be conserved in β decay. Finally, recent experiments in which the recoil momenta of nuclei as well as the corresponding β-particle momenta were measured seem to indicate that linear momentum conservation is also violated in β decay.

To avoid the necessity of abandoning all these conservation laws for the case of β-decay processes, W. Pauli postulated that in each β disintegration an additional unobserved particle is emitted. The properties attributed to this hypothetical particle which has come to be known as the neutrino are such that the conservation difficulties are eliminated. The neutrino is supposed to have zero charge, spin $\frac{1}{2}$, and Fermi statistics, and it is thought to carry away the appropriate amount of energy and momentum in each β process to conserve these quantities. To account for the fact that neutrinos have never been detected, it is in addition necessary to assume that they have a very small or zero rest mass and a very small or zero magnetic moment. By careful measurements of the maximum energy of a β spectrum and determination of the masses of the corresponding β emitter and product nucleus, experimenters have set an upper limit to the rest mass of the neutrino at about 0.05 electron mass (or 25 kev).

The difficulties regarding conservation laws are entirely analogous in positron decay. In this case the hypothetical particle emitted is sometimes called the antineutrino. Since the properties of neutrino and antineutrino are supposed to be the same we shall speak only of neutrinos.

Theory of β Decay. Using the neutrino hypothesis, E. Fermi in 1934 constructed a theory of β decay, somewhat analogous to the theory of emission of radiation from atoms. In this theory β emission is treated as the transition of a nucleon from the neutron state to the proton state (or the reverse) with the simultaneous creation of an electron (or positron) and a neutrino. Choosing a form for the interaction between nucleons and the electron–neutrino field analogous to that between atoms and the radiation field, Fermi arrived at an equation relating the decay constant with the maximum momentum of the β particles and

with the shape of the β spectrum.[3] Although the Fermi theory accounts qualitatively for the observed shapes of β spectra it appears that the theory may not in all cases give quantitatively correct results. The best fit at the upper-energy limit is obtained when the neutrino mass is taken to be zero or negligibly small.

Selection Rules. According to the Fermi theory the disintegration probability of a β emitter depends on, among other factors, the spin difference ΔI between initial nucleus and final nucleus. In first approximation only transitions with $\Delta I = 0$ and no change in parity are allowed. In higher approximations it turns out that transitions with $\Delta I = \pm 1$ should be roughly 100 times less probable than those with $\Delta I = 0$; those with $\Delta I = \pm 2$ another factor of 100 less probable, etc. These are called the Fermi selection rules for β decay. If the possibility that a nucleon may reverse its spin orientation during β decay is taken into account, transitions with $\Delta I = 0$ or ± 1 and no change in parity become allowed; those with $\Delta I = \pm 1$ or ± 2 and no change in parity "singly forbidden," etc. These latter rules, the so-called Gamow–Teller selection rules, appear to agree much better with experimental evidence than do the Fermi rules.

For maximum β energies considerably greater than 0.5 Mev the Fermi equation for the decay constant reduces to the approximate form $\lambda = k(E_{\max})^5$, where $E_{\max}$ is the maximum energy of the β spectrum, and the value of the constant k depends on the "degree of forbiddenness" of the transition. Thus for a given

[3] Once an interaction between nucleons and electron–neutrino field is chosen, one might hope this same interaction would account for the nuclear binding forces in terms of an exchange of electron–neutrino pairs between protons and neutrons (and perhaps an exchange of electron–positron or neutrino–antineutrino pairs between like nucleons). The Fermi theory leads to exchange forces much too small to account for nuclear binding. Exchange of a single, positive, negative, or neutral particle (rather than a pair of particles) to account for nuclear forces was first proposed by H. Yukawa; in his theory β decay was thought of as the emission of such a particle followed by immediate breakup into electron (or positron) and neutrino. This theory gives better agreement with known nuclear binding energies than the Fermi theory if the particle responsible for the exchange forces has a mass approximately 200 to 300 times the electron mass. Attempts have been made to identify this particle with some of the types of mesons found in cosmic rays. It is now thought that the π mesons (table II–3) may be responsible for nuclear binding. However, none of the meson theories of nuclear forces proposed so far appears to be entirely satisfactory.

degree of forbiddenness a linear relation would be expected between $\log \lambda$ and $\log E_{max}$, and on a plot of $\log \lambda$ vs. $\log E_{max}$ the points representing the various β emitters should fall on a series of parallel lines, the top line representing the allowed transitions, the next lower one the first forbidden transitions, etc. Prior to the development of the Fermi theory B. W. Sargent called attention to the fact that the natural β emitters could be represented on two such lines, and these lines are called Sargent curves. For the quantitative correlation of decay constants, maximum energies, and spin changes for the large number of artificially produced β emitters, Sargent diagrams have proved of little value. The data scatter over a large area in a Sargent plot, with little evidence for definite lines. However, it may be noted that few if any points fall above the line of allowed transitions; thus for a given E_{max} an approximate lower limit for the half-life can be determined, and for a given half-life an approximate lower limit for E_{max} can be found.

Complex β Spectra. In connection with the relation between decay constant and E_{max} it should be noted that a given β emitter may decay to several different quantum states of the product nucleus, with corresponding β-particle spectral distributions of different maximum energies. The relative probability of the transition to a particular state will then, of course, depend on the selection rules governing that transition. However, for the different transitions to be observed their relative probabilities must not differ by many orders of magnitude.

Whenever a β transition leads to an excited state of the product nucleus, it is followed by the emission of one or more γ quanta (or the excitation energy is lost in some other way discussed in section C). A complete diagram of the energies and genetic relationships of the β and γ rays is called a decay scheme. A few typical decay schemes are shown in figure VI–5.

The analysis of complex β spectra into components is very difficult. The existence of more than one component can be rather easily established from β-ray spectrograph data (and sometimes even from absorption data) if the intensities are not too different and the energies not too similar; but it is hard to determine the maximum energy of the lower-energy components. Sometimes this can be accomplished by the coincidence methods mentioned in chapter VIII, section D.

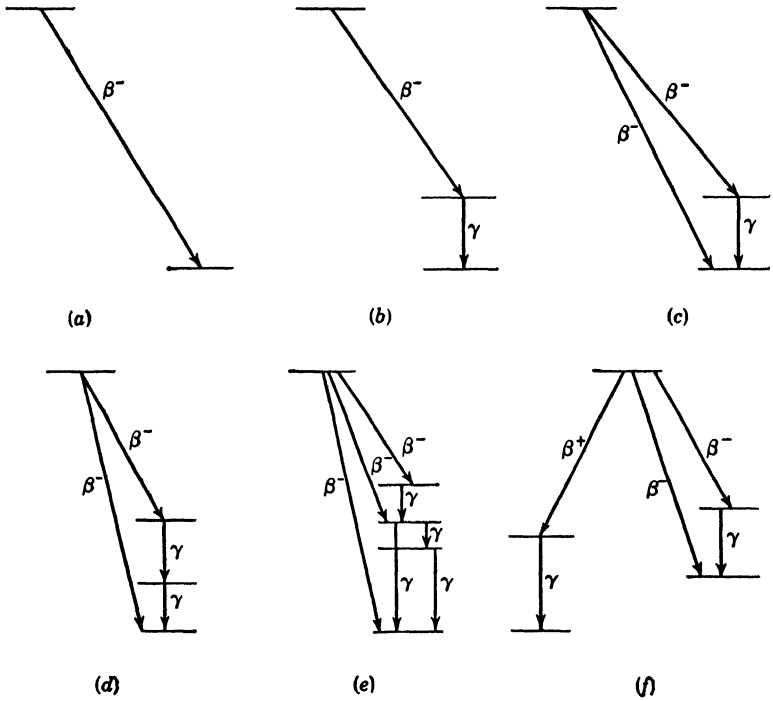


FIGURE VI–5. A few typical β-decay schemes.

(*a*) Simple β spectrum.
(*b*) Simple β spectrum followed by γ quantum.
(*c*) Two β spectra; the γ energy equals the difference between the maximum β energies.
(*d*) Two β spectra; the lower-energy one being followed by two γ quanta in cascade.
(*e*) Three β spectra with four γ rays.
(*f*) β^+–β^- branching.

***K*-electron Capture.** As we have seen, positron emission may be considered as the transformation of a proton into a neutron with the simultaneous emission of a positron (and a neutrino). An alternative way for a proton to transform into a neutron (and thus for a nucleus to decrease its charge by one unit) is by the capture of an electron, and this process is also observed. As the K electrons in an atom are, on the average, closest to the nucleus (or quantum-mechanically the wave functions of the K electrons have larger amplitudes at the nucleus than those of the L, M,

etc., electrons) the capture probability is greatest for the K electrons; the process is therefore called K-electron capture, or K capture, although L, M, etc., electrons may also be captured, but with smaller probabilities.[4] (In the sense of the Dirac theory, positron emission is the capture of an electron from the continuum of negative-energy states.) In K-electron capture, as in other β processes, momentum, angular momentum, and statistics cannot be conserved unless a neutrino is simultaneously emitted. However, since the electron is captured from a definite energy state, the neutrinos emitted in this process are presumably monoenergetic.

The electron-capture process may be difficult to observe because it is not necessarily accompanied by the emission of any detectable nuclear radiation, except in cases where the product nuclei are left in excited states and γ rays are emitted. The most characteristic radiations accompanying electron capture are the X rays emitted as a consequence of the vacancy created in the K (or L, etc.) shell. These X rays ordinarily include the entire X-ray spectrum of the product element although the K lines are usually by far the most prominent ones. Emission of Auger electrons from the excited extranuclear structure accompanies K capture. Auger electrons result from what may be described as an internal photoelectric effect; for example, the emission of a K X ray may be replaced by the ejection of an L electron with a kinetic energy equal to the difference between the K X-ray energy and the L binding energy.

Stability Considerations. In general, nuclei with neutron–proton ratios greater than that corresponding to the stability region decay by β^- emission, and most nuclei with neutron–proton ratios less than that required for stability decay by β^+ emission or K-electron capture. Occasionally a nuclide with two adjacent stable isobars may decay both by β^- emission and by one or both

[4] It is clear that a nucleus stripped of all extranuclear electrons cannot undergo K capture. By the same reasoning it is evident that it should be possible to change the half-life for a K-capture process by altering the electron density near the nucleus. Attempts to detect differences in the half-life of K-capturing Be^7 in different states of chemical combination have recently been reported [see E. Segrè and C. E. Wiegand, *Phys. Rev.* **75**, 39 (1949)]. Although this is a very favorable case for testing the effect, the reported differences in half-life are surprisingly small and may not be real.

of the other modes (for example, Cu^{64} and As^{74}). Electron capture [5] by a nucleus Z^A is energetically possible if

$$M_{Z^A} > M_{(Z-1)^A} + \mu \qquad \text{(VI-1)}$$

where the M's are the atomic masses,[6] and μ is the mass of the neutrino. Positron emission by the nucleus Z^A requires that

$$M_{Z^A} > M_{(Z-1)^A} + 2m_e + \mu, \qquad \text{(VI-2)}$$

where m_e is the electron mass (5.486×10^{-4} atomic weight unit). In cases intermediate between conditions VI-1 and VI-2, that is, when

$$M_{(Z-1)^A} + \mu < M_{Z^A} < M_{(Z-1)^A} + 2m_e + \mu,$$

protons can transform into neutrons only by electron capture; for nuclei to which relation VI-2 applies, both electron capture and positron emission are possible. The ratio of the probability of β^+ emission to that of K capture increases with increasing disintegration energy and decreases with increasing Z. No case of β^+ emission has been definitely established for any element heavier than the rare earths.

The condition for instability of a nucleus $(Z-1)^A$ with respect to β^- emission can be written

$$M_{(Z-1)^A} > M_{Z^A} + \mu. \qquad \text{(VI-3)}$$

Considering now two isobars of atomic numbers Z and $Z-1$ we see from relations VI-1 and VI-3 that, if μ is zero,

[5] Relation VI-1 is the condition for capture of a free electron. If the available energy is very little, it may not permit the capture of an electron bound in the K shell; in such cases either a free electron or one from a shell of sufficiently low binding energy would have to be captured, and this probability might be quite small.

[6] Conditions VI-1,2,3 can perhaps be more readily derived by considering the masses of the bare nuclei; denoting these by small m's, we can write down the following conditions:

For electron capture by Z^A: $m_{Z^A} + m_e > m_{(Z-1)^A} + \mu$;
For β^+ emission by Z^A: $m_{Z^A} > m_{(Z-1)^A} + m_e + \mu$;
For β^- emission by $(Z-1)^A$: $m_{(Z-1)^A} > m_{Z^A} + m_e + \mu$.

one of the two isobars must be unstable: if $M_{Z^A} > M_{(Z-1)^A}$, the nucleus Z^A is unstable with respect to electron capture; if $M_{Z^A} < M_{(Z-1)^A}$, the nucleus $(Z-1)^A$ is unstable with respect to β^- emission. If the neutrino has a small but finite rest mass it may be possible for a pair of neighboring isobars to be energetically stable.[7]

Three pairs of apparently stable neighboring isobars are known, and are listed in table VI–1. Whether both members of each

TABLE VI–1

PAIRS OF APPARENTLY STABLE NEIGHBORING ISOBARS

	$_{48}Cd^{113}$	$_{49}In^{113}$	$_{49}In^{115}$	$_{50}Sn^{115}$	$_{51}Sb^{123}$	$_{52}Te^{123}$
Relative abundance (%)	12.3	4.2	95.8	0.4	42.8	0.89
Spin (units of ħ)	½	9/2	9/2	½	7/2	

pair are really stable or whether the half-lives are so long or the radiations so soft that no activity has yet been detected cannot be stated with certainty; indeed long half-lives could be rationalized in terms of the large spin differences. The fact that the member of each pair which has the higher Z has a relatively low abundance has sometimes been taken as an indication that these substances may have been disappearing by K-capture activity. Another pair of neighboring isobars until recently regarded as stable is $_{75}Re^{187}$ and $_{76}Os^{187}$; however Re^{187} (62.9 per cent abundant) has been shown to decay to Os^{187} (1.64 per cent abundant) by β^- decay with a half-life of $(4 \pm 1) \times 10^{12}$ years. The recently discovered naturally occurring La^{138} (abundance 0.089%) has two stable neighboring isobars (Ba^{138} and Ce^{138}) and is very

[7] Conditions similar to equations VI–1 to VI–3 can be set up for isobars differing by two units of charge. For example the nucleus $(Z-2)^A$ is unstable with respect to decay into Z^A with the simultaneous emission of two β^- particles if $M_{(Z-2)^A} > M_{Z^A}$. It is interesting to note that conservation laws do not make it necessary to postulate neutrino emission in such a double β decay process. The expected half-lives for simultaneous emission of two β particles (or simultaneous capture of two orbital electrons) are exceedingly long, and it is therefore not surprising that many pairs of apparently stable isobars differing by two units of charge are known. Recently some experimental evidence has been presented for the double β^- decay of Sn^{124} with a half-life of about 10^{16} years. [E. L. Fireman, *Phys. Rev.* **75**, 323 (1949)].

probably radioactive although its activity has not yet been found. Three other naturally occurring nuclides with stable neighboring isobars have long been known to be radioactive. These are K^{40}, Rb^{87}, and Lu^{176}; their properties may be found in table I–2, or in table A in the appendix.

An interesting consequence of the prohibition (or near prohibition) of stable neighboring isobars is the absence of stable isotopes of the elements of atomic numbers 43 (technetium) and 61. All mass numbers from 94 to 102 are occupied by isotopes of $_{42}$Mo or $_{44}$Ru or both, and all mass numbers from 142 to 150 are occupied by isotopes of $_{60}$Nd or $_{62}$Sm or both. Yet it is just in these mass regions that stable isotopes of technetium and element 61, respectively, would be expected.

C. GAMMA DECAY AND ISOMERISM

Gamma Radiation and Internal Conversion. In the discussion of the high-energy α particles from RaC′ and ThC′ (on page 126), we mentioned that a study of these α particles and the γ rays emitted from the same nuclear levels permits an estimate of the "lifetimes" of these levels with respect to γ emission. These lifetimes are of the order of 10^{-13} sec. In general, theoretical calculations indicate that ordinarily the half-lives for γ emission are unobservably short ($<10^{-9}$ sec) if dipole or quadrupole radiation is emitted. This is the case for the vast majority of γ rays following α- or β-decay processes as well as neutron capture and other nuclear reactions. Gamma-ray energies between about 10 kev and about 6 Mev have been observed in radioactive decay.

Gamma-ray emission may be accompanied, or even replaced, by another process, the emission of internal-conversion electrons. Internal conversion has been pictured as a photoelectric effect produced by a γ ray in the electron shell surrounding the γ-emitting nucleus. An extranuclear electron is emitted with a kinetic energy equal to the difference between the γ energy and the binding energy of the electron in the atom. Actually the emission of internal-conversion electrons may be regarded as an additional alternative process for the de-excitation of a nucleus. Internal-conversion electrons have line spectra, and for a β transition followed by internal conversion the conversion electron lines are superimposed on the β spectrum.

We define for a given transition between two quantum states an internal conversion coefficient [8]

$$\alpha = \frac{\text{number of internal-conversion electrons emitted}}{\text{number of } \gamma \text{ quanta emitted}}.$$

This coefficient α may have any value between 0 and ∞. The internal-conversion coefficient generally decreases with increasing energy of the transition, and γ rays of 0.5 Mev or greater energy are usually not accompanied by appreciable numbers of conversion electrons. The magnitude of the internal-conversion coefficient increases with increasing multipole order of the transition (see below).

Internal conversion involving emission of electrons from the K shell is most probable (if K conversion is energetically possible), and the probabilities decrease for successive shells. Some cases are known in which the energy of the transition is less than the binding energy of the K electrons; then only L, M, etc., conversion can take place (for example, the 1.5-min Ir^{192} isomer). Partial conversion coefficients for the various shells are often used; for example the K-conversion coefficient

$$\alpha_K = \frac{\text{number of conversion electrons emitted from the } K \text{ shell}}{\text{number of } \gamma \text{ quanta emitted}}.$$

The total conversion coefficient equals the sum of the partial coefficients. The energy differences between the conversion electrons from different shells but due to transitions between the same pair of nuclear levels are characteristic of the element; this fact is often used either to identify the element or to decide which electron lines (in an electron spectrogram) result from the same nuclear transition.

Since internal conversion leaves a vacancy in one of the inner electron shells, the emission of conversion electrons is always accompanied by the emission of characteristic X rays and Auger electrons. The mode of decay of an active X-ray-emitting isotope

[8] Caution should be exercised in the use of data on internal-conversion coefficients from the literature. Some authors define the internal-conversion coefficient as the ratio of the number of internal-conversion electrons to the total number of transitions (number of quanta + number of conversion electrons). The coefficient defined in that way is restricted to values between 0 and 1.

of element Z can often be determined from a study of the characteristic X-ray spectrum by critical-absorption methods as is discussed in chapter X, section B.

Isomerism. The phenomenon of nuclear isomerism, that is, the existence of a nucleus of given Z and A in more than one energy state of measurable lifetime,[9] is generally explained in terms of highly forbidden γ-ray transitions making the excited states metastable. Attempts have been made to formulate on a theoretical basis quantitative relations between the lifetime of an excited state, the energy of the transition, the spin change, and the internal-conversion coefficient. The predicted correlations appear to be in fairly good agreement with experimental observations. Some of the theoretical results are briefly summarized in the next paragraph.

Electric dipole radiation is emitted when the initial and final states of the nucleus have opposite parity and a spin difference ΔI of 0 or ± 1. Electric quadrupole radiation corresponds to $\Delta I = \pm 1$ or ± 2 with no parity change. Lifetimes of excited states increase with increasing multipole order and with decreasing energy of the transition. For electric octopole radiation ($\Delta I = \pm 2$ or ± 3, parity change) half-lives of about 10^{-5} sec to minutes may be expected for energies between 200 kev and 20 kev; 2^4-pole radiation corresponds to half-lives of seconds to days for that energy range, etc. When, for a given $\Delta I = l$, electric 2^l-pole radiation is forbidden by the parity selection rule, magnetic 2^l-pole radiation may compete with electric 2^{l+1}-pole radiation, with comparable lifetimes.[10] It has also been shown that a transition between two states of zero spin is highly forbidden if the two states have the same parity. A few cases of excited states of measurable lifetimes have been tentatively ascribed to this effect, for example, a 5×10^{-7}-sec isomeric state of Ge^{72}. For low transition energies and fairly large spin differences, emission of conversion electrons is a more probable process than γ emission, and lifetimes calculated from γ-emission probabilities must be

[9] With present techniques the lower limit for a measurable lifetime is about 10^{-8} sec.

[10] M. L. Wiedenbeck [*Phys. Rev.* **69**, 567 (1946)] has shown that most known isomeric transitions fit quite well the simple relations $\lambda = 42E^{3.67}$ (for electric 2^4- or magnetic 2^3-pole radiation) and $\lambda = 45.8 \times 10^{-5}E^{3.67}$ (for electric 2^5- or magnetic 2^4-pole radiation), where λ is in sec^{-1} and E in Mev.

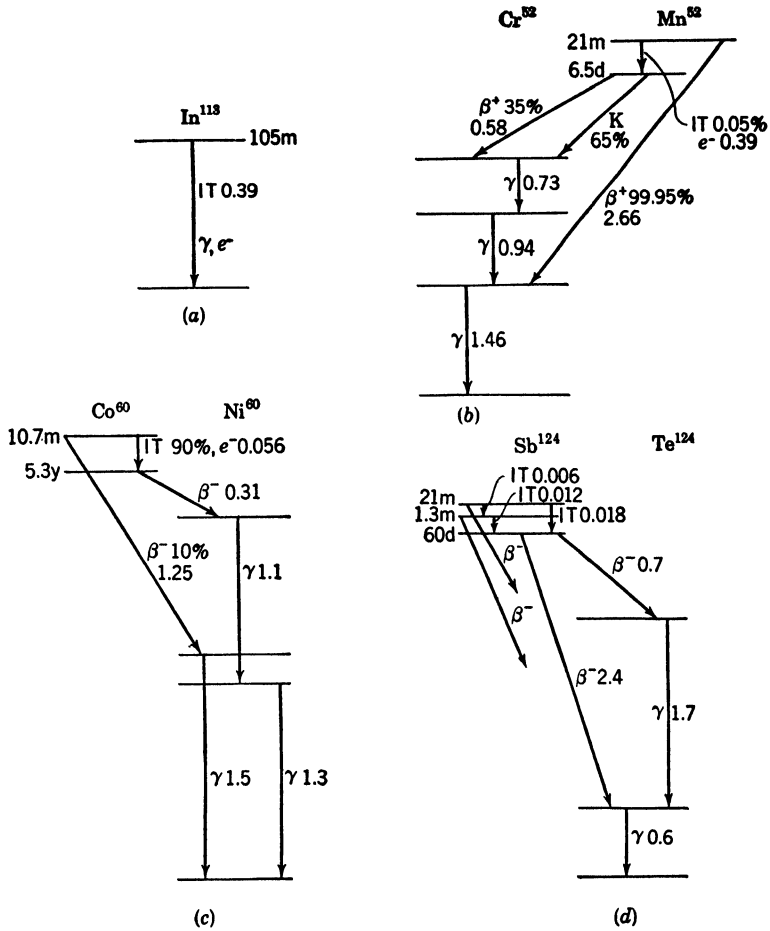


FIGURE VI–6. Some typical decay schemes of nuclear isomers. Energies are given in million electron volts.

(*a*) 105-min excited state of In^{113} decays entirely to ground state by isomeric transition.

(*b*) 21-min upper state of Mn^{52} decays 99.95 per cent by β^+ emission to Cr^{52}.

(*c*) 10.7-min upper state of Co^{60} decays 90 per cent by isomeric transition to 5.3-year lower state, 10 per cent by β^- emission to Ni^{60}.

(*d*) The first case of triple isomerism reported is in Sb^{124}. The 21-min and 1.3-min excited states decay by partially *L*-converted isomeric transitions as well as by β^- emission. The decay scheme shown is incomplete; for example several other β^- and γ energies are known to be associated with the 60-day activity.

correspondingly reduced $\left(\text{multiplied by the factor } \frac{1}{1+\alpha}\right)$ when the conversion coefficients are high. Experimentally, very high conversion coefficients are common in low-energy isomeric transitions.

Excited isomeric states with half-lives ranging from about 10^{-7} sec to several months are known. Depending on the relative decay probabilities an excited isomeric state may transform predominantly into its associated lower isomeric state by γ-ray or conversion-electron emission or predominantly to a neighboring isobar by a β or K-capture process. For example, the 105-min In^{113} transforms into stable In^{113} by emission of conversion electrons, and the 24.5-min Ag^{106} emits positrons to become Pd^{106}; in 21-min Mn^{52} both the transition to the lower isomeric state and β^+ emission occur to an observable extent. Transition to a lower isomeric state is called isomeric transition, abbreviated IT. Some typical decay schemes for isomers are shown in figure VI–6. At least one case of triple isomerism has been established; this is in $_{51}Sb^{124}$, with 21-min, 1.3-min, and 60-day periods (figure VI–6).

D. OTHER MODES OF DECAY

Spontaneous Fission. The explanation of the fission process in terms of the liquid drop model naturally leads one to inquire whether a nucleus that can be split by the addition of a relatively small amount of excitation energy may not have a finite probability of undergoing fission spontaneously. This process of spontaneous fission has indeed been discovered in uranium. The isotope U^{238} accounts for most of the spontaneous fission processes observed; its disintegration constant for spontaneous fission is about 2×10^{-24} sec^{-1}. If spontaneous fission were the only process by which U^{238} decayed, this would correspond to a "half-life" of 10^{16} years. Also Pu^{239} has been investigated, and its "half-life" for spontaneous fission reported as $>10^{14}$ years. Obviously spontaneous fission contributes only a negligible fraction of the decay of these nuclear species, and their half-lives are practically entirely determined by their α decay. However, it is quite possible that still heavier elements may have much higher disintegration constants for spontaneous fission and that this

may at least in part account for the absence of transuranium elements in nature.

"Delayed" Neutrons. As has been pointed out before, nucleon emission with measurable lifetimes has not been observed and is not expected on theoretical grounds. The emission of so-called "delayed" neutrons, with half-lives of from fractions of a second to about 1 min, following fission, has been shown to be due to practically instantaneous emission of neutrons from highly excited states of fission products following β^- decays. This process can occur when β^- decay leaves the product nucleus in a state of such high excitation (at least several million electron volts) that neutron emission can compete successfully with γ emission. The neutron activity then persists with the half-life of the preceding β^- decay. Some of the delayed neutron periods have been identified with particular β^--emitting fission products by chemical analysis; the 56-sec β^- decay of Br^{87} and the 22-sec β^- decay of I^{137}, for example, are followed by neutron emission. Recently the 4.1-sec β^- decay of N^{17} (formed in various nuclear reactions) has been shown to be followed by neutron emission.

EXERCISES

1. In the decay of AcC(Bi^{211}) to AcC″(Tl^{207}) the following α- and γ-ray energies have been observed: α 6.611, 6.262 Mev; γ 0.354 Mev. Construct a reasonable decay scheme for this disintegration. Indicate the *total* energy difference between each energy state involved and the ground state of AcC″.

2. Estimate the decay constant for the 10.54-Mev α-particle emission in ThC′, and the decay constant for γ emission from the same ThC′ level.
Answer: $\lambda_\alpha \approx 5 \times 10^{11}$ sec^{-1}; $\lambda_\gamma = 100\lambda_\alpha$.

3. Represent on a Sargent diagram the β^- decays of H^3, C^{14}, Na^{24}, P^{32}, S^{35}, K^{40}, Co^{60} (5.3 y), Zn^{69} (57 m), Br^{82}, Sr^{89}, Y^{91} (57 d), Cb^{95} (35 d), Te^{127} (9.3 h), Te^{129} (70 m), I^{131}. What conclusions, if any, can you draw?

4. Show that the production of a positron–electron pair by a photon in vacuum is impossible. (*Note*: Set up the conditions for momentum and energy conservation, using relativistic expressions, and show that they lead to a contradiction, for example to the inequality $\cos\theta > 1$, where θ is the angle between the directions of motion of positron and electron.)

5. Estimate the multipole order of the isomeric transition in Te^{125}.

6. Collect from the literature as complete information as you can on the decay schemes of V^{48}, Na^{22}, I^{131}. (*Note.* The Physical Review is a good source of information.)

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VII. INTERACTION OF RADIATIONS WITH MATTER

A. ALPHA PARTICLES

Processes Responsible for Energy Loss. Nuclear radiations, both corpuscular and electromagnetic, are detectable only through their interactions with matter. If this interaction is sufficiently small, as in the case of the neutrino, the radiation remains undetected. For an understanding of the methods and instruments used for the detection, measurement, and characterization of nuclear radiations it is necessary to consider the manner in which these radiations interact with matter.

In passing through matter α particles lose energy chiefly by interaction with electrons.[1] This interaction may lead to the dissociation of molecules or to the excitation or ionization of atoms and molecules. The effect which is most easily measured and most often used for the detection of α particles is ionization. The details of the ionization processes and other effects associated with α-particle passage are more readily investigated in gases than in liquids or solids, although the processes are presumably about the same. We shall therefore speak mostly of phenomena observed in the passage of α particles through gases.

Because α particles have relatively short ranges a known number of α particles of known initial energy can be made to spend their entire energy inside an ionization chamber, and thus the total ionization produced per α particle is readily measured. These experiments show that on the average about 35 ev (35 electron volts of energy) are dissipated for each ion pair formed in air. At least in the case of air this value is quite independent of the initial energy of the α particles. The energy required to form an ion pair is listed for a number of other gases in table VII–1, together with the first ionization potentials of these gases. In the noble gases a larger fraction of the α-particle energy is spent

[1] The interactions of α particles with nuclei (scattering and nuclear reactions) have been discussed in previous chapters. The contribution of these processes to energy loss of α particles in passing through matter is entirely negligible.

ionization processes than in the diatomic and polyatomic gases where dissociation of molecules is also possible.

Part of the energy loss of α particles is accounted for by the kinetic energy given to the electrons removed from atoms or molecules in close collisions with the α particle. It can easily be shown from conservation of momentum that the maximum velocity which an α particle of velocity v can impart to an electron is about $2v$; therefore, the maximum energy which an electron can

TABLE VII-1

AVERAGE ENERGY LOST BY α PARTICLES IN PRODUCING ONE ION PAIR IN VARIOUS GASES

	Energy per Ion Pair (K) (ev)	First Ionization Potential (I) (volts)	Difference (K − I)
H_2	33.0	15.4	17.6
He	27.8	24.5	3.3
N_2	35.0	15.5	19.5
O_2	32.3	12.5	19.8
Ne	27.4	21.5	5.9
A	25.4	15.7	9.7
Kr	22.8	13.9	8.9
Xe	20.8	12.1	8.7
CH_4	30.0	14.5	15.5
NH_3	38.9	11.2	27.7
CS_2	25.6	10.4	15.2

receive from the impact of a 6-Mev α particle, for example, is about 3000 ev. The average energy imparted to electrons by α particles in their passage through matter is of the order of 100 to 200 ev. Many of these secondary electrons or δ rays are fast enough to ionize other atoms. In fact about 60 to 80 per cent of the ionization produced by α particles is due to secondary ionization; the exact ratio of primary to secondary ionization is very difficult to determine. Delta-ray tracks are often seen in cloud-chamber pictures of α-particle tracks.

Range. Because an α particle loses only a very small fraction of its energy in a single collision with an electron and is not appreciably deflected in the collision, α-particle paths are very nearly straight lines. Furthermore, because of the very large number of collisions (of the order of 10^5) necessary to bring an α particle of a few million electron volts initial energy to rest, the ranges

of all α particles of the same initial energy are the same within narrow limits. In figure VII–1 the number of α particles found in a gas at a distance r from the source is plotted against r for the case of a source which emits α particles of a single energy. It is seen that the ranges of all the particles in a given medium are not exactly the same but show a small spread of about 3 or 4 per cent. This phenomenon, called the straggling of α-particle

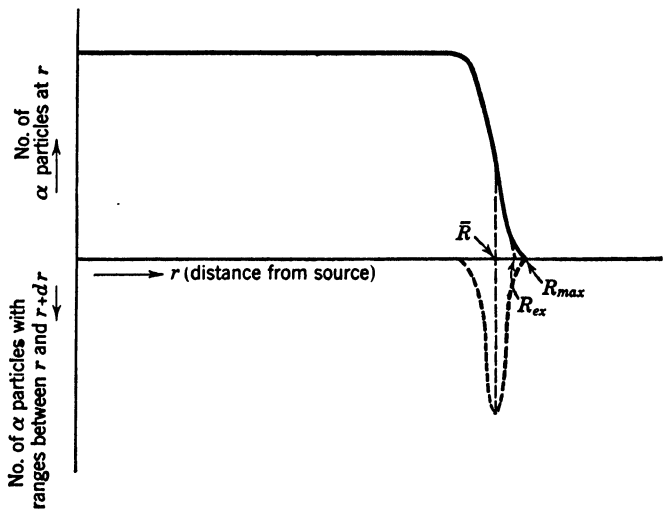


FIGURE VII–1. Number of α particles from a point source as a function of the distance from the source (full curve). The derivative of that function is also shown (dotted curve); the latter represents the distribution in ranges.

ranges, is caused by the statistical fluctuations in the number of collisions and in the energy loss per collision. The dotted curve in figure VII–1 is obtained by differentiating the other (integral) curve and represents the distribution of ranges or the amount of straggling; it is approximately a Gaussian curve. The distance r corresponding to the maximum of the differential curve (point of inflection of the integral curve) is called the mean range $\bar{R}$ of the α particles. The distance r obtained by extrapolating to the abscissa the approximately straight portion of the integral curve is the extrapolated (or practical) range R_{ex}. The largest observed range is called the maximum (or true) range R_{max}. Thus, although the range of an α particle is a measure of its energy and can be rather readily determined experimentally, it is necessary to specify

which range is expressed. The mean range is least dependent on the particular experimental setup and is now generally used in range tables and in range–energy relations. Extrapolated ranges were often given in the older literature and are more easily determined experimentally.

Alpha-particle ranges are usually given for air at 15°C and 760 mm pressure (although 0°C is used as this standard temperature by some authors). The range is inversely proportional to the density of air. Range–energy relations based on calculations and on energy determinations by magnetic-deflection experiments coupled with range measurements have been published by many authors.[2] For mean α-particle ranges between 3 and 7 cm the empirical equation $\bar{R} = 0.318\ E^{3/2}$, with $\bar{R}$ in centimeters of air at 15°C and 760 mm pressure and E in millions of electron volts, holds fairly well. Ranges of α particles are generally determined by absorption methods, either with solid absorbers or, more accurately, by varying the pressure of an absorbing gas. Cloud-chamber photographs of α-particle tracks are also used. Ranges may be determined with a precision of about one part in 5000.

Stopping Power. Comparisons of the ranges of α particles in different substances introduce the concept of the stopping power of a particular substance. Formally the stopping power F is defined as the space rate of energy loss of an α particle in the substance, $F = -dE/dx$, and thus has the dimensions of a retarding force. Because F is generally not independent of energy E we write $dx = -dE/F(E)$, and the range $R = -\int_{E_0}^{0} dE/F(E)$ for a particle of initial energy E_0. Experimentally one can determine $F(E)$ by measuring the range as a function of energy and then taking the derivative of that function: $dR/dE = 1/F(E)$. In common practice the energy dependence of stopping power is often neglected and an average value for the whole range used in approximate calculations.

Bragg's empirical rule for stopping-power variations among different substances is that the stopping effect per atom, called the atomic stopping power s, is about proportional to $A^{1/2}$, that

[2] See, for example, M. S. Livingston and H. A. Bethe, *Rev. Mod. Phys.* **9**, 261–276 (1937). The same paper (p. 285) also gives the difference between extrapolated and mean range as a function of mean range.

is to the square root of the atomic weight. Because the number of atoms per cubic centimeter is proportional to the ratio of the density ρ to the atomic weight, another statement of this rule is that the stopping power per unit weight, called the mass stopping power, is about proportional to $A^{-\frac{1}{2}}$. The similarity of these two statements is just enough to be confusing; it is to be remembered that for stopping α particles a heavier atom is more effective, according to $A^{\frac{1}{2}}$, but is less effective per unit weight, according to $A^{-\frac{1}{2}}$.

Another empirical rule for the atomic stopping power, resembling Bragg's rule but giving a better result in most instances, is stated in terms of Z, the atomic charge, rather than A:

$$s = 0.563 \frac{Z}{\sqrt{Z + 10}}. \qquad \text{(VII-1)}$$

The proportionality constant 0.563 is chosen to give the best fit with experimental data in terms of a value of $s = 1$ for air.

In most actual cases, indeed for air, the stopping substance is not a single element but rather is a compound or mixture of elements. For practical purposes in using the approximate rules just stated we make the further approximation that the stopping power of a molecule or of a mixture of atoms or molecules is given by the sum of the stopping powers of all the component atoms. In view of the fact that a considerable fraction of the α-particle energy is expended in molecular excitation and dissociation processes, this simple additivity relation is somewhat surprising. The stopping power of water vapor has been measured to be about 3 per cent less than that of the equivalent mixture of hydrogen and oxygen; measurements for a number of organic isomers show that their molecular stopping powers are the same within less than 1 per cent. For systems containing deuterium equation VII–1 should be used rather than Bragg's rule, because H^1 and H^2 have very nearly the same atomic stopping power.

Probably the best procedure for making calculations of ranges in different substances compared to air is to apply to the given range appropriate correction factors for density, atomic or molecular stopping power, and so on, just as the pressure–volume–temperature relations are commonly handled in gas-law problems.

For example, a range in aluminum R_{Al} is given in terms of the range in air:

$$R_{Al} = R_{air} \times \frac{\rho_{air}}{\rho_{Al}} \times \sqrt{\frac{A_{Al}}{A_{air}}} = R_{air} \times \frac{0.001226}{2.70} \times \sqrt{\frac{27.0}{14.4}};$$

$$R_{Al} = 0.000622\, R_{air}.$$

The value 14.4 for the "atomic weight of air" is the square of the weighted average square-root value: $(0.8\sqrt{14} + 0.2\sqrt{16})^2 = 14.4$. Other mixtures and compounds are handled in the same way when Bragg's rule is used. For the same calculation but using equation VII–1, we obtain a slightly different result:

$$R_{Al} = R_{air} \times \frac{\text{no. atoms air per cm}^3}{\text{no. atoms Al per cm}^3} \times \left(0.563\, \frac{13}{\sqrt{13 + 10}}\right)^{-1},$$

$$R_{Al} = R_{air} \times \frac{0.001226/14.4}{2.7/27.0} \times \left(0.563\, \frac{13}{\sqrt{23}}\right)^{-1},$$

$$R_{Al} = 0.000558\, R_{air}.$$

As another illustration we calculate the range in SO_2 compared to the range in air, taking for the density of SO_2 at 15°C the ideal-gas-law value, $(64.1/22{,}400) \times (273/288) = 0.00272$; we have:

$$R_{SO_2} = R_{air} \times \frac{\text{no. atoms air per cm}^3}{\text{no. molecules } SO_2 \text{ per cm}^3} \times \frac{1}{\text{molecular stopping power of } SO_2},$$

$$R_{SO_2} = R_{air} \times \frac{0.001226/14.4}{0.00272/64.1} \times \frac{1}{s_{SO_2}},$$

$$s_{SO_2} = s_S + 2s_O = 0.563\, \frac{16}{\sqrt{16 + 10}} + 2 \times 0.563\, \frac{8}{\sqrt{8 + 10}} = 3.89,$$

$$R_{SO_2} = 0.515 R_{air}.$$

Often it is convenient to express absorber thickness not in centimeters but in milligrams per square centimeter. As an illustration of the use of this unit, a foil of gold of thickness 12.0 mg per cm^2 would be equivalent to about $12.0 \times \sqrt{14.4/197} = 3.24$ mg per cm^2 of air, or $3.24/1.226 = 2.65$ cm of air at 15°C.

Specific Ionization. The energy and velocity of an α particle decrease in each interaction. If the velocity of an α particle in an absorbing medium is plotted against distance from the source, a curve such as the one sketched in figure VII–2 is obtained. The

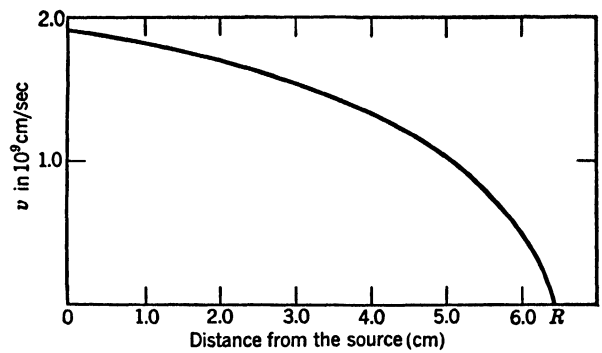


FIGURE VII–2. Velocity of a typical α particle versus distance from the source.

velocity of an α particle of E million electron volts is about $6.9 \times 10^8 E^{1/2}$ cm sec^{-1} (in the nonrelativistic region).

The specific ionization—the number of ion pairs formed per millimeter of path—varies with the velocity of the α particle and is approximately proportional to $1/v$ at energies in excess of about 1 Mev ($v > 7 \times 10^8$ cm sec^{-1}). The maximum specific ionization is reached by an individual α particle about 3.0 mm from the end of its range where it has a velocity of 4.2×10^8 cm sec^{-1}, or an energy of about 370 kev. After that the specific ionization falls rapidly to zero. For a beam of initially homogeneous α particles the maximum of the specific ionization occurs at about 4.7 mm from the extrapolated range. A plot of specific ionization versus distance from the source is called a Bragg curve (figure VII–3). For α particles of different energies the Bragg curves are almost identical over corresponding regions measured from the end of

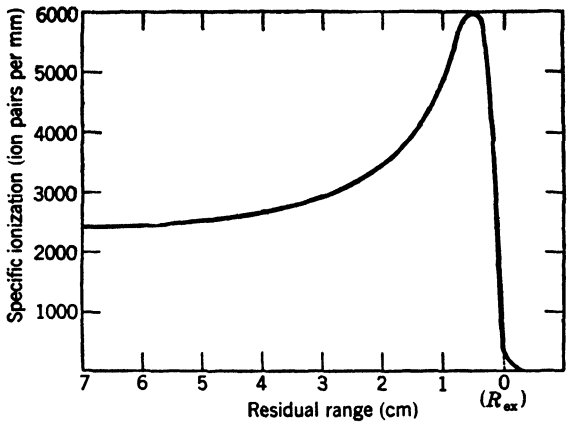


FIGURE VII-3. Bragg curve for initially homogeneous α particles.

the range. In table VII-2 specific ionization values are given for a number of residual ranges, that is for various distances from the end of the extrapolated range.

TABLE VII-2

SPECIFIC IONIZATIONS AND ENERGIES OF α PARTICLES AT VARIOUS RESIDUAL RANGES *

Residual Range (in cm of air) (15°C and 760 mm pressure)	Energy (Mev)	Ion Pairs per mm of air (15°C and 760 mm pressure)
0.21		4500
0.47		6000
1.0	1.9	4800
1.5	2.7	3960
2.0	3.4	3440
4.0	5.5	2680
7.0	7.8	2440

* The data in columns one and three are taken from E. Rutherford, J. Chadwick, and C. D. Ellis, *Radiations from Radioactive Substances*, Cambridge University Press, England, and The Macmillan Co., New York.

It should be noted that for a given α-particle source the extrapolated range obtained from a Bragg curve is not exactly the same as that obtained from a number-versus-distance curve, the latter being a few tenths of 1 per cent larger.

The straggling in specific ionization is complicated by the fact that near the end of their ranges the α particles frequently pick up electrons (becoming He^+ or even He) and subsequently lose them in later collisions. Several thousand such fluctuations in charge occur for each α particle, but they are almost completely confined to the last few millimeters of the range where the velocity of the α particle becomes comparable to the orbital-electron velocities in helium atoms. An α particle spends over 90 per cent of its entire path as He^{++}. The relative abundances of He^{++}, He^+, and He at various energies were studied by magnetic-deflection measurements with varying thicknesses of absorbers between source and measuring device.

Other Heavy Charged Particles. Any ion moving at high speed through matter loses its energy by essentially the same mechanism. Rather elementary calculations show that the rate of energy loss at a given velocity may be expected to vary approximately as Z^2, where Z is the net charge of the ion. This is found to be true experimentally. Alpha particles and protons of the same velocity have equal ranges because the α particle, with Z^2 four times as large, loses energy four times as fast and has just four times as much energy to lose. Deuterons and protons of the same velocity lose energy at the same rate, with $Z^2 = 1$ for both; then because the deuteron has twice the energy it has just twice the range of the proton. Range–energy relations for protons and deuterons are found in the literature.[3]

Fission fragments are 20 to 40 times heavier than α particles and have initial energies 10 to 20 times greater; thus their initial velocities are close to those of α particles. If a fission fragment were completely stripped of electrons the large value of Z^2, several hundred times greater than for an α particle, would lead to a range of the order of 1 mm in air. Actually the particle probably will be stripped only of electrons with orbital velocities smaller than its own velocity and so will hold all electrons with binding energies greater than about 1 kev.[4] Therefore, the fragment

[3] For example, Livingston and Bethe, *Rev. Mod. Phys.* **9**, 268 (1937).

[4] The kinetic energy of a bound electron (which is equal to its binding energy; see chapter II, page 25) with velocity equal to that of a fission fragment is in a nonrelativistic approximation the energy of the fission fragment multiplied by the ratio of the electron-to-fragment masses. For a 100-Mev fragment with mass 100, the binding energy of the corresponding electron is $100 \times 10^6 \times (0.00055/100) = 550$ ev.

starts with net $Z \approx 25$ and gains electrons as its velocity decreases until $Z \approx 0$ at about 1 Mev (approximately 3 mm before the end of the range). Measured fission fragment ranges are about 1.9 to 2.9 cm in air; a curve of specific ionization versus residual range looks about as shown in figure VII–4. If particles like fission fragments but of very much higher energy were available, the curve in figure VII–4 could be extended to larger residual ranges, and, no doubt, the same general features as in the Bragg curve

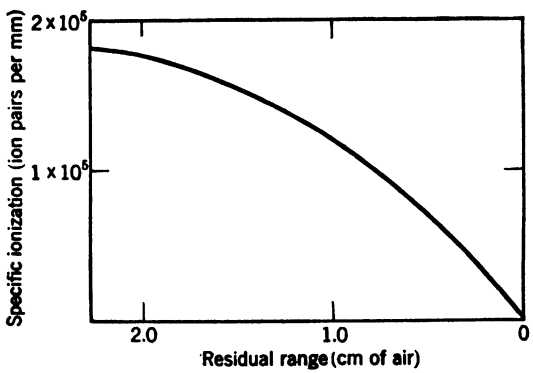


FIGURE VII–4. "Bragg curve" for a fission fragment.

for α particles would be seen. The relative stopping powers of various substances are very nearly the same for fission fragments as for α particles.

B. ELECTRONS

Comparison with α-particle Behavior. The interaction of electrons with matter is in many ways fundamentally similar to that of α particles. The processes which are responsible for the energy loss are the same in both cases. In fact, the average energy loss per ion pair formed is almost the same for electrons as for α particles (about 32.5 ev for electrons in air). The primary ionization by electrons accounts for only about 20 to 30 per cent of the total ionization; the remainder is due to secondary ionization.

We must consider a number of differences between the interactions of the two types of particles with matter. First, for a given energy the velocity of an electron is much larger than that of an α particle (see table VII–3), and, therefore, the specific ionization

is less for electrons. In table VII–3 the specific ionization in air is given for electrons of a few different energies. In the region from about 10 kev to 2 Mev the specific ionization has been shown to be nearly proportional to the inverse square of the velocity. The largest specific ionization, 770 ion pairs per millimeter, occurs

TABLE VII–3

VELOCITY AND SPECIFIC IONIZATION FOR ELECTRONS OF VARIOUS ENERGIES

E (Mev)	Velocity (in units of the velocity of light, c)	Ion Pairs per mm of air
10^{-6}	0.001979	0
10^{-5}	0.006257	0
10^{-4}	0.01978	
1.46×10^{-4}	0.0240	770 (maximum)
10^{-3}	0.06248	
10^{-2}	0.1950	~110
0.05	0.4127	20
0.10	0.5483	15
0.50	0.8629	6
1.0	0.9411	5
2.0	0.9791	4.5
3.0	0.9893	
4.0	0.9934	
5.0	0.9957	
10	0.9988	
20	0.99969	

at 146 ev (velocity = $0.024c$), which is a much lower energy but somewhat higher velocity than corresponds to the peak in the Bragg curve for α particles. In air, ionization stops when the electron energy has been reduced to 12.5 ev (the ionization potential of oxygen molecules).

An electron may lose a large fraction of its energy in one collision; therefore, a statistical treatment of the energy-loss processes is much less justified than for α particles, and straggling is much more pronounced. In the passage of an initially homogeneous beam of electrons through matter the apparent straggling is further increased by the pronounced scattering of the electrons into different directions, which makes possible widely different path lengths for electrons traversing the same thickness of ab-

sorber. Nuclear scattering as well as scattering by electrons is important.

For electrons of high energy an additional mechanism for losing energy must be taken into account: the emission of radiation (bremsstrahlung) when an electron is accelerated in the electric field of a nucleus. The ratio of energy loss by this radiation to energy loss by ionization in an element of atomic number Z is approximately equal to $\frac{E \cdot Z}{800}$, where E is the electron energy in millions of electron volts. Thus, in heavy materials such as lead the radiation loss becomes appreciable even at 1 Mev, whereas in light materials (air, aluminum) it is unimportant for the energies available from β emitters.

Finally, the additional fact that β particles are emitted with a continuous energy spectrum makes their absorption in matter a phenomenon too complicated for theoretical analysis.

Absorption of β Particles. The combined effects of continuous spectrum and scattering lead—quite fortuitously—to an approximately exponential absorption law for β particles of a given maximum energy. Absorption curves, that is, curves of activity versus thickness of absorber traversed, are for this reason usually plotted on semilogarithmic paper. The nearly exponential decrease applies both to numbers and specific ionizations of β particles, although absorption curves taken with counters and ionization chambers cannot be expected to be completely identical. The exact shape of an absorption curve depends also on the shape of the β-ray spectrum and, because of scattering effects, on the geometrical arrangement of active sample, absorber, and detector. If sample and absorber are as close as possible to the detector, the semilog absorption curve becomes most nearly a straight line; otherwise, some curvature toward the axes is generally found. When β particles belonging to two spectra of widely different maximum energies are present in a source, this is apparent from the change of slope in the absorption curve; such an absorption curve is roughly analogous to the semilog decay curve of an activity containing two different half-life periods.

If the absorption of β rays is represented by an exponential law $\mathbf{A}_d = \mathbf{A}_0 e^{-\mu d}$, where $\mathbf{A}_0$ is the measured activity without absorber and $\mathbf{A}_d$ the activity observed through absorber of thickness d, then μ is known as the absorption coefficient. The ratio of the

absorption coefficient to the density ρ, known as the mass absorption coefficient, is nearly independent of the nature of the absorber. More accurately it varies about as Z/A; that is, the number of electrons per unit mass determines the mass stopping power of a substance for β particles. The thickness required to reduce the activity to one half of its initial value is called the half-thickness $d_{1/2} = 0.693/\mu$; more frequently the half-thickness is expressed in grams per square centimeter, and then is equal to $0.693\rho/\mu$ and varies about as A/Z. Absorption coefficients and half-thickness values given in the literature usually refer to the initial portions of absorption curves. These values cannot be relied on as accurate measures of β-particle energies.

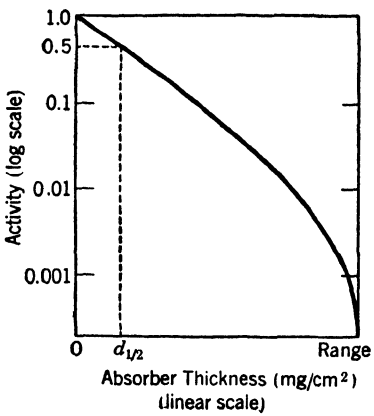


FIGURE VII–5. Idealized β-ray absorption curve (semilog plot).

Determination of β-particle Ranges. It is generally the purpose of absorption measurements to determine the upper energy limit of a β-ray spectrum. We should say at the outset that precision determinations of upper energy limits can be made only with electron spectrographs; yet for most purposes absorption measurements are much more convenient.

To get a measure of the upper energy limit of a β-ray spectrum one must find the range in the absorber of the most energetic β particles. The fact that a range exists for a given β-ray spectrum means that the absorption curve cannot continue as an approximate exponential but must eventually turn downward toward $-\infty$ on a semilog plot (see figure VII–5). The ratio of range to initial half-thickness is generally between 5 and 10. In practice a β-ray absorption curve is never found to reach $-\infty$ on a semilog plot and may not even turn in that direction, because of the presence of more penetrating radiation beyond the range of the β rays. Even if neither nuclear γ radiation nor characteristic X rays are present, there is always some background of bremsstrahlung from the deceleration of the β particles in the

sample itself and in the absorbers. If elements of low Z are used as absorbers, the difference in slopes between β-ray and γ- or X-ray absorption curves is particularly marked; the absorption curve then exhibits a fairly sharp break where the β-ray component turns over into the photon "tail" (see figure VII–6). For this reason β-ray absorption curves are always taken with absorbers

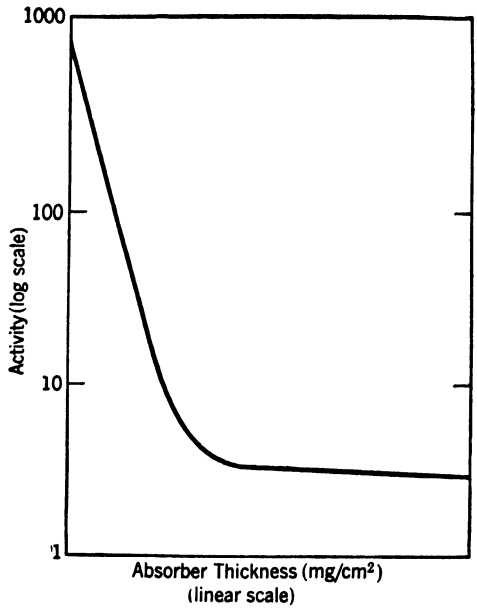


FIGURE VII–6. Typical β-ray absorption curve in aluminum (semilog plot). A γ-ray component is present.

of low atomic number; aluminum or plastic absorbers are most commonly used, and for differentiating β particles from soft X rays beryllium absorbers are particularly useful. (Beta-ray ranges expressed in milligrams per square centimeter vary only about as A/Z, whereas the absorption of electromagnetic radiation increases rapidly with Z as is discussed in section C.)

The relative efficiencies of measuring instruments for β and γ rays vary with the energies of the radiations and with different instruments; most Geiger–Müller counters and air-filled ionization chambers are about 100 times as efficient for β particles as for 1-Mev γ quanta, and the efficiency for γ rays is roughly pro-

portional to the γ-ray energy from a few hundred kiloelectron volts to a few million electron volts. Thus a typical β-ray absorption curve for a β spectrum accompanied by γ rays will have a γ-ray "tail" with intensity of the order of 1 per cent of the initial β activity. A pure bremsstrahlung tail is usually at least an order of magnitude smaller, about 0.05 per cent or less of the initial β activity. In β^+ absorption curves there is, in addition to other electromagnetic radiation which may be present, always a background of annihilation radiation, about 1 per cent of the initial β^+ intensity in a typical detector.

The maximum range for a β-particle spectrum may be obtained from an experimental absorption curve in various ways. Visual inspection gives a rough value (usually too small) for the point at which the β activity ceases to be detectable above the γ- or X-ray background; the lower the γ- or X-ray background, the better is the visual method. Better results can sometimes be obtained by subtraction of the penetrating background radiation from the total absorption curve, which should result in a curve similar to the one in figure VII–5.

The best method for the determination of β-ray ranges from absorption curves is the comparison method suggested by N. Feather. Here the absorption curve to be analyzed is compared with the absorption curve (measured under identical conditions) of a standard β emitter, usually RaE (E_{max} = 1.17 Mev, range = 476 mg per cm^2 in aluminum) or UX_2 (E_{max} = 2.32 Mev, range = 1105 mg per cm^2 in aluminum).[5] The net β-ray absorption curves after subtraction of all backgrounds due to electromagnetic radiation are used. If the same percentage reduction of initial activity would correspond to the same fraction of the range for each β emitter, the absorber thickness corresponding to a certain fraction (say 0.5) of the range could be readily determined for the unknown by comparison with the standard of known range. Actually this procedure gives a somewhat different apparent range for each fraction of the range at which the comparison is made, because of the different shapes of the absorption curves.

[5] The ranges given for the standard substances are those corresponding to the spectrographically determined E_{max} values according to the best range–energy relations (see p. 164). These, rather than the visual ranges determined for the standard substances in a particular experimental arrangement should be used in the Feather analysis.

But, if these apparent ranges are plotted against the fractions of the range at which they were determined, a smooth curve results which can be extrapolated to fraction 1.0 of the range to find the true range. This curve is called the Feather plot; one is shown in figure VII–7. If the spectra of the unknown and the standard have the same shape, the Feather plot is a straight line parallel to the abscissa. When RaE, which has an unusually large percentage of low-energy electrons in its spectrum, is used as a stand-

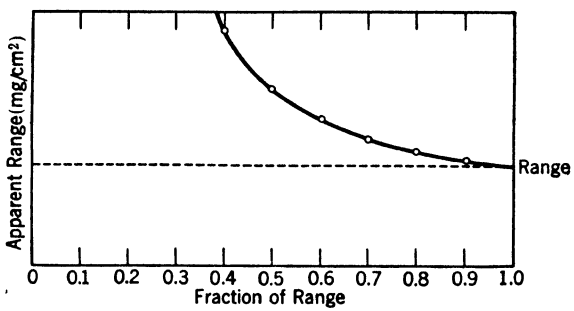


FIGURE VII–7. A typical Feather plot.

ard, the Feather plot often has a shape similar to that shown. The UX_2 spectrum is more nearly normal, as is also that of P^{32} which has been suggested as a standard. A more complicated comparison method has recently been proposed;[6] it uses as standards a number of absorption curves and takes into account the fact that the shapes of β spectra vary with the atomic number of the β emitter and with E_{max}.

When two β-spectral components are emitted from a sample it is usually very difficult to obtain a reliable end point for the softer component; extrapolation of the line representing the harder component to zero absorber thickness and subtraction of this extrapolated curve from the total absorption curve is the best that can be done. In this case too the Feather method gives much better results than does visual determination.

Absorption of Monoenergetic Electrons. When soft conversion electrons are emitted in addition to β particles, the absorption curve (on semilog paper) usually has an initial portion which is

[6] E. Bleuler and W. Zünti, "On the Absorption Method for the Determination of β and ... Energies," *Helv. Phys. Acta* 19, 375 (1946).

concave toward the origin (see figure VII–8*a*). The range of the conversion electrons cannot be obtained reliably from such a graph. If conversion electrons are emitted without accompanying β rays, for example following K capture or in an isomeric transition, their range can often be determined better from an extrapolation to zero activity on a linear plot, because the absorption

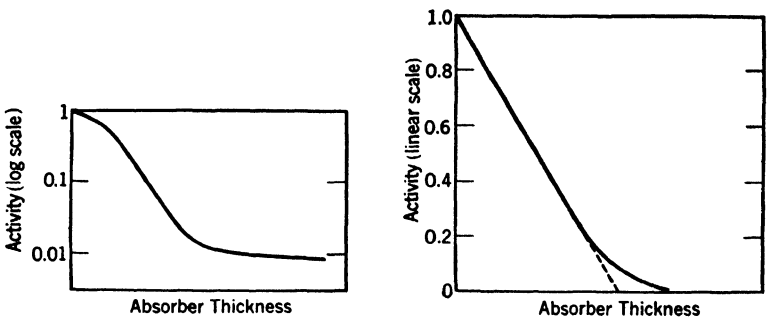


FIGURE VII–8. Typical absorption curves of conversion electrons.
(*a*) Semilog absorption curve of soft conversion electrons superimposed on a β spectrum.
(*b*) Linear absorption curve of pure conversion electrons.

of monoenergetic electrons turns out to be more nearly linear than exponential (see figure VII–8*b*). The only really good method for measuring energies of conversion electrons uses the electron spectrograph.

It should be noted that in plotting absorption curves the plotted absorber thickness must include not only the added absorbers but also the window or wall of the measuring instrument, the air between sample and instrument, and any material covering the sample. The sample itself should be thin compared to the half-thickness value for the radiation.

Range–energy Relations. Once the range of β particles or conversion electrons is known, a range–energy relation can be used to deduce the maximum energy.[7] Many empirical relations have

[7] Monoenergetic electrons of a given energy and β particles of the same maximum energy should, of course, have the same ranges, and at low energies this is verified experimentally. Above 0.5 Mev, apparent β-particle ranges drop off a few per cent compared to the observed ranges of corresponding monoenergetic electrons; this is probably due to the relatively small number of electrons with energies near the upper energy limit in a β-ray spectrum.

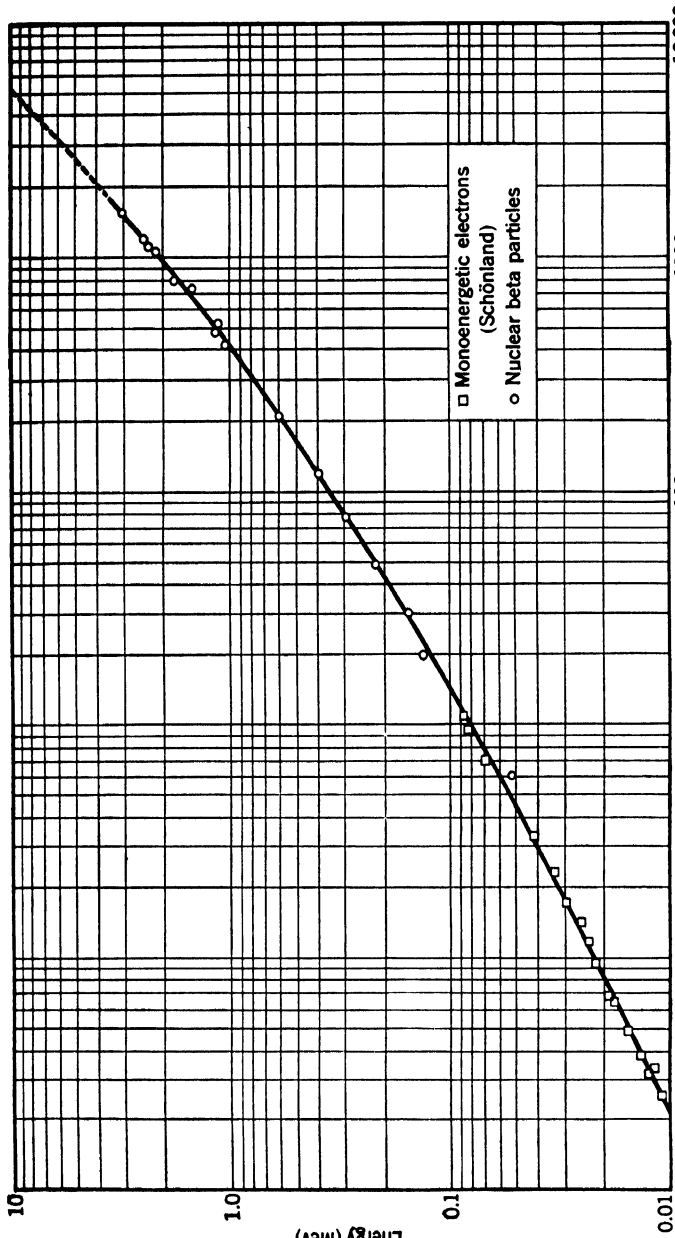


FIGURE VII–9. Range–energy relation for β particles and electrons. (Reproduced from a paper by L. E. Glendenin, *Nucleonics* **2**, no. 1, 16 (1948), by permission of the McGraw-Hill Publishing Co.)

been proposed. One given by Feather for energies above 0.6 Mev has been most widely used: $R = 0.543E - 0.160$, where E is the maximum β energy in million electron volts and R the range in aluminum in grams per square centimeter. Another relation proposed by C. D. Coryell and L. E. Glendenin for energies above 0.8 Mev appears to give a somewhat better fit at higher energies (2–3 Mev): $R = 0.542E - 0.133$. In the lower-energy region (below about 0.7 Mev) it is best to use a range–energy curve such as the one plotted in figure VII–9.

Back-scattering and Self-absorption. As already mentioned, scattering of electrons, both by nuclei and by electrons, is much more pronounced than scattering of heavy particles. A very significant fraction of the number of electrons falling on a piece of material may be reflected (that is, scattered through more than 90°) as a result of single and multiple scattering processes. The reflected intensity increases with increasing thickness of reflector until the thickness equals about one-fifth of the range of the electrons; further increase in thickness does not add to the reflected intensity. The amount of back-scattering increases also with atomic number of the reflector. Table VII–4 lists the rela-

TABLE VII–4 *

Reflector	Z of Reflector	Amount of Ionization
None		I_0
C	6	$1.17I_0$
Al	13	$1.30I_0$
S	16	$1.32I_0$
Cu	29	$1.45I_0$
Ag	47	$1.57I_0$
Au	79	$1.68I_0$
Pb	82	$1.70I_0$

* The data for this table were taken from G. Hevesy and F. A. Paneth, *A Manual of Radioactivity*, Oxford University Press, 1938, p. 46.

tive amounts of ionization produced in an electroscope by a given sample of RaE ($E_{max} = 1.17$ Mev) with various reflectors of effectively infinite thickness behind it. Numerical values will depend, of course, on the particular geometrical arrangement. It is clear that in the measurement of radioactive materials care must be taken to mount all samples whose activities are to be compared

on the same thickness of the same backing material; otherwise, cumbersome and uncertain corrections for back-scattering are necessary. It is sometimes convenient to increase measured β-ray activities by placing heavy reflectors (such as lead) immediately behind the samples.

A very complex effect compounded of absorption and back-scattering occurs in all but the thinnest β-particle sources. This so-called self-absorption effect, for a given sample thickness in milligrams per square centimeter, appears to decrease with increasing Z, perhaps about as $Z^{-\frac{1}{2}}$. Some other empirical facts about self-absorption will be given in chapter X, section E.

C. ELECTROMAGNETIC RADIATION

Processes Responsible for Energy Loss. The specific ionization caused by a γ ray is about $\frac{1}{100}$ of that caused by an electron of the same energy, at least for energies greater than 100 kev. The practical ranges of γ rays are very much greater than those of β particles. The ionization observed for γ rays is almost entirely secondary in nature as we shall see from a discussion of the three processes by which γ rays (and X rays) lose their energy. The average energy loss per ion pair formed is the same as for β rays, namely, 32.5 ev.

At low energies (and, therefore, of particular significance for characteristic X rays) the most important process is the photoelectric effect. In this process the electromagnetic quantum of energy $h\nu$ ejects a bound electron from an atom or molecule and imparts to it an energy $h\nu - E$, where E is the energy with which the electron was bound. The quantum of radiation completely disappears in this process, and momentum conservation is possible only because the remainder of the atom can receive some momentum.

In the energy region of characteristic X rays the probability for photoelectric absorption has sharp discontinuities at energies equal to the binding energies of the K, L, etc., electrons. For $h\nu$ greater than the K-binding energy the photoelectric absorption first falls off rapidly (about as $E_\gamma^{-\frac{7}{2}}$), then more slowly (eventually as E_γ^{-1}) with increasing energy. It is also approximately proportional to Z^5. Except in the heaviest elements photoelectric absorption is relatively unimportant for energies above 1 Mev.

The ionization produced by photoelectrons accounts largely for the ionization effect of low-energy photons. The photoelectric effect is frequently used to determine γ-ray energies. This is accomplished by measurements in an electron spectrograph of the energies of photoelectrons ejected from a thin foil, called the "radiator" or "converter," placed over the γ-active sample; an element of high atomic number such as gold is used as the radiator.

Instead of giving up its entire energy to a bound electron a photon may transfer only a part of its energy to an electron, which in this case may be either bound or free; the photon is not only degraded in energy but also deflected from its original path. This process is called the Compton effect or Compton scattering. The relation between energy loss and scattering angle can be derived from the conditions for conservation of momentum and energy. The Compton scattering per electron is independent of Z, and, therefore, the scattering coefficient per atom is proportional to Z. For energies in excess of 0.5 Mev it is also approximately [8] proportional to E_γ^{-1}. Thus Compton scattering falls off much more slowly with increasing energy than photoelectric absorption, at least at moderate energies (up to 1 or 2 Mev), and even in heavy elements it is the predominant process in the energy region from about 0.6 to 2.5 Mev. Photon energies can be determined from the upper energy limits of Compton electrons. For this purpose a radiator of relatively low Z, often copper, is used in the electron spectrograph, so that the Compton effect predominates over the photoelectric effect.

The third mechanism by which electromagnetic radiation can be absorbed is the pair-production process (discussed in chapter VI, section B). Pair production cannot occur when $E_\gamma < 1.02$ Mev. Above this energy the atomic cross section for pair production first increases slowly with increasing energy and above about 4 Mev becomes nearly proportional to $\log E_\gamma$. It is also proportional to Z^2. The energy dependence of pair production is satisfactorily predicted by a theory due to H. A. Bethe and W. Heitler. At high energies, where pair production is the predominant process, γ-ray energies can best be determined by measurements of the total energies of positron–electron pairs.

[8] A formula for the scattering coefficient which contains a very complicated function of E_γ has been derived from relativistic quantum mechanics by O. Klein and Y. Nishina.

Pair production is always followed by annihilation of the positron, usually with the simultaneous emission of two 0.51-Mev photons. The absorption of quanta by the pair-production process is, therefore, always complicated by the appearance of this low-energy secondary radiation.

The atomic cross sections for all three processes discussed increase with increasing Z, except for the photoelectric effect at very low energies. For this reason heavy elements, atom for atom, are much more effective absorbers for electromagnetic radiation than light elements, and lead is most commonly used as an absorber. Because photoelectric effect and Compton effect decrease and pair production increases with increasing energy, the total absorption in any one element has a minimum at some energy. For lead this minimum absorption, or maximum transparency, occurs at about 2.7 Mev; for copper at about 10 Mev; and for aluminum at about 22 Mev.

Determination of Photon Energies by Absorption. The only mechanism for absorption of quanta which gives rise to a true exponential absorption is the photoelectric effect. The production of degraded electromagnetic radiation in the Compton-

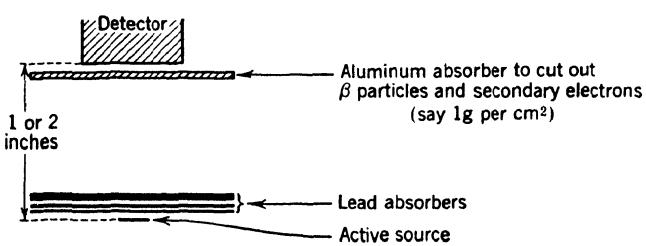


FIGURE VII–10. Recommended arrangement for γ-ray absorption measurements.

scattering and pair-production processes tends to distort the exponential absorption. However, if an appropriate experimental arrangement is used, these distortions can be minimized to such an extent that exponential absorption curves can be obtained in practice. One such arrangement is shown schematically in figure VII–10. Active source and absorbers are as far as practicable from the detector to prevent most of the scattered quanta and secondary electrons from falling on the detector. The additional absorber of low Z near the detector stops a large frac-

tion of the secondary electrons (as well as any β particles emitted by the source which otherwise might enter the detector when little or no lead absorber is used). In an ideal arrangement, source

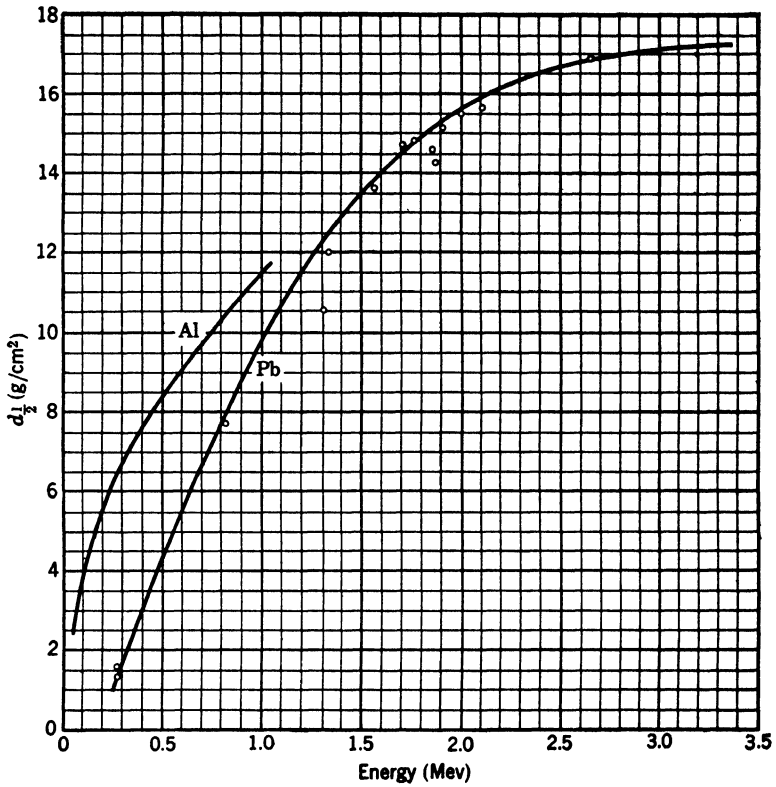


FIGURE VII–11. Half-thickness values in lead and aluminum for photons of various energies. (Reproduced from a paper by L. E. Glendenin, *Nucleonics* **2**, no. 1, 19 (1948), by permission of the McGraw-Hill Publishing Co.)

and detector would be far apart, with the lead absorbers midway between them.

Absorption curves for electromagnetic radiation are plotted on semilog paper. For a single energy a line results which is straight over a factor of 10 or 20 in intensity if the afore-mentioned experimental arrangement is used. When two components differing by at least a factor of two in energy are present, the absorption curve can often be resolved into two straight lines, in the

same manner as a decay curve is resolved. Resolution into more than two components with any precision is generally not possible.

For an exponential absorption law the intensity I_d measured through an absorber thickness d is given by $I_d = I_0 e^{-\mu d}$ where I_0 is the intensity without absorber and μ is called the absorption coefficient. The half-thickness $d_{1/2}$ is defined as the thickness which makes $I_d = \frac{1}{2}I_0$; $d_{1/2} = 0.693/\mu$. Absorber thicknesses are

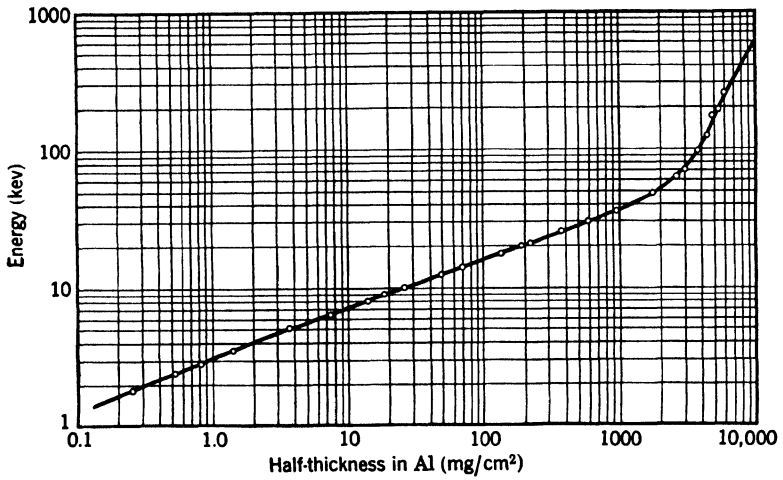


FIGURE VII–12. Half-thickness values in aluminum for low-energy photons. (Reproduced from a paper by L. E. Glendenin, *Nucleonics* **2**, no. 1, 21 (1948), by permission of the McGraw-Hill Publishing Co.)

more frequently given in terms of surface density (ρd, expressed in grams per square centimeter). Then $I_d = I_0 e^{-(\mu/\rho)\rho d}$; μ/ρ is called the mass absorption coefficient. In the energy region where the Compton effect predominates the mass absorption coefficient varies only slowly with Z (as in β-ray absorption), but in the low- and high-energy ranges it increases rapidly with increasing Z. This dependence of the absorption on Z is sometimes used to distinguish low-energy X rays from electrons.

The energy of γ or X rays is usually deduced from measured half-thickness values. Curves of half-thickness versus energy are reproduced in figures VII–11 and VII–12 for different energy regions and for lead and aluminum absorbers. It should be remembered that a given half-thickness may correspond to two different energies because of the minimum in the curve of absorp-

tion versus energy. For example, a half-thickness of 13.5 g per cm^2 in lead may correspond either to an energy of 1.5 Mev or to an energy of about 5.5 Mev. One can always eliminate this ambiguity by taking absorption curves in two different materials

D. NEUTRONS

Because neutrons carry no charge, their interaction with electrons is exceedingly small, and primary ionization by neutrons is a completely negligible effect. The interaction of neutrons with matter is confined to nuclear effects; these include elastic and inelastic scattering and nuclear reactions such as (n, γ), (n, p), (n, α), $(n, 2n)$, and fission. All these subjects have been discussed in chapter III, and here we shall merely indicate how each of these types of interaction may be applied to the detection and measurement of neutrons.

The recoil protons produced by fast neutrons in hydrogenous material are often used for the detection of such neutrons. About 7 protons leave a thick paraffin layer per 10^4 incident neutrons of 1 Mev energy, and for other energies the ratio of protons to neutrons is roughly proportional to neutron energy. The energy of the fastest recoil protons equals the neutron energy.

The ionization produced by protons or α particles created in n, p or n, α reactions can also be used for neutron detection. Ionization chambers or proportional counters (described in chapter VIII) may be lined with boron or filled with gaseous BF_3, and the particles from the B^{10} (n, α) Li^7 reaction may be detected. The separated isotope B^{10} is particularly effective. Fission fragments may be detected in an ionization chamber lined with fissionable material and exposed to a neutron source.

Neutron-capture reactions leading to radioactive products are frequently used for detection of neutrons by means of the induced activity. This technique is especially useful in the resonance region, where the number of neutrons of a particular energy can often be determined by the amount of resonance capture observed.

E. BIOLOGICAL RADIATION UNITS

The subject of interaction of radiations with matter is of great importance in the study of the biological effects of radiation. These effects are generally assumed to be almost entirely due to

ionization processes. Actually the disruption of molecules by recoiling atoms is undoubtedly also a factor, especially in the case of neutron irradiation, but the effects of such processes are not easily isolated for study.

In determining radiation effects on living organisms, whether from external radiation or from ingested or inhaled radioactive material, one has to take into consideration not only the total dosages of ionization produced in the organism but also such factors as the density of the ionization, the dosage rate, the localization of the effect, and the rates of administration and elimination of radioactive material.

The units which are used for biological radiation dosage are derived from the roentgen unit. One roentgen unit, or r unit, is "that quantity of X or γ radiation such that the associated corpuscular emission per 0.001293 g [9] of air produces, in air, ions carrying 1 esu of quantity of electricity of either sign." This means that 1 r produces 1.61×10^{12} ion pairs per g of air which corresponds to the absorption of 83.8 ergs of energy per g of air.

The r is a unit of the total quantity of ionization produced by γ or X rays, and dosage rates for these radiations are therefore expressed in terms of roentgens per unit time. The maximum allowable daily dose for humans exposed to X or γ radiation is usually taken (in the United States) as 0.1 r, or 100 mr. (It is believed by some that this value should be lowered to 50 mr.)

Because of its definition, the r unit should not be used for radiations other than X or γ rays. Another unit, the roentgen-equivalent-physical, rep, has therefore been proposed to express ionization in tissues caused by other radiations (electrons, protons, α particles, neutrons). One roentgen-equivalent-physical is the quantity of ionization produced when 83 ergs are dissipated by the radiation per gram of tissue.

The same amount of energy dissipation per gram of tissue may cause different amounts of biological damage when brought about by different radiations. For this reason still another unit, the roentgen-equivalent-man, rem, has been introduced. One rem unit corresponds to an energy dissipation in tissue which is biologically equivalent in man to 1 r of γ or X rays. For example, since the secondary ionization due to recoil protons from fast neutrons has been found to be about 5 times as effective biologically as the

[9] This is the weight of 1 cc of dry air at 0°C and 760 mm pressure.

same quantity of ionization due to γ rays, for fast neutrons 1 rem = 83/5 ergs per g of tissue = 0.2 rep. Therefore, the maximum allowable daily dose is 0.02 rep for fast neutrons. Similarly the maximum allowable daily doses for other types of radiation are estimated; these are usually taken as 0.1 rep for β radiation, 0.01 rep for α radiation, 0.02 rep for protons, and 0.05 rep for thermal neutrons.

EXERCISES

1. Show that the maximum velocity an electron can receive in an impact with an α particle of velocity v is approximately $2v$.

2. Estimate the ranges in air of (*a*) 10-Mev H^3 ions, (*b*) doubly charged 10-Mev He^3 ions. *Answer*: (*a*) 51.3 cm.

3. What is the velocity of a 20-Mev α particle (*a*) in a nonrelativistic approximation and (*b*) calculated with the relativistic correction?

4. An absorption curve of a sample emitting β and γ rays was taken, using a Lauritsen electroscope, with aluminum absorbers. The data obtained were:

Absorber Thickness (g/cm^2)	Activity (divisions/min)
0	5.8
0.070	3.5
0.130	2.2
0.200	1.3
0.300	0.60
0.400	0.28
0.500	0.12
0.600	0.11
0.700	0.11
0.800	0.10
1.00	0.10
2.00	0.092
4.00	0.080
7.00	0.065
10.00	0.053
14.00	0.040

(*a*) Find the maximum energy of the β spectrum (in million electron volts).

(*b*) Find the energy of the γ ray.

(*c*) What would be the absorption coefficient of that γ ray in lead?

5. What are the approximate initial charges of the following fission fragments: (*a*) Kr^{97} of 100-Mev initial energy, (*b*) Xe^{140} of 70-Mev initial energy? *Answer:* (*a*) 26.

6. What are the mean and the extrapolated ranges of α particles from Th^{232} in 15°C air? Estimate their mean ranges (in milligrams per square centimeter) in argon, uranium hexafluoride, and gold.

Answer: $\bar{R}$ in argon = 5.5 mg/cm^2 by Bragg's rule or 4.8 mg/cm^2 by equation VII–1.

7. In a certain measuring arrangement the β rays of 13.7-day Cs^{136} are absorbed as follows (γ-ray background has been subtracted).

Absorber Thickness (mg Al/cm^2)	Relative Intensity
0	100
12	47
27	17
41	7.3
53	2.7
72	0.30
85	0.037

For comparison the absorption of P^{32} β rays is measured with the same arrangement; the results are

Absorber Thickness (mg Al/cm^2)	Relative Intensity
0	250
160	100
245	50
360	20
420	10
480	5.0
530	2.30
580	0.95
620	0.45
680	0.15
725	0.05
780 (range)	0.0

Determine the maximum β energy of 13.7-day Cs^{136} by means of a Feather plot, using the P^{32} as a standard.

8. At 1.00 meter from 1.00 g radium (in equilibrium with its decay products and enclosed in 0.5 mm of platinum) the γ-ray dosage rate is 0.84 r per hr. What is the minimum safe working distance from a 1 mg radium source for an 8-hr day? *Answer:* 26 cm.

9. Estimate the linear absorption coefficient in air (at 15°C and 760 mm pressure) for 1-Mev γ rays.

10. (*a*) Estimate the r-dosage rate at a distance of 30 cm from a 2×10^3 rd source of 1.5-Mev γ rays. (*b*) What is the minimum thickness of lead that must be placed around this source to allow an experimenter to work at 30 cm from it for 2 hr every day?

11. At sea level the cosmic radiation produces about 2 ion pairs per sec per cm^3 of air. At higher altitudes the intensity depends on the latitude but for much of the United States is about 10 ion pairs sec^{-1} cm^{-3} at 10,000 feet and about 200 ion pairs sec^{-1} cm^{-3} at 40,000 feet above sea level. Estimate the radiation dosage received per 24 hr in r units (probably we should say in rep units) from this source at (*a*) sea level, (*b*) 10,000 feet, (*c*) 40,000 feet.

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VIII. INSTRUMENTS FOR RADIATION DETECTION AND MEASUREMENT

A. METHODS NOT BASED ON ION COLLECTION

In the preceding chapter we saw that the principal interactions of the radioactive radiations with matter result in the production of ions with a reduction in energy of the radiation of about 33 ev per ion pair formed. All methods for detection of radioactivity are based on interactions of the charged particles or electromagnetic rays with matter traversed. The uncharged neutron is detected only indirectly, through recoil protons (from fast neutrons) or through nuclear transmutations or induced radioactivities (from fast or slow neutrons). Neutrinos have no charge and do not seem to interact measurably with matter to produce either ions or recoil particles, and, therefore, are not detectable by any of these methods. (It may be presumed that neutrinos should be capable of causing nuclear transmutations, but the cross section for the process is estimated from the principle of microscopic reversibility to be less than 10^{-40} cm^2—being hard to emit they must be hard to absorb.)

Photographic Film. The historical method for the detection of radioactivity was the general blackening or fogging of photographic negatives, apparent on chemical development in the usual way. This method was soon supplanted by ionization measurements but has reappeared recently in the "film badge" for personnel exposure control (see section E) and in the γ raying (analogous to X raying) of castings and other heavy metal parts for hidden flaws. Also, in the radioautograph technique the distribution of a radioactive tracer (preferably an α or soft-β emitter) is revealed when a thin section, perhaps of biological material, is kept in contact with a photographic plate. An improvement over this "contact radioautograph" technique may eventually be achieved through perfection of an "enlargement" method, in which soft β rays from the specimen are accelerated electrostatically and then focused on the photographic plate as in an

electron microscope; however, the range of useful magnification is not likely to be very great.

Photographic emulsions exposed to densely ionizing radiations such as α rays, protons, and mesons, on development show blackened grains along the path of each particle; since the range of such rays is small, these tracks are observed under a microscope. The direction and range of each particle are indicated, and nuclear transmutations may be studied. The number of developed grains along a track is smaller by several orders of magnitude than the number of ion pairs produced. The technique is particularly useful for the recording of very rare events, such as are of interest in cosmic-ray studies.

Cloud Chamber. A pictorial representation of the paths of ionizing particles similar to the photographic track but capable of much finer detail is given by the cloud chamber (Wilson chamber). In this instrument the particle track through a gas is made visible by the condensation of water droplets on the ions produced. To accomplish this, an enclosed gas saturated with vapor (water, alcohol, and the like) is suddenly cooled by adiabatic expansion to produce supersaturation. Ordinarily a fog would be formed, but, if conditions are right and the gas is free of dust, scattered ions, and so on, the supersaturation is maintained except for local condensation along the track where the ions serve as condensation centers. The piston or diaphragm causing the expansion is operated in a cyclic way, and a small electrostatic gradient is provided to sweep out ions between expansions. There is usually an arrangement of lights, camera, and mirrors to make stereoscopic photographs of the fresh tracks at each expansion.

The α tracks appear as straight lines of dense fog droplets, with thousands of droplets per centimeter. The β tracks are much less dense, with discrete droplets visible, several per centimeter along the path. In both cases δ rays are visible, and scattering and straggling may be studied. Electron energies may be determined from track curvature in a magnetic field, and positrons are distinguished from negative electrons by the curvature if beginning and end of the tracks can be recognized. Cosmic-ray experts have learned to tell much about a particle's charge, mass, and energy from the relation of magnetic curvature and track density. Gamma rays in the cloud chamber produce scattered droplets and δ-ray tracks, with no obvious indication of a particular photon

path; the Compton-recoil electrons and photoelectric-conversion electrons may be studied.

Scintillation Counting. When α particles strike a prepared fluorescent screen of zinc sulfide, discrete flashes of light may be seen by the dark-accustomed eye. The counting of α rays by this scintillation method was of great value in the early studies of radioactivity. Although it is no longer used in this way, there is a modern adaptation of scintillation counting for β and γ rays. The rays produce light in a naphthalene or anthracene crystal (or polycrystalline mass); the light can produce photoelectrons from the first, photosensitive electrode of a photomultiplier tube such as the RCA 931A, 1P21, or 1P28, and the output pulse may be recorded. Other fluorescent materials recommended for this purpose include calcium tungstate and corundum (synthetic sapphire). Insofar as the substance is translucent, the effective detector thickness may be quite great (more than 1 cm in models already working); consequently, high γ-ray efficiencies are possible in principle and have been indicated in current reports. Also, because the time for light emission is probably very short, the instrument should be capable of high counting rates and short coincidence resolving times. A special type of electron multiplier tube (not yet commercially available) has been used for counting in a more direct way also: the rays are caused to fall on its first electrode, and the secondary electrons produced there initiate pulses.

Other Methods. A few other detection methods not based on ion collection have historical and occasional current interest. The heating effect of the radiations can be measured with precision for very active preparations to the extent that the radiations are stopped in the calorimeter; with α emitters this condition is readily met. One of the more curious (and certainly not practical) detection methods described involves a calibration of the error in weighing when a strong radium sample (presumably many curies) is placed under one pan of an analytical balance. (This should not be confused with the quite ordinary use of much smaller γ-active sources near balance cases to prevent the accumulation of disturbing static electric charges.) The partial precipitation of certain colloids by radioactive rays has been used as a simple indicator of radiation dosage.

B. SATURATION ION CURRENT COLLECTION

The Ionization Chamber; Relation of Current to Voltage and Ionization Intensity. Many common radiation detectors make use of the electric conductivity of a gas resulting from the ionization produced in it. This conductivity is somewhat analogous to the electric conductivity of solutions caused by the presence of electrolyte ions. In gas conduction as produced by radiation the ion current first increases with applied voltage (as in the electrolyte

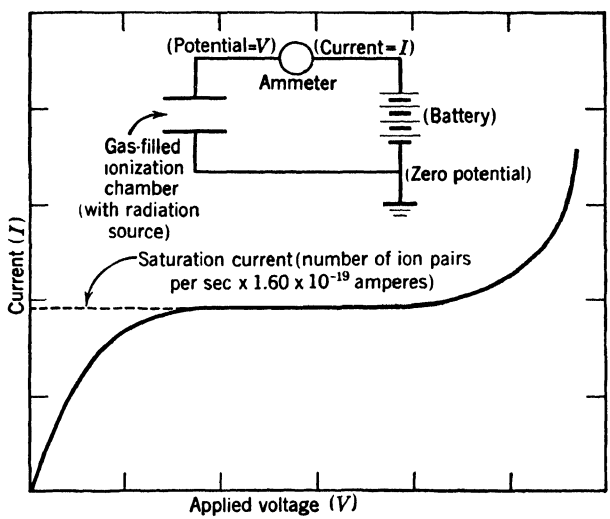


FIGURE VIII–1. Ionization current.

case); with increasing voltage the current eventually reaches a constant value which is a direct measure of the rate of production of charged ions in the gas volume. This constant value of the current is called the saturation current. A schematic representation of a gas volume and collecting electrodes, with potential difference V and meter to measure the ionization current I, is shown in figure VIII–1, along with a plot of I vs. V that might be obtained.

In the region of applied voltage below that necessary for the saturation current, recombination of positive and negative ions reduces the current collected. As the applied voltage is increased beyond the upper limit for saturation collection, the current

increases again, and, finally, the gap breaks down into a glowing discharge or arc, with a very sharp rise in the current. In the measurement of gas ionization it is obviously of some advantage to measure the saturation current: the current is easily interpreted in terms of the rate of gas ionization, and the measured current does not depend critically on the applied voltage or other like factors. The range of voltage over which the saturation current is obtained depends on the geometry of the electrodes and their spacing, the nature and pressure of the gas, and the general and local density and spatial distribution of the ionization produced in the gas. In air, for many practical cases, this region may be taken to extend from $\sim 10^2$ to $\sim 10^4$ volts per cm of distance between the electrodes.

We may classify detection systems (of the ion-collection type) according to whether saturation collection is employed or whether the multiplicative collection region is used. In the multiplicative region, where V is above the maximum value for saturation collection, the additional current is due to secondary ionization processes which result from the high velocities reached by the ions (particularly electrons) moving in the high field gradient. The use of this current amplification makes multiplicative collection methods inherently sensitive but unfortunately also inherently critical to many experimental variables.

Lauritsen-type Electroscope. We will call the gas-filled electrode systems designed for saturation collection ionization chambers. Saturation current instruments consist of the ionization chamber, in which ions produced are collected with as little recombination or multiplication as possible, and an electric system for measuring the very small currents obtained. The essential differences between the various instruments of this sort are in the nature of the current-measuring systems. In one common and relatively inexpensive instrument, the Lauritsen-type electroscope, a sensitive quartz-fiber electrometer measures the change in voltage produced on the fiber and its support by collection of the ionization charge. An external battery or rectifier is used to provide the initial voltage V (by means of a temporary connection to the fiber support); then, the fiber position is observed through a small telescope to measure ΔV as a function of time. For a collected charge q, the resulting $\Delta V = q/\mathrm{C}$, where C is the approximately constant capacitance of the fiber and electrode system.

(The order of magnitude of C may be guessed from the dimensions; the fiber and support arrangement are about 1 cm long, and C is of the order of 1 cm, which is the electrostatic unit of capacitance equal to about 10^{-12} farad or ~ 1 $\mu\mu$f.) The Lauritsen electroscope is simple and rugged and can be used to detect a β activity of about 2000 disintegrations per min (so arranged that about 50 per cent of the rays enter the ionization chamber). With the same arrangement samples up to about 1000 times this activity may be measured accurately. To set corresponding limits of usefulness for γ-ray measurements, account must be taken of the roughly 100-fold smaller specific ionization produced by these rays. Care must be used in measurement because the instrument is not strictly linear over different portions of the eyepiece scale and may be somewhat erratic in behavior when first charged. Most workers find it best to use only one chosen portion of the scale and to have the fiber charged for several hours before use.

D-c Amplifiers. Instruments of another type use ionization chambers with electronic d-c amplifiers. The ionization current I is caused to flow through a very high resistance R, and the volt-

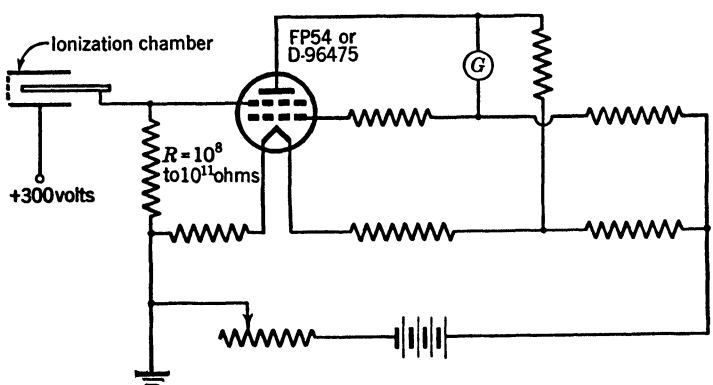


FIGURE VIII–2. Ionization chamber with balanced d-c amplifier.

age developed, $V = IR$, is applied to the control grid of a vacuum tube and measured in terms of the plate current of the tube by a galvanometer G. (See the schematic circuit in figure VIII–2.) To measure the smallest currents the vacuum tube must be chosen for low inherent grid current. A common amplifier has been one using the General Electric FP54 or Western Electric D-96475

vacuum tube; the Victoreen VX-41A is also used, especially in portable equipment. High stability of the circuit, particularly against variations in the battery voltage, is essential; balanced circuits are in use in which a single battery supplies plate, filament, and bias voltages, and through proper adjustment of the voltage-dividing networks the plate current is made insensitive to the battery voltage over a limited range, in the balanced condition. An instrument of this type with $R = 10^{11}$ ohms is easily sensitive to 1000 β disintegrations per min (with the same assumption as in the last paragraph that about 50 per cent of the rays enter the ionization chamber). With $R = \infty$ the time rate of drift of the plate current may be measured; by this means the sensitivity may be extended to about 200 disintegrations per min. Ordinarily a switch is provided so that R may be selected from values such as ∞, 10^{11}, 10^{10}, 10^{9}, and 10^{8} ohms. On this last position a sample of ~20,000,000 disintegrations per min may be measured in the 50-per cent geometry.

The vacuum tube, grid resistors R, selector switch, and so on, for the FP54-type instrument are enclosed in an evacuated or at least sealed and desiccated can to minimize stray leakage currents, and insulators of the highest quality are needed in the grid circuit. The ionization chamber may be connected through a screw-in fitting so that different chambers can be used as desired. A chamber containing air at atmospheric pressure and closed with an exceedingly thin aluminum leaf window is common for particles of low penetration; γ and X rays are detected with considerably improved sensitivity in a closed chamber filled to a pressure of 2 or 3 atmospheres with Freon (a chlorofluoromethane) or methyl bromide.

As an example we might estimate the ionization current I and the voltage drop IR for an ionization chamber under these assumptions: $R = 10^{11}$ ohms; the sample is an emitter of moderately energetic β rays with 1000 disintegrations per min; the geometry is such that 50 per cent of the β particles enter and spend an average 8-cm path length in the effective volume of an air-filled chamber. The number of ion pairs to be expected [1] is about $1000 \times 0.50 \times 80 \times 10 = 4 \times 10^{5}$ per min, or 6.7×10^{3} per sec. The current I will be the corresponding charge per second:

[1] The estimate of 10 ion pairs per mm over the 80-mm path is taken from the information on β-ray ionization in chapter VII.

$$I = 6.7 \times 10^3 \times 1.6 \times 10^{-19}$$

$$= 1.1 \times 10^{-15} \text{ coulomb per sec, or ampere.}$$

$$IR = 1.1 \times 10^{-15} \times 10^{11}$$

$$= 1.1 \times 10^{-4} \text{ volt, or 0.11 mv.}$$

If the use of the number of ion pairs rather than the total number of ions in this calculation is not entirely clear, remember that only half of the ions—those with the proper sign of charge—are collected at either electrode. (Also, although the current in the gas space consists of moving ions of both signs, for each ion pair formed neither ion traverses the whole path length between the electrodes, but rather the sum of the two ion paths is that distance.)

Vibrating-reed Electrometer. Even though special tubes and balanced circuits may be used with ionization chambers, d-c amplifiers are more susceptible to disturbance and drift and more difficult to arrange with several successive stages of amplification than those designed for amplifying alternating currents. A recent development is the use of a continuously vibrating reed which, through its oscillating electrostatic capacitance to a fixed electrode, converts the IR voltage to an approximately sinusoidal alternating potential; the a-c signal is then amplified in a highly stable audio-frequency amplifier. This instrument has a sensitivity comparable to the FP54 unit and can be unusually free from troublesome zero drift and external disturbances. The output signal level may be used to operate a continuously recording milliammeter to give a permanent record of the ionization-chamber current. The vibrating reeds are constructed with great care, and the entire set of equipment is rather expensive.

Linear Pulse Amplifier. An ionization chamber with directly connected a-c amplifier, as in the schematic representation of figure VIII–3, will, of course, give no response to any steady ionization current. A short burst of intense ionization, such as results from the passage of an α particle through the chamber, will give a sudden change of voltage on the first grid; this grid voltage will return to normal in a time of the order of RC, where C represents the distributed capacitance of the grid and collecting-

electrode system and R is the effective resistance to ground.[2] With sufficient amplification a large pulse will appear at the amplifier output terminal; the shape in time of this voltage pulse will depend on several factors, including the value of RC and the frequency-response characteristics of the amplifier. It is ordinarily desirable to have the height of the output pulse proportional to the amount of ionization produced by the particle in the

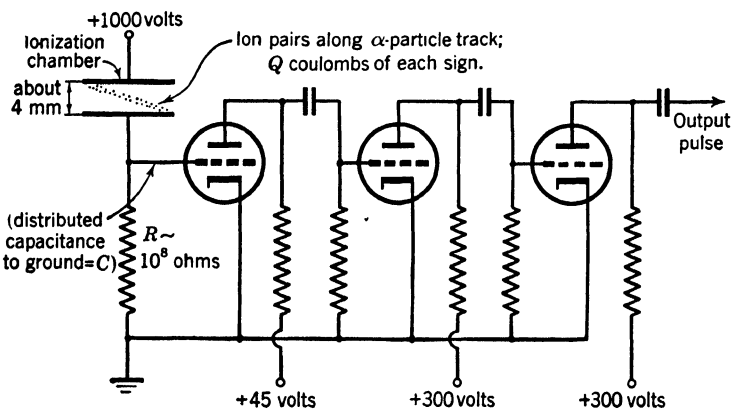


FIGURE VIII–3. Schematic representation of ionization chamber with linear pulse amplifier.

chamber; thus, the name linear amplifier or linear pulse amplifier is often applied to this instrument.

Since the instrument is used for counting single α particles we may estimate the voltage amplification factor (gain) needed. A fast α particle traveling 1 cm in the chamber would give in air about 25,000 ion pairs, and the collected ion charge, $q = \sim 25{,}000 \times 1.6 \times 10^{-19} = \sim 4 \times 10^{-15}$ coulomb; and guessing $C = \sim 10\ \mu\mu f$ we have then $V = q/C = \sim 4 \times 10^{-4}$ volt. If an output pulse of 100 volts is wanted (convenient for oscillographic observation and photographic recording) the required gain is $100/(4 \times 10^{-4}) = 2.5 \times 10^{5}$. Four amplifier stages might be used, each with gain of roughly 22.

[2] The charge in a capacitor of capacitance C short-circuited with a high resistance R will be dissipated exponentially; the half-time for the process is given by 0.693 RC; RC is known as the time constant of the circuit and is the time required for the charge to be reduced to $1/e$ of its value.

A practical ionization chamber may have a background rate of the order of 0.1 to 1 α per min; the lower limit of sample strength easily detectable we may take as $\sim$1 α disintegration per min (with 50 per cent geometry). With appropriate amplifier and recording equipment the maximum usable rate is limited by the duration ($\sim$RC) of the voltage pulse, because, if the average rate of arrival of pulses is such that there is an appreciable chance of one following another within the time RC, appreciable counting error results. With R = 10^8 ohms, RC = $\sim 10^{-3}$ sec, and a few thousand counts per minute would be the useful upper limit. Of course R is easily made smaller, but the full voltage q/C is achieved only if RC is long compared to the time of collection of ions in the chamber. The velocity v of ions in air under a voltage gradient E volts per centimeter is about (perhaps 1.5 times) E centimeters per second; with 1000 volts applied to a 0.4 cm chamber, $v = \sim 4000$ cm per sec, and the ion collection time is $\sim 0.4/4000 = \sim 10^{-4}$ sec. In practice RC is usually made somewhat longer than this time; to waste much of the voltage pulse is not advisable because with higher amplifier gains much trouble would be caused by tube "noise" and by microphonic effects (sensitivity of the chamber and amplifier to vibration). However, in a closed ionization chamber filled with pure argon or nitrogen the negative ions will be principally free electrons, which may be collected very much faster than heavy gas ions; then with an appropriate amplifier much higher counting rates may be used.

If an ionization chamber is large enough to contain the entire range of the most energetic α rays, then the ionization produced will be an accurate measure of the α-particle energy, which is characteristic of the particular α emitter. Instruments which record in many separate channels the counting rates of α particles of various energies have been constructed, and they are very useful for the analysis of complex mixtures of the heavy α-active nuclides. Linear amplifiers may be used to count fissions; because fission fragments have roughly ten times the specific ionization of α particles they are easily distinguished.

It has recently been shown that certain crystals, including selected diamonds, and AgCl crystals at low temperatures, when fitted with electrodes and connected to a source of high voltage and to a fast amplifier, give pulses under the action of radioactive rays. The crystal presumably acts as an ionization chamber,

with electrons and vacant electron sites moving through the crystal in the potential gradient. That this detector can respond to β and γ rays in addition to α particles, is due to the much greater specific ionization produced in a solid crystal than in a gas. The special requirements on the nature of the crystal may be related to the random production of free electrons by thermal processes and to the existence of many electron traps in ordinary imperfect crystals. This crystal counting technique may be developed to have a favorable sensitivity for γ rays; it is also of interest in coincidence counting because of its short characteristic time delay.

C. MULTIPLICATIVE ION COLLECTION

In the preceding section we discussed detection techniques utilizing saturation collection of ions in ionization chambers. The arrangements of electrodes for the multiplicative collection of ions as described in the following we will call counters. Usually the currents collected in counters, even from as little as one initial ion pair, may be large enough so that no very sensitive amplifiers or extremely low-capacitance or extremely high-resistance circuits are required. Practical difficulties are found rather in the construction and operation of the counters themselves. To obtain multiplicative collection of the type desired one might at first think of simply increasing the voltage applied to an ordinary parallel-plate ionization chamber. This is ordinarily [3] not practical, for several reasons which are suggested by the following discussion.

Voltage Gradients and Electrode Shapes. Figure VIII-4 shows the electrostatic lines of force between parallel-plate electrodes. The density of the lines of force is a measure of the voltage gradient E in any region. The voltage gradient is the same everywhere between the plates, except for effects near the edge, and is given by the applied voltage difference ΔV divided by the plate separation. Lines of force converge on a curved electrode such as a sphere or wire or point, and indeed unless the parallel plates are

[3] This qualification is made because some success has been obtained with parallel-plate counters. They are poor counters in most respects but do seem to have shorter time lags between ionization and counting action, and this characteristic can be very valuable in some coincidence counting work.

perfectly smooth high local gradients will exist at surface irregularities.

In counters one electrode is usually a cylinder, the other an axial wire. Figure VIII–5 shows a cross-sectional view, with the wire radius exaggerated; the lines of force are sketched in. It is readily seen that the density of these lines is inversely proportional to the radial distance r; that is, $E = k/r$. Now E is by definition

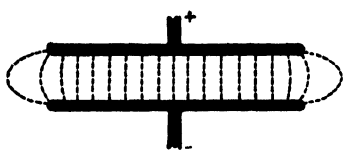


FIGURE VIII–4. Electrostatic lines of force between parallel-plate electrodes.

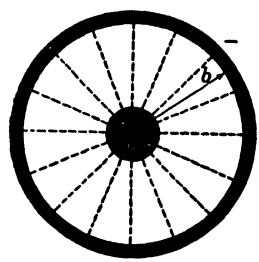


FIGURE VIII–5. Electrostatic lines of force between coaxial cylindrical electrodes.

dV/dr, and we may represent the voltage difference between the electrodes of radii a and b:

$$\Delta V = \int_{r=a}^{r=b} dV = \int_a^b E\,dr = \int_a^b \frac{k}{r}\,dr = k \ln\left(\frac{b}{a}\right).$$

In a practical case we might have $b = 1$ cm, $a = 4 \times 10^{-3}$ cm, $\Delta V = 1000$ volts. Then:

$$1000 = k \ln\left(\frac{1}{4 \times 10^{-3}}\right) = 5.5k; \qquad k = 180.$$

The voltage gradients at wall and wire are

$$E_b = 180 \text{ volts per cm};$$

$$E_a = \frac{180}{4 \times 10^{-3}} = 4.5 \times 10^4 \text{ volts per cm.}$$

The gradient at the wire and for a small space around it is above the maximum value for saturation collection (say $\sim 10^3$ volts

per cm in a practical counter gas). The voltage difference ΔV is always applied with the wall (cathode) negative with respect to the wire (anode); in this way free electrons and heavy negative ions move to the wire.

The Geiger–Müller (G–M) Counter. If an electrode system like that just described is filled with a suitable gas such as 90 per cent argon and 10 per cent ethyl alcohol (total pressure about 10 cm) and connected to a high-gain amplifier, and the pulses produced are studied as a function of the applied voltage with different types of ionizing particles, the following voltage regions are observed:

1. At relatively low voltages (of the order of 100 volts) there is no multiplication of the ionization current; the system operates as an ordinary ionization chamber, and only the pulses produced by α particles are seen and these only at very high amplification.

2. At moderate voltages (several hundred volts) there is amplification (10- to 100-fold or more) of the pulse heights; α-particle pulses are seen with moderate external amplification, and even β particles are detectable with high external amplification. The pulse height at fixed voltage is approximately proportional to the amount of ionization caused by the particle; when operated in this region the device is known as a proportional counter. (At least one commercial instrument uses this principle to count β particles.)

3. As the voltage is increased further (to about 1000 volts) the pulse heights increase, and their dependence on the initial ionization intensity disappears; this is the beginning of the Geiger counting region, where a single ion pair or the intense ionization from an α particle produces the same large pulse (perhaps about 10 volts on the counter wire and requiring little or no amplification for observation or recording).

To investigate the extent of the Geiger counting region we often arrange the counter with a fixed source of radiation and determine the counting rate produced as a function of the applied voltage. Figure VIII–6 shows this curve for a good counter. The region BC in which the rate is very nearly independent of the voltage is the "plateau" region; its length may be as much as several hundred volts; the voltage is always set in this region for counting. At voltages below B pulses exist, but are not uniform in size, and only some trip the recording circuits. We call A the starting

voltage, where the largest of these pulses just begin to be counted. The voltage difference between A and B obviously will depend on the circuit characteristics; if it exceeds 30 or 40 volts for a counter connected directly to the amplifier, a reduction of the stray capacitance to ground of the anode wire connections or an increase in the amplifier gain will probably lengthen the plateau (move B nearer to A).

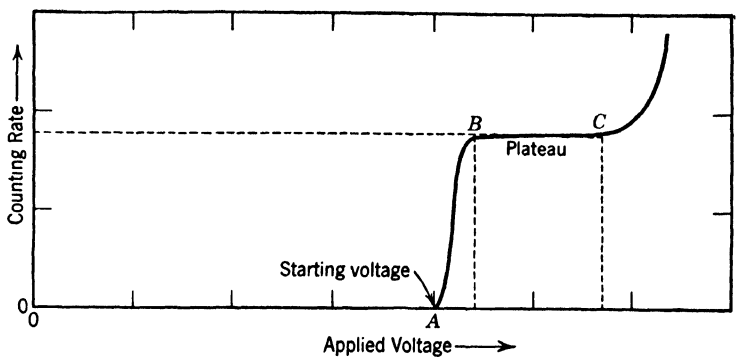


FIGURE VIII–6. Plateau curve for a good Geiger–Müller counter.

To understand the rise in counting rate beyond C we must consider (very briefly) some of the things that happen when the counting action occurs:

(*a*) The negative ion of the original ion pair moves toward the wire, traveling most or all the way as a free electron and thus at high speed, and it very quickly reaches the region of pronounced multiplicative processes.

(*b*) The intense region of secondary, tertiary, etc., ions and electrons formed in the high field gradient immediately around the wire spreads along the wire over all its effective length; spreading occurs at least partly through the photoelectric ionization of the gas by photons of high absorption coefficient (short mean path). There may also be some effect of photoelectrons from the cathode wall.

(*c*) The negative ions formed, mostly free electrons, very quickly reach the wire, and the intensely ionized region is left as a sheath of positive ions surrounding the wire. The effect of this positive charge is to reduce the voltage gradient below the value necessary for ion multiplication. All this has occurred in less than about

0.5 microseconds (0.5 μ sec), and now the counting action is complete except that the counter is left insensitive and must recover before another event can be counted.

(*d*) Recovery is effected through migration of the positive gas ions away from the wire. From the rough formula already given for ion mobilities, correcting for a roughly linear dependence of velocity on reciprocal pressure and taking account of the variable voltage gradient, we can estimate that migration of an ion from wire to wall will require $\sim$200 μ sec, and this is about the dead time found experimentally.

(*e*) When positive ions reach the cathode secondary electrons might be emitted from the surface; this would produce a new counter discharge just about 200 μ sec after the first, and quite independent of the source of radiation that the counter is intended to measure. Double, triple, and other multiple pulses with about this time spacing are observed with counters operating above the upper voltage limit of the plateau.

The various recipes for counter construction contain provisions designed to repress the emission of secondary electrons from the cathode. The argon–alcohol filling mixture seems to be effective because the positive ions are by electron transfers all converted to alcohol ions while moving to the cathode, and the polyatomic alcohol ions may dissipate energy by predissociation and so reduce enormously the probability of secondary-electron emission. Also the alcohol may serve to quench metastable states of the argon atoms. It is significant that the alcohol is considerably consumed after 10^8 or 10^9 counts and that a polyatomic filling gas, tetramethyl lead, requires no additive such as alcohol. The special cathode treatments recommended by some are probably related to changes in surface work function. The fact that various counter recipes differ widely probably means that the nature of the counter discharge action may be quantitatively different in different styles of counter tubes. The references at the end of this chapter will supply details on a number of construction techniques. Our feeling is that, although it is easy to make a useful counter, it is difficult to make one that does not sacrifice at least one of the following "ideal" features, chosen for their general usefulness and with the understanding that any given feature may readily be had at the expense of others:

1. The counter should operate without quench circuit, with a series resistor of ~0.25 megohm.

2. The plateau should be at least 50 volts long, preferably longer, and have a slope of not more than about 3 per cent per hundred volts.

3. The counter should be indefinitely stable against aging, though it may require refilling after the usual 10^8 counts, or thereabouts.

4. For the measurement of soft β particles a window not more than about 3 mg/cm^2 in thickness and with an area of several square centimeters should be provided close to the sensitive region.

5. The counter should be free of the troublesome hysteresis effects, in which counting rates and plateau curves may be different by a few per cent before and after the counting of active samples.

6. The dead time should not be more than a few hundred microseconds.

As suggested in item 4 provision for the effective introduction of low-energy β rays into the closed counter gas volume offers some problems. The window specified can be made of mica without a supporting grid and is thin enough to be usable with serious though not prohibitive loss of sensitivity for C^{14} and S^{35}, but not H^3. For very soft rays, there are several counter modifications that may be used:

1. With helium-plus-alcohol-vapor filling, the counters can be made to work at atmospheric pressure and used with very thin windows or even no window at all. Other gases can be used similarly.

2. In the screen wall counter the cathode is an open screen, and the sample is placed between it and an outer tube which is the counter gas container; with the sample on the inner surface of a cylindrical holder, it can be moved into counting position and moved away for background determination, by gravity, without opening the counter seal.

3. The counter may be made as part of a larger gas space which encloses a rotating wheel with positions for assorted samples or absorbers, and many measurements can be made before the system must be opened.

4. If the sample can be converted into gaseous form it may be introduced directly in the filling mixture; because strange gases

may upset the counter properties this is a tricky technique. However, with ionization chambers rather than counters it is a more useful method. In both cases considerable precautions are necessary to minimize errors caused by adsorbed radioactive gas.

Counter Quench Circuits. Before self-quenching counters were made it was common procedure to avoid spurious counts from double, triple, etc., pulses by connecting the counter with a very high series resistor (R = 10^8 to 10^9 ohms). See figure VIII–7. With the discharge a large negative voltage appears on the anode wire and leaks away with a time constant RC = $\sim 10^{-3}$ sec = $\sim$1000 μ sec. In this way, the voltage across the tube may be kept below the starting voltage until after the positive ion sheath

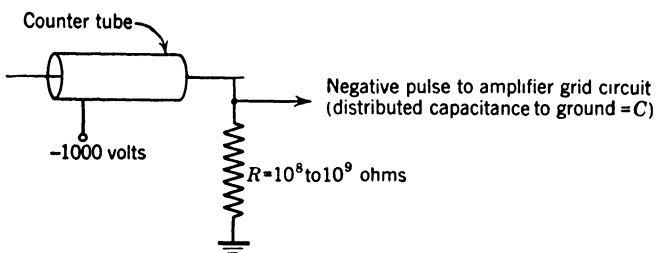


FIGURE VIII–7. Resistor-quenched counter circuit. With self-quenching counter tube the value of R is reduced to about 10^5 to 10^6 ohms.

is discharged. Use of this resistor quench seriously reduces the maximum useful counting rate. A number of electronic quenching circuits have been devised that are faster or more effective or both. Essentially these circuits use the amplification factor of a vacuum tube to provide a more positive and better-timed voltage reduction pulse applied to the wire or wall of the counter tube.

Practical Counting Instruments. In addition to the G–M counter tube and the optional quenching circuit, some auxiliary circuits are always required. Today excellent complete circuits are available from a number of manufacturers. The high-voltage supply ($\sim$1000 volts) might be a bank of batteries but is usually a combination of transformer, rectifier, and filter. Regulation of the output voltage is essential; a good commercial unit should provide a stabilized high voltage which will vary not more than 1 or 2 volts ($\sim$0.1 per cent) for a 10-volt ($\sim$10 per cent) change in line voltage. In almost all sets scaling circuits are used to reduce the rate for easier recording; with few exceptions these employ multi-

vibrator [4] scaling pairs, each pair reducing the rate by a factor of 2; common scaling factors include $2^3 = 8$; $2^6 = 64$; $2^8 = 256$. The scaled impulses are recorded mechanically, with electric power supplied by a suitable output circuit. Although with a fast mechanical recorder a scaling factor of about 8 can be adequate for most purposes, the present trend is toward higher scaling factors with slow mechanical recorders; the costs of register plus scaler are about the same in both cases.

There are some modifications of the basic circuit arrangements useful with Geiger counters and other types of counting instruments. In one design the time required for a fixed number of input pulses is automatically measured; this unit has no mechanical register, but rather a very high scaling factor, and the first output pulse turns off the counter and stops an electric timer. In other variations of the same principle the time for each output pulse is printed by a triggered time clock, or the output pulses are recorded by a recording galvanometer on paper tape moving at a constant known speed; these modifications are particularly useful for determinations of decay curves. In the counting rate meter the pulses are integrated electrically, and their average rate of arrival (averaged over a suitably long time interval) is indicated directly by a meter deflection.

The limit of sensitivity of a G–M counter is set by the background counting rate. Even in a laboratory not contaminated by radiochemical work small amounts of activity are present as impurities in construction materials. Also the air contains an appreciable and variable amount of radon and thoron and their decay products. It has been estimated that in free air at the earth's surface most of the ionization is from these two causes, with the cosmic radiation contributing a smaller part. However, because the counter is itself closed, and enclosed in a building, it is not accessible to most of the radioactive α, β, and even γ radiation, and the cosmic-ray effect is the most significant. A counter with a radius of 1 cm and length 10 cm may have a background rate

[4] In this application both tubes of the basic multivibrator pair (square wave-form oscillator) are biased to prevent oscillation; each incoming pulse triggers the pair through a half-cycle; completion of each cycle provides an output pulse to trigger the next pair. One of the most dependable circuits is that employing direct rather than capacitor coupling, devised by W. A. Higinbotham.

of about 50 counts per min; this may be reduced to about 25 counts per min by the usual lead shield of a few centimeters thickness. We may take about 10 β disintegrations per min as the minimum sample strength easily detected by a counter (with the 50 per cent geometry estimate as before). Most counter sets are not dependable at counting rates greater than that from a sample of about 3000 times this activity placed in the same position; however, with rather special precautions some workers use counters up to rates higher by another order of magnitude. For γ rays samples must have roughly 100-fold higher disintegration rates to give comparable counting rates.

D. AUXILIARY INSTRUMENTS

Electron Spectrograph; Coincidence Spectrometer. A number of other special instruments, designed more for the study of radiations than for intensity measurements, are likely to be found in laboratories working in many phases of radiochemistry. The β-ray spectrograph, or electron spectrograph, permits a much more accurate study of β-ray spectra than is possible by absorption measurements. In the various forms of this instrument the β particles are deflected by a magnetic (or possibly electrostatic) field, with some provision for angular focusing, and in this way are resolved according to energy. They may be recorded by a photographic plate or by a counter; in the latter case the instrument is called a β-ray spectrometer. Internal-conversion electrons are best studied in the same instrument, and the energies of γ rays and X rays may be deduced from measured energies of photoelectrons (external-conversion electrons). In studies of complex β spectra for the elucidation of disintegration schemes a very valuable technique is the coincidence spectrometer, in which a spectrum of β rays coincident with characteristic γ rays is distinguished by coincidence counting.

Curved-crystal Spectrograph. An instrument for the precision measurement of X rays and low-energy γ rays is the curved-crystal spectrograph. This is analogous to an optical spectrograph of the grating type, with the atomic planes of a bent crystal replacing the ruled lines of the curved grating. The detector may be a photographic plate, or a counter in the curved-crystal spectrometer.

E. HEALTH PHYSICS INSTRUMENTS

By the term health physics instruments we refer to detection and measuring instruments designed for the monitoring of personnel radiation exposures, and for the surveying of laboratories, equipment, clothing, hands, and the like, for biologically harmful radioactive contaminations. Very many types of such instruments have been made and used, especially within the wartime Manhattan Project laboratories and the Atomic Energy Commission laboratories and affiliated installations. Those which are generally available commercially may be divided into a few categories, and are all derived from the instrumentation principles already discussed in this chapter.

Pocket Ion Chambers; Film Badges. Perhaps the most widely used radiation monitor is the pocket ionization chamber. This is an ordinary ionization chamber in most respects, made small enough to be worn clipped in the pocket like a fountain pen. The charging potential is applied through a temporary connection, and at the end of the day or end of exposure the residual charge is read on an electrometer. A pocket ionization chamber with a built-in electrometer and scale is more convenient, but also more expensive; this style of chamber is initially charged on an external device and then may be read directly at any time without auxiliary apparatus and without effect on the indication.

These pocket meters are calibrated in roentgen units, with full scale corresponding most commonly to 0.1 or 0.2 r, so that they easily detect general radiation dosage below tolerance levels. They may not give a measure of local exposure (say, of the hands, while other parts of the body are shielded by a lead screen) and, of course, are not sensitive to soft radiations that do not penetrate the chamber wall. These limitations are also found in the photographic-film badge, which may be worn for determination of exposures integrated over longer times—usually over several days or weeks.

Portable Counters and D-c Amplifiers. More sensitive detection instruments are used to determine the rate at which exposure is being received in a given radiation field. These may be larger ionization chambers with compact d-c amplifiers operated from self-contained batteries, so that a readily portable survey meter weighing perhaps 10 pounds is achieved. Models of this type

ordinarily have several calibrated scale ranges, from about 0–20 mr per hr to about 0–3000 mr per hr. Battery-operated portable Geiger-counter sets of about the same size and weight are available and can be used for the same purpose. They are usually arranged as counting-rate meters, with full-scale readings calibrated at about 0.2 to about 20 mr per hr. Although the counter type of meter is much more sensitive than the ionization chamber, the latter is usually sensitive enough and may be expected to give a response more nearly proportional to the biological effects of the radiation. Both types are ordinarily provided with a movable shield to permit a distinction between hard and soft radiations. These two types of instruments are also useful for surveying the laboratory and its apparatus for radioactive contamination. The G–M counter instrument with its higher sensitivity, especially when used with earphones so that each count may be heard, is more convenient in rapid surveys for small amounts of activity but in its usual form is not useful for very soft β rays such as those from C^{14}. The ionizaiton-chamber instrument is more easily fitted with a window than enough for this purpose (not more than a few milligrams per squire centimeter), and some available models have very thin windows (or simply open screens) that will pass even α particles.

Other Procedures. A number of other more specialized instruments have been devised. Geiger counters and atmospheric-pressure proportional counters may be arranged particularly to detect β and α contaminations on the hands. The monitoring of air-borne contamination requires special instruments, and may be particularly important in laboratories handling long-lived α activities. One method for air-borne dusts is to filter a large volume of the air and to assay the activity left on the filter paper with a standard counter or linear-amplifier instrument. (The radon decay products ordinarily present in air can be detected in this way.) A very simple and widely applicable semiquantitative method of contamination monitoring which requires no special instrumentation is worth mentioning here; this is the so-called "swipe" method. A small piece of clean filter paper of a standard size is wiped over a roughly uniform path length on the suspected desk top, floor, wall, laboratory ware, or almost anywhere, and then measured for α, β, or γ activity on a standard instrument. Even air-borne contamination may be checked in a rough way by

swipe samples of accumulated dust from an electric-light fixture or some such place which is exposed only to contamination from the air.

EXERCISES

1. Estimate roughly the voltage (IR) applied to the grid of a d-c amplifier tube; use these assumptions: R = 10^{11} ohms; the sample emits 1-Mev γ rays at the rate of 10^6 per min; the geometry is such that 30 per cent of the γ's spend an average 8-cm path length in the ionization chamber which is filled with CF_2Cl_2 at 2 atmospheres total pressure.

2. (*a*) What is the thickest mica window that will pass some β particles from C^{14}? Give the answer in milligrams per square centimeter and in inches of thickness.

(*b*) How thin must a window be to pass some of the H^3 β particles?

(*c*) What is the energy of the softest β ray that could enter a counter tube with a 0.10-mm Pyrex-glass wall?

3. A β particle of 2 Mev enters a G–M counter and spends a 1.0-cm path length in the gas, which is $Pb(CH_3)_4$ at a pressure of 1.0 cm. What is the (average) number of ion pairs that should be expected to result from ionization in the gas? *Answer:* 5

4. Calculate the time required for a positive ion to move from the wire to the wall of a Geiger counter; take 0.005 inch for the wire diameter, 1⅛ inch for the cathode diameter, 1000 volts as the applied voltage, 10 cm as the gas pressure, and 1.5 cm per sec for the mobility of the ion at 1 volt per cm gradient and at 76 cm pressure. *Answer:* 490 μs

5. A Lauritsen electroscope is to be used for work with Cl^{38}. Its fiber system has a capacity of 0.3 esu. Its chamber has a diameter of 5 cm. In order to get a sufficiently accurate reading, its discharge rate must be at least 0.10 volt per min (corresponding to about 0.1 division per min). Making appropriate assumptions, estimate the minimum sample strength that should be used.

6. What type of instrument would you use for each of the following: (*a*) detection of 10^{-5} rd of H^3; (*b*) detection of 10^{-3} rd of Mn^{56}; (*c*) detection of 10^{-6} rd of At^{211}; (*d*) following the decay of a sample of Cu^{64} (initially 0.3 rd) over a period of 8 days. Briefly state the reason for each choice.

7. The radiations emitted by a certain radioactive species were studied in a β-ray spectrometer. The β^- spectrum was resolved into two components of 0.61 $\pm$ 0.01 Mev and 1.438 $\pm$ 0.007 Mev maximum energies. The higher-energy component was about four times as abundant as the lower-energy one. When γ rays were allowed to strike a thin silver radiator

placed in the source position of the spectrometer, the following five photoelectron energies were measured:

Energy in Mev	Intensity
0.216 ± 0.002	Strong
0.237 ± 0.002	Weak
0.801 ± 0.003	Weak
0.823 ± 0.003	Very weak
1.046 ± 0.005	Very weak

The K and L binding energies in silver are 25 and 4 kev, respectively. Draw a plausible decay scheme for the radioactive species under investigation.

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IX. STATISTICAL CONSIDERATIONS IN RADIOACTIVITY MEASUREMENTS

A. RANDOM PHENOMENA

The occurrence of nuclear disintegrations is a random phenomenon subject to established methods of statistical analysis. We must study in some detail the application of these methods and the nature of the statistical laws. First consider the set of data actually obtained with a Geiger counter measuring a "steady" source, as given in table IX–1. The number of counts recorded per minute (the counting rate) is clearly not uniform. Which minute gave the most accurate result? The best thing we can do is to compute the arithmetic mean (the average value) and consider this to represent the proper counting rate.

Average Value. If the determinations, minute by minute, are denoted by x_1, x_2, $\cdots$ x_i for the 1st, 2d, $\cdots$ ith minute, then the arithmetic mean value $\bar{x}$ is, by definition,

$$\bar{x} = \frac{1}{N_0} \sum_{i=1}^{i=N_0} x_i,$$

where N_0 is the number of values of x to be averaged. For the counting rates in the table $\bar{x} = 990/10 = 99.0$.

Standard Deviation. After the experiment which gave these data we might have repeated the measurements and so obtained another average rate. The best figure would then have been the average of all the results. Knowing merely the average value we do not know anything about the statistical dependability of the data from which the average was computed, that is, the degree of agreement between the individual results. We want to define a quantitative measure of the closeness of agreement. Consider for a moment the deviation Δ_i of each number x_i, defined as the difference between x_i and $\bar{x}$: $\Delta_i = x_i - \bar{x}$. These deviations are tabulated in the third column of the table. The average value of Δ_i cannot be taken as a measure of internal agreement because it is just zero:

$$\frac{1}{N_0}\sum_{i=1}^{i=N_0}\Delta_i = \frac{1}{N_0}\sum_{1}^{N_0} x_i - \frac{1}{N_0}\sum_{1}^{N_0}\bar{x} = \bar{x} - \bar{x} = 0.$$

The average of the squares of the individual deviations, called the dispersion and denoted by σ_x^2, gives a useful measure of the degree of agreement among the results.

$$\sigma_x^2 = \frac{1}{N_0}\sum_{1}^{N_0}\Delta_i^2 = \frac{1}{N_0}\Sigma(x_i - \bar{x})^2.$$

The standard deviation σ_x, which is just the square root of the dispersion, is commonly used. For the data of table IX–1 we

TABLE IX–1

Minute	Counts	Δ_i	Δ_i^2
1	89	−10	100
2	120	+21	441
3	94	−5	25
4	110	+11	121
5	105	+6	36
6	108	+9	81
7	85	−14	196
8	83	−16	256
9	101	+2	4
10	95	−4	16
Totals	990	0	1276

compute $\sigma_x^2 = 1276/10 = 127.6$; $\sigma_x = 11.3$. A useful relation that may be derived is: $\sigma_x^2 = \overline{x^2} - \bar{x}^2$; that is, the dispersion is given by the difference between the average of the squares of the x values and the square of the average value.

B. PROBABILITY AND THE COMPOUNDING OF PROBABILITIES

The ideas and definitions just presented may be applied, with varying degrees of usefulness, to any set of data, whether or not strictly random phenomena are involved. Before proceeding further, we must consider very carefully the concept of probability. As illustrations we will investigate the answers to questions such as:

(*a*) What is the probability that a card drawn from a deck be an ace?

(*b*) If a coin is flipped twice, what is the probability of it falling "heads up" both times?

(*c*) Given a sample of a radioactive material what is the probability that exactly 100 disintegrations occur during the next minute? We shall define probability in this way: Given a set of N_0 objects (or events, or results, etc.) containing n_1 objects of the 1st kind, n_2 objects of the 2d kind, and n_i objects of the ith kind; the probability p_i that an object specified only as belonging to the set is of the ith kind is given by: $p_i = n_i/N_0$. By applying this definition we find that the probability that one card drawn from a full deck be an ace is just 4/52.

We may now rewrite the definition of the average value $\bar{x}$ of a set of quantities x_i taking into account the possibility that any particular value may appear several, say n_i, times. Then:

$$\bar{x} = \frac{1}{N_0} \Sigma n_i x_i = \Sigma p_i x_i.$$

This may be generalized, and the expression for the average value of any function of x is

$$\overline{f(x)} = \Sigma p_i f(x_i). \qquad \text{(IX–1)}$$

In experimental measurements we may make a large number K of observations and find the ith result k_i times. Now the ratio k_i/K is not the probability p_i of the ith result as we have defined it, but for our purposes we assume that k_i/K approaches arbitrarily closely to p_i as K becomes very large:

$$\lim_{K \to \infty} \frac{k_i}{K} = p_i.$$

This assumption is not subject to mathematical proof, because a limit may not be evaluated for a series with no law of sequence of terms.

Addition Theorem. We turn now to the compounding of several probabilities, and consider first the addition theorem. Given a set of N_0 objects (or events, or results, etc.) containing n_i objects of the kind a_i, and given that the kinds a_1, a_2, $\cdots$ a_i have no members in common, then the probability that one of the N_0

objects belongs to a combined group $a_1 + a_2 + \cdots a_j$ is just $\sum_{i=1}^{i=j} p_i$. Thus for two mutually exclusive events with probabilities p_1 and p_2 the probability of one or the other occurring is just $p_1 + p_2$. When one card is drawn from a full deck the chance of its being either a five or a ten is $4/52 + 4/52 = 2/13$. (When one draws one card while already holding, say, four cards none of which is a five or ten, the probability then of getting either a five or a ten is slightly greater, $4/48 + 4/48 = 1/6$, provided there is available no information as to the identity of other cards that may already have been withdrawn.) When a coin is tossed the probability of either "heads" or "tails" is $\frac{1}{2} + \frac{1}{2} = 1$.

Multiplication Theorem. Another type of compounding of probabilities is described by the multiplication theorem. If the probability of an event i is p_i, and if after i has happened the probability of another event j is p_j, then the probability that first i happens and then j happens is $p_i \times p_j$. If a coin is tossed twice the probability of getting "heads" twice is $\frac{1}{2} \times \frac{1}{2} = \frac{1}{4}$. If two cards are drawn from an initially full deck the probability of two aces is $4/52 \times 3/51$. The probability of four aces in four cards drawn is $4/52 \times 3/51 \times 2/50 \times 1/49$. (The probability of drawing five aces in five cards is $4/52 \times 3/51 \times 2/50 \times 1/49 \times 0/48 = 0$.)

Binomial Distribution. The binomial distribution law treats one fairly general case of compounding of probabilities. Given a very large set of objects in which the probability of occurrence of an object of a particular kind w is p, then, if n objects are withdrawn from the set, the probability $W(r)$ that exactly r of the objects are of the kind w is given by

$$W(r) = \frac{n!}{(n-r)!r!} p^r(1-p)^{n-r}. \qquad \text{(IX-2)}$$

To see how this combination of terms actually represents the probability in question, think for a moment of just r of the n objects. That the first of these is of the kind w has the probability p; that the first and second are of the kind w has the probability p^2, etc., and the probability that all r objects are of the kind w is p^r. But, if exactly r of the n objects are to be of this kind, the remaining $n - r$ objects must be of some other kind; this proba-

bility is $(1 - p)^{n-r}$. Thus we see that, for a particular choice of r objects out of the n objects, the probability of exactly r of kind w is $p^r(1 - p)^{n-r}$; this particular choice is not the only one. The first of the r objects might be chosen (from the n objects) in n different ways; the second in $n - 1$ ways; the third in $n - 2$ ways, and the rth in $n - r + 1$ ways. The product of these terms, $n(n - 1)(n - 2)\cdots(n - r + 1)$, is $\dfrac{n!}{(n - r)!}$, and this coefficient must be used to multiply the probability just found. But this coefficient is actually too large in that it not only gives the total number of possible arrangements of the objects in the way required but also includes the number of arrangements which differ only in the order of selection of the r objects. So we must divide by the number of permutations of r objects which is $r!$. Thus, the final coefficient is $n!/(n - r)!r!$, which is that in equation IX–2. The law (equation IX–2) is known as the binomial distribution law because this coefficient is just the coefficient of the x^r term in the binomial expansion of $(1 + x)^n$.

C. RADIOACTIVITY AS A STATISTICAL PHENOMENON

Binomial Distribution for Radioactive Disintegrations. We may apply the binomial distribution law to find the probability $W(m)$ of obtaining just m disintegrations in time t from N_0 original radioactive atoms. We think of N_0 as the number n of objects chosen for observation (in our derivation of the binomial law) and we think of m as the number r that are to have a certain property (namely, that of disintegrating in time t), so that for this case the binomial law becomes

$$W(m) = \frac{N_0!}{(N_0 - m)!m!} p^m (1 - p)^{N_0 - m}.$$

Now the probability of an atom not decaying in time t, $1 - p$ in the equation above, is given [1] by the ratio of the number N that survive the time interval t to the initial number N_0,

$$\frac{N}{N_0} = e^{-\lambda t};$$

[1] See chapter I, p. 7.

and p is then $1 - e^{-\lambda t}$. We now have

$$W(m) = \frac{N_0!}{(N_0 - m)!m!}(1 - e^{-\lambda t})^m(e^{-\lambda t})^{N_0 - m}. \quad \text{(IX-3)}$$

Time Intervals Between Disintegrations. Since the time of Schweidler's derivation of the exponential decay law from probability considerations a number of experiments have been made to test the applicability of these statistical laws to the phenomena of radioactivity. As an example of the positive evidence obtained we consider the distribution of time intervals between disintegrations. The probability of this time interval having a value between t and $t + dt$, which we write as $P(t)\,dt$, is given by the product of the probability of no disintegration between 0 and t and the probability of a disintegration between t and $t + dt$. The first of these two probabilities is given by equation IX-3 with $m = 0$:

$$W(0) = \frac{N_0!}{N_0!0!}(1 - e^{-\lambda t})^0(e^{-\lambda t})^{N_0} = e^{-N_0\lambda t}.$$

(Notice that $0! = 1$.) The probability of one of the N_0 atoms disintegrating in the time dt is clearly $N_0\lambda\,dt$. (See chapter I, page 6, or obtain this result as $W(1)$ from equation IX-3 with $m = 1$, t replaced by dt, and all terms in $(dt)^2$ and higher neglected.) Then,

$$P(t)\,dt = N_0\lambda e^{-N_0\lambda t}\,dt.$$

Experiments designed to test this result usually measure a large number s of time intervals between disintegrations and classify them into intervals differing by the short but finite length Δt; then the probability for intervals between t and $t + \Delta t$ should be $N_0\lambda e^{-N_0\lambda t}\,\Delta t$, and the number of measured intervals between t and $t + \Delta t$ should be $sN_0\lambda e^{-N_0\lambda t}\,\Delta t$. For example, Feather has found experimentally that the logarithm of the number of intervals between t and $t + \Delta t$ is proportional to t, as required by this formula.

Average Disintegration Rate. Another application of the binomial law to radioactive disintegrations may be seen if we calculate the expected average value for a set of numbers obeying the binomial distribution law. We will for the moment revert to the

notation of equation IX–2 and for further convenience will represent $1 - p$ by q.

$$W(r) = \frac{n!}{(n-r)!r!} p^r q^{n-r}.$$

The average value to be expected for r is obtained from equation IX–1:

$$\bar{r} = \sum_{r=0}^{r=n} W(r)r = \sum_{r=0}^{r=n} r \frac{n!}{(n-r)!r!} p^r q^{n-r}.$$

To evaluate this awkward-appearing summation consider the binomial expansion of $(px + q)^n$:

$$(px+q)^n = \sum_{r=0}^{r=n} \frac{n!}{(n-r)!r!} p^r x^r q^{n-r} = \sum_{r=0}^{r=n} x^r W(r).$$

Differentiating with respect to x we obtain

$$np(px+q)^{n-1} = \sum_{r=0}^{r=n} r x^{r-1} W(r). \qquad \text{(IX–4)}$$

Now letting $x = 1$ and using $q = 1 - p$, we have the desired expression:

$$np = \sum_{r=0}^{r=n} rW(r) = \bar{r}.$$

This result should not be surprising; it means that the average number $\bar{r}$ of the n objects which are of the kind w is just n times the probability for any given one of the objects to be of the kind w.

The foregoing result may be interpreted for radioactive disintegration if n is set equal to N_0 and $p = 1 - e^{-\lambda t}$, as before. Then the expected average number M of atoms disintegrating in the time t is: $M = N_0(1 - e^{-\lambda t})$. For small values of λt, that is, for times of observation short compared to the half-life, we may use the approximation $e^{-\lambda t} = 1 - \lambda t$, and then $M = N_0 \lambda t$. The disintegration rate $\mathbf{R}$ to be expected is: $\mathbf{R} = M/t = N_0\lambda$. (This corresponds to the familiar equation $-dN/dt = \lambda N$.)

Expected Standard Deviation. A more interesting question is: What may we expect for the standard deviation σ_r for this expected average value $\bar{r}$ (or M)? If we differentiate equation IX–4 again with respect to x we get

$$n(n-1)p^2(px+q)^{n-2} = \sum_{r=0}^{r=n} r(r-1)x^{r-2}W(r).$$

Again letting $x = 1$ and using $p + q = 1$ we have

$$n(n-1)p^2 = \sum_{r=0}^{r=n} r(r-1)W(r) = \sum_{r=0}^{r=n} r^2W(r) - \sum_{r=0}^{r=n} rW(r);$$

$$n(n-1)p^2 = \overline{r^2} - \bar{r}.$$

Recall that the dispersion σ_r^2 is given by

$$\sigma_r^2 = \overline{r^2} - \bar{r}^2;$$

now combining we have

$$\sigma_r^2 = n(n-1)p^2 + \bar{r} - \bar{r}^2,$$

and with $\bar{r} = np$:

$$\sigma_r^2 = n^2p^2 - np^2 + np - n^2p^2 = np(1-p) = npq;$$

$$\sigma_r = \sqrt{npq}.$$

For the case of radioactive disintegration this becomes

$$\sigma = \sqrt{N_0(1 - e^{-\lambda t})e^{-\lambda t}} = \sqrt{Me^{-\lambda t}}.$$

In counting practice λt is usually small; that is, the observation time t is short compared to the half-life, and when this is so $\sigma = \sqrt{M}$. If a reasonably large number m of counts has been obtained that number m may be used in the place of M for the purpose of evaluating σ. Thus if 100 counts are recorded in 1 min the expected standard deviation is $\sigma \approx \sqrt{100} = 10$, and the counting rate might be written 100 ± 10 counts per min. If 1000 counts are recorded in 10 min the standard deviation of this number is $\sigma = \sqrt{1000} = 32$; the counting rate is $\dfrac{1000 \pm 32}{10} = 100 \pm 3.2$ counts per min. Thus we see that for a given counting

rate **R** the σ for the rate is inversely proportional to the square root of the time of measurement:

$$\mathbf{R} = \frac{m}{t};$$

$$\sigma_{\text{rate}} = \frac{\sqrt{m}}{t} = \frac{\sqrt{\mathbf{R}t}}{t} = \sqrt{\frac{\mathbf{R}}{t}}. \qquad \text{(IX–5)}$$

What is the result in an experiment in which the counting time is long compared to the half-life? As $\lambda t \to \infty$, $e^{-\lambda t} \to 0$, and in this limit $\sigma = \sqrt{Me^{-\lambda t}} = 0$. The explanation is clear; if we start with N_0 atoms and wait for all to disintegrate, then the number that disintegrate is exactly N_0. However, in actual practice we observe not the number of disintegrations but that number times a coefficient c which denotes the probability of a disintegration resulting in an observed count. Taking this into account we see that in this limiting case the proper representation of $\sigma = \sqrt{npq}$ is $\sigma = \sqrt{N_0c(1-c)}$. If $c \ll 1$ then $\sigma = \sqrt{N_0c} = \sqrt{\text{no. of counts}}$ as before. For cases where $\lambda t \sim 1$ and c is neither unity nor very small a more exact analysis based on $\sigma = \sqrt{npq}$ should be made, with the result that $\sigma = \sqrt{Mc(1 - c + ce^{-\lambda t})}$.

The introduction of the detection coefficient c in the preceding paragraph may raise the question as to why it is not necessary to take account of this coefficient in the more familiar case with λt small, where we have written $\sigma = \sqrt{m}$. If we do consider c in this case, we have for the probability of one atom producing a count in time t, $p = (1 - e^{-\lambda t})c$; and $q = 1 - p = 1 - c + ce^{-\lambda t}$. Then,

$$\sigma = \sqrt{N_0(1 - e^{-\lambda t})c(1 - c + ce^{-\lambda t})},$$

and for λt small and the same approximations as before:

$$\sigma = \sqrt{N_0\lambda tc} = \sqrt{Mc} = \sqrt{\text{no. of counts recorded}}.$$

This is just the conclusion we had reached without bothering about the detection efficiency. However, it should be emphasized that actual counts and not scaled counts from a scaling circuit must be used in these equations.

D. POISSON AND GAUSSIAN APPROXIMATIONS OF THE DISTRIBUTION LAW

Poisson Distribution. The binomial distribution law may be put into a more convenient approximate form if we impose the restrictions $\lambda t \ll 1$, $N_0 \gg 1$, $m \ll N_0$, that is, if we consider a large number of active atoms observed for a time short compared to their half-life. It is also necessary to make use of these mathematical approximations:

(1) $$e^{\lambda t} = 1 + \lambda t, \quad \text{neglecting subsequent terms.}$$

(2) $$x! = \sqrt{2\pi x}\, x^x e^{-x} \quad \text{(Stirling's approximation).}$$

(3) $$\left(1 - \frac{m}{N_0}\right)^{N_0} = \lim_{N_0 \to \infty} \left(1 - \frac{m}{N_0}\right)^{N_0} = e^{-m}, \quad \text{since } N_0 \gg 1.$$

With these restrictions and approximations and with $M = N_0 \lambda t$, equation IX–3 may in a straightforward way be put into the form known as the Poisson distribution:

$$W(m) = \frac{M^m e^{-M}}{m!}.$$

In words, the probability of obtaining the particular number of counts m, where the average to be expected is M, is $M^m e^{-M}/m!$. This approximation is very good even for N_0 as small as 100 and λt as large as 0.01. Two features of this distribution might be noticed in particular. The probability of obtaining $m = M - 1$ is equal to the probability of obtaining $m = M$, or $W(M) = W(M - 1)$. For large M the distribution is very nearly symmetrical about $m = M$ if values of m very far from M be excluded.

Gaussian Distribution. A further approximation of the distribution law may be made for large m (say >100) and for $|M - m| \ll M$. With these additional restrictions and with the approximate expansion,

$$\ln\left(1 + \frac{M - m}{m}\right) = \frac{M - m}{m} - \frac{(M - m)^2}{2m^2},$$

neglecting subsequent terms, we may modify the Poisson distribution to obtain the Gaussian distribution:

$$W(m) = \frac{1}{\sqrt{2\pi M}}\, e^{-\frac{(M-m)^2}{2M}}.$$

It will be noticed that this distribution is symmetrical about $m = M$. For both the Poisson and Gaussian distributions we may derive $\sigma = \sqrt{M}$, or $\sigma = \sqrt{m}$ for large m.

E. EXPERIMENTAL APPLICATIONS

Addition and Subtraction of Counting Rates. An important practical consideration is the addition and subtraction of counting results or counting rates. The Poisson distribution expression is suitable for the treatment of these cases, but the derivations are much too tedious to be included here. The very significant results are these:

1. The sum of two Poisson distributions is itself a Poisson distribution. Hence, the dispersion σ_s^2 and standard deviation σ_s of a sum are given by: $\sigma_s^2 = \sigma_1^2 + \sigma_2^2 + \cdots$ and $\sigma_s = \sqrt{\sigma_1^2 + \sigma_2^2 + \cdots}$.

2. The difference of two Poisson distributions is not a Poisson distribution; the dispersion σ_d^2 and standard deviation σ_d of the difference are given by: $\sigma_d^2 = \sigma_1^2 + \sigma_2^2$ and $\sigma_d = \sqrt{\sigma_1^2 + \sigma_2^2}$.

As an example suppose that the background counting rate of a counter is measured, and 600 counts are recorded in 15 min. Then with a sample in place the total counting rate is measured, and 1000 counts are recorded in 10 min. We wish to know the net counting rate due to the sample and the standard deviation of this net rate. First the background rate $\mathbf{R}_b$ is

$$\mathbf{R}_b = \frac{600 \pm \sqrt{600}}{15} = \frac{600 \pm 24}{15} = 40 \pm 1.6 \text{ counts per min.}$$

The total rate $\mathbf{R}_t$ is

$$\mathbf{R}_t = \frac{1000 \pm \sqrt{1000}}{10} = \frac{1000 \pm 32}{10} = 100 \pm 3.2 \text{ counts per min.}$$

The net rate $\mathbf{R}_n = 100 - 40 = 60$ counts per min, and its standard deviation is $\sigma_n = \sqrt{1.6^2 + 3.2^2} = 3.6$; and $\mathbf{R}_n = 60 \pm 3.6$ counts per min.

Ratios and Products of Counting Rates. In many types of experiments the ratio of two counting rates is wanted. What is the standard deviation of this ratio $Q = \mathbf{R}_1/\mathbf{R}_2$, if the two standard deviations σ_1 and σ_2 are known? It may be shown by straightfor-

ward algebraic operations that for small deviations the ratio σ_Q^*/Q is given by

$$\frac{\sigma_Q^*}{Q} = \pm\frac{\sigma_1}{\mathbf{R}_1} \pm \frac{\sigma_2}{\mathbf{R}_2},$$

where for the moment we mean by σ_Q^* the particular deviation in Q resulting from possible combinations of the deviations σ_1 and σ_2 in $\mathbf{R}_1$ and $\mathbf{R}_2$. To obtain instead the standard deviation σ_Q we must assume that the two fractional deviations of the rates are not simply additive but on the average combine to give a root-mean-square deviation:

$$\frac{\sigma_Q}{Q} = \sqrt{\left(\frac{\sigma_1}{\mathbf{R}_1}\right)^2 + \left(\frac{\sigma_2}{\mathbf{R}_2}\right)^2}.$$

This is the formula to be used for evaluation of the standard deviation σ_Q of the ratio Q of two quantities. A similar expression is applicable for the product P of two or more rates:

$$\frac{\sigma_P}{P} = \sqrt{\left(\frac{\sigma_1}{\mathbf{R}_1}\right)^2 + \left(\frac{\sigma_2}{\mathbf{R}_2}\right)^2 + \cdots}.$$

(See exercise 10 at the end of this chapter.)

Gaussian Error Curve. Knowledge of the distribution law permits a quantitive evaluation of the probability of a given deviation of a measured result m from the proper average M to be expected. The Gaussian distribution is convenient for this purpose. With the absolute error $|M - m| = \epsilon$, and with the assumption that the integral numbers are so large that the distribution may be treated as continuous, the probability $W(\epsilon)d\epsilon$ of an error between ϵ and $\epsilon + d\epsilon$ is given by

$$W(\epsilon)\,d\epsilon = \frac{2}{\sqrt{2\pi M}}\, e^{-\frac{\epsilon^2}{2M}}\, d\epsilon.$$

The factor 2 arises from the existence of positive and negative errors with equal probability within the limits of validity of this approximation. Recalling that $\sigma = \sqrt{M}$ we have

$$W(\epsilon)\,d\epsilon = \frac{1}{\sigma}\sqrt{\frac{2}{\pi}}\, e^{-\frac{\epsilon^2}{2\sigma^2}}\, d\epsilon.$$

The probability of an error greater than $k\sigma$ is obtained by integration from $\epsilon = k\sigma$ to $\epsilon = \infty$. Numerical values of this integral as a function of k may be found in handbooks. For example, we have taken for table IX–2 some representative values from the table, "Probability of Occurrence of Deviations" in the Chemical Rubber Publishing Company *Handbook of Chemistry and Physics.*

TABLE IX–2

k	0	0.674	1	2	3	4
Probability of $\epsilon > k\sigma$	1.00	0.50	0.32	0.046	0.0027	0.00006

Notice that errors greater than 0.674σ and errors smaller than 0.674σ are equally probable; 0.674σ is called the "probable error" and is sometimes given rather than the standard deviation when counting data are reported. In plots of experimental curves it can be very convenient to indicate the probable error of each point (by a mark of the proper length); then on the average the smooth curve drawn should be expected to pass through about as many "points" as it misses.

Comparison with Experiment. We may now return to a consideration of the typical counting data in table IX–1. We have already found from the deviations between the ten measurements $\sigma = \sqrt{\frac{1}{N_0}\Sigma(x_i - \bar{x})^2} = 11.3$. Now, if the counting rate measured there represents a random phenomenon, as we expect it should, we may evaluate the expected σ for the result in any minute as the square root of the number of counts. For a typical minute, the 9th, we find $\sigma = \sqrt{101} = 10$, and for other minutes other values not much different. Because these agree reasonably with the 11.3 there is evidence for the random nature of the observed counting rate. This test should occasionally be made on the data from a counting instrument.

In addition to estimating σ for any minute's counting in table IX–1, we may now estimate the σ for the average of the ten observations (which we could not do directly from the definition of

σ). The average counting rate with its standard deviation is $\mathfrak{x} = \frac{990 \pm \sqrt{990}}{10} = 99.0 \pm 3.1$ counts per min. This means that the probability that the true average is between 95.9 and 102.1 is from table IX–2 just $1 - 0.32 = 0.68$. Actually, when the counting data given in table IX–1 were obtained the average rate was measured much more accurately in a 100-min count, and the result was $\frac{10{,}042 \pm \sqrt{10{,}042}}{100} = 100.4 \pm 1.0$ counts per min.

Case of Very Few Counts. A question sometimes met in counting experiments is what can be done with results which show very few total counts (not net counts), or even no counts at all. A treatment of this problem has been given by R. W. Dodson. He shows that for the Poisson distribution the best value to assume for the true average number of counts M is not the measured value m, but rather $m + 1$. (This is clearly related to one of the features already pointed out for the Poisson distribution.) The best guess for the standard deviation is $\sigma = \sqrt{m + 1}$. Thus, if the observed count is $m = 0$, we take for the answer $M = 1 \pm 1$; if the observed $m = 1$ we take $M = 2 \pm 1.4$; etc.

Counter Efficiencies. As another application of the methods of this chapter to counting techniques we may estimate the efficiency of a Geiger counter for rays of a given ionizing power, with the assumptions that any ray which produces at least one ion pair in the counter gas is counted and that effects at the counter walls are negligible. Knowledge of the nature of the radiation and the information given in chapter VII permit an estimate of the average number of ion pairs a to be expected within the path length of the radiation in the counter filling gas. The problem then is to find the probability that a ray pass through the counter leaving no ion pairs and thus not be counted. We think of the path of the ray in the counter as divided into n segments of equal length; if n is very large, each segment will be so small that we may neglect the possibility of having two ion pairs in any segment. Then just a of the n segments will contain ion pairs, and by definition the probability of having an ion pair in a given segment is $p = a/n$. Now by equation IX–2 for the binomial distribution

we have the probability for no ion pairs in n segments; that is, for $r = 0$:

$$W(0) = \frac{n!}{n!0!} p^0(1-p)^n = (1-p)^n = \left(1 - \frac{a}{n}\right)^n.$$

Since the probability [2] is evaluated correctly only as n becomes very large,

$$W(0) = \lim_{n \to \infty} \left(1 - \frac{a}{n}\right)^n = e^{-a}.$$

The probability of counting the ray, which is the efficiency to be determined, is then $1 - W(0) = 1 - e^{-a}$. As a particular example consider a fast β particle with the relatively low specific ionization of 5 ion pairs per mm in air and a path length of 10 mm in a counter gas which is almost pure argon at 7.6 cm pressure. We estimate a from these assumptions, correcting for the relative densities of air and the argon:

$$a = 5 \times 10 \times \frac{7.6}{76} \times \frac{40}{29} = 7.$$

The corresponding estimated counter efficiency is $1 - e^{-7} = 99.9$ per cent. It should not be expected that an efficiency calculated in this way is very precise; wall effects may be important, and the assumption of random distribution of ion pairs along the β-ray path is not entirely consistent with the mechanism of energy loss by ionization presented in chapter VII.

Coincidence Correction. If a counter has a recovery time (or dead time or resolving time) τ after each recorded count during which it is completely insensitive, the total insensitive time per unit time is $\mathbf{R}\tau$, where $\mathbf{R}$ is the observed counting rate. If $\mathbf{R}^*$ is the rate that would be recorded if there were no coincidence losses then the number of lost counts per unit time is $\mathbf{R}^* - \mathbf{R}$, and is given by the product of the rate $\mathbf{R}^*$ and the fraction of insensitive time $\mathbf{R}\tau$:

$$\mathbf{R}^* - \mathbf{R} = \mathbf{R}^*\mathbf{R}\tau,$$

$$\mathbf{R}^* = \frac{\mathbf{R}}{1 - \mathbf{R}\tau}. \qquad \text{(IX–6)}$$

[2] We might have evaluated this probability more easily from the Poisson distribution expression: $W(0) = a^0 e^{-a}/0! = e^{-a}$.

A number of variants of this formula are also in use. One expression (the Schiff formula) is $\mathbf{R}^* = \mathbf{R}e^{\mathbf{R}^*\tau}$; this is derived from a calculation of the probability $W(0)$ of having had no event during the time τ immediately preceding any event, but it neglects the possibility that any preceding event itself may not have led to a count through coincidence loss.[3] Another approximate expression is derived from the first two terms in the binomial expansion of $(1 - \mathbf{R}\tau)^{-1}$ appearing in equation IX–6:

$$\mathbf{R}^* = \mathbf{R}(1 + \mathbf{R}\tau) = \mathbf{R} + \mathbf{R}^2\tau.$$

This form is especially convenient for the interpretation of an experiment designed to measure τ by measuring the rates $\mathbf{R}_1$ and $\mathbf{R}_2$ produced by two separate sources and the rate $\mathbf{R}_t$ produced by the two sources together, all these rates including the background effect $\mathbf{R}_b$. Obviously,

$$\mathbf{R}_1{}^* + \mathbf{R}_2{}^* = \mathbf{R}_t{}^* + \mathbf{R}_b,$$

where we have neglected the coincidence loss in the measurement of the low background rate. Replacing by $\mathbf{R}_1{}^* = \mathbf{R}_1 + \mathbf{R}_1{}^2\tau$, etc., and rearranging we have,

$$\tau = \frac{\mathbf{R}_1 + \mathbf{R}_2 - \mathbf{R}_t - \mathbf{R}_b}{\mathbf{R}_t{}^2 - \mathbf{R}_1{}^2 - \mathbf{R}_2{}^2}.$$

Integrating Measuring Instruments. For measurements with counting instruments we have seen the convenience of the simple expression $\sigma = \sqrt{\text{no. of counts}}$. In the counting rate meter, where a steady meter deflection is observed, what value may be assigned to the standard deviation? In this instrument a combination of resistance R and capacitance C effectively averages the rate of arrival of pulses over an interval of the order of magnitude of the time constant RC; a quantitative analysis shows that the effective interval is 2RC. Representing the counting rate in counts per minute by $\mathbf{R}_1$ we obtain the standard deviation of this

[3] It may be noticed that the Schiff formula might be expected to correspond more closely to the conditions of coincidence loss in a mechanical register, where a new pulse within a dead time could initiate a new dead-time period although it would not be recorded. There exists also the opportunity for coincidence losses in the electric circuits.

rate from equation IX–5, where $t = 2RC/60$ min for R in ohms and C in farads:

$$\sigma_{\text{rate}} = \sqrt{\frac{R_1}{t}} = \sqrt{\frac{30R_1}{RC}}.$$

For an instrument using an ionization chamber with a d-c amplifier the same expression may be used, provided the activity can be evaluated approximately as a rate of arrival of ionizing particles and the value of the longest time constant is known (ordinarily that of the collecting electrode and first grid circuit unless a very slow galvanometer is employed). If necessary the time constant may be approximated as the time for a deflection to be reduced to $1/e$ of its value after removal of an active sample.

In a rate-of-drift-measuring method, as in the d-c amplifier with infinite grid resistance or in the Lauritsen electroscope, the number m of ionizing particles arriving during the time of measurement is estimated, and the standard deviation of the activity **A** is given by $\sigma = \mathbf{A}\frac{\sqrt{m}}{m} = \frac{\mathbf{A}}{\sqrt{m}}$. These considerations are not easily applied to γ-ray measurements in ionization chambers, but the statistical uncertainties would be at least as great as indicated by these formulas.

EXERCISES

1. Derive the relation given on p. 200: $\sigma_x^2 = \overline{x^2} - \bar{x}^2$.

2. Mr. Jones' automobile license carries a six-digit number. What is the probability that it has (*a*) exactly one 4, (*b*) at least one 4?

Answer: (*b*) 0.46856.

3. Given an atom of a radioactive substance with decay constant λ, what is

(*a*) The probability of it decaying between 0 and dt?
(*b*) The probability of it decaying between 0 and t?

4. The following numbers were obtained in the measurement of a physical quantity x.

Set 1: 90,110,100.
Set 2: 99,101,100.

What is the average value obtained in each set? In which set would you consider the measurements more reliable? What is the standard deviation for each set?

5. A given Geiger counter has a measured background rate of 900 counts in 30 min. With a sample of a long-lived activity in place, the total measured rate was 1100 counts in 20 min. What is the net sample counting rate and its standard deviation?

Answer: 25.0 ± 1.9 counts per min.

6. Denote by $\mathbf{R}_t$ and $\mathbf{R}_b$ the total and background counting rates for a long-lived sample, and calculate the optimum division of available counting time between sample and background for minimum σ on the net counting rate.

Answer: $\frac{t_t}{t_b} = \sqrt{\frac{\mathbf{R}_t}{\mathbf{R}_b}}$

7. Refer to exercise 3, chapter VIII. What would be the detection efficiency for that counter and that β ray?

8. (*a*) Sample A, sample B, and background alone were each counted for 10 min; the observed total rates were 110, 205, and 44 counts per min, respectively. Find the ratio of the activity of sample A to that of sample B and the standard deviation of this ratio. (*b*) Sample C was counted on the same counter for 2 min, and the observed total rate was 155 counts per min. Find the ratio, and its standard deviation, of the activity of C to that of A. *Answer:* (*a*) 0.41 ± 0.027.

9. Show that equation IX–6 may be derived by summing up the probabilities of losing one count, two counts, three counts, etc., during each dead time interval. (The Poisson distribution is convenient for this.)

10. The expressions given for the standard deviations of sums, differences, quotients, and products, and indeed for any function F of the individual rates, $F = F(\mathbf{R}_1, \mathbf{R}_2 \cdots)$, may be found by the following approximate method, provided all deviations are small and all rates are large numbers: replace the usual differential expression,

$$dF = \frac{\partial F}{\partial \mathbf{R}_1} d\mathbf{R}_1 + \frac{\partial F}{\partial \mathbf{R}_2} d\mathbf{R}_2 + \cdots,$$

by a form showing the root-mean-square differential deviation,

$$dF = \sqrt{\left(\frac{\partial F}{\partial \mathbf{R}_1} d\mathbf{R}_1\right)^2 + \left(\frac{\partial F}{\partial \mathbf{R}_2} d\mathbf{R}_2\right)^2 + \cdots}.$$

Use this equation to derive for the sum or difference of two rates $\mathbf{R}_1$ and $\mathbf{R}_2$, that is, for $F = \mathbf{R}_1 + \mathbf{R}_2$ and $F = \mathbf{R}_1 - \mathbf{R}_2$, the standard deviation dF corresponding to the individual standard deviations $d\mathbf{R}_1$ and $d\mathbf{R}_2$. Find the formulas for standard deviations of quotients and products in the same way, with $F = \mathbf{R}_1/\mathbf{R}_2$ and $F = \mathbf{R}_1\mathbf{R}_2$.

11. What is the probability of a penny turning heads up *at least* once in n throws?

12. Use the data given in exercise V–1 to find the half-lives by the method of least squares rather than by graphical solution. Assume that there is no error in the measured time intervals and that all the measured counting rates have standard deviations of ± 1 per cent.

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X. TECHNIQUES FOR MEASUREMENT AND STUDY OF RADIATIONS

A. GENERAL TECHNIQUES

The principal types of instruments used for detection and measurement of radiations from radioactive substances have been discussed in chapter VIII. Here we wish to emphasize the important role of proper techniques in the course of measurements of this kind. The choice of instruments and techniques will be determined in large part by the kinds of information desired. In a simple tracer application, employing one active isotope of favorable properties and available with ample activity and known purity, a single instrument (counter, electrometer, or electroscope) is likely to be sufficient, and measuring techniques may offer no problems. The opposite extreme would be a large radiochemical laboratory devoted to the identification and characterization of new radioactive species as made in a nuclear chain reactor or a new high-energy accelerator; here a large number and wide variety of instruments, including highly specialized types, must be available, and the associated techniques and manipulations will be complex and ingenious. Most radiochemical laboratories represent intermediate situations. A number of tracers are likely to be in use, calling for different choices of detectors and sample handling procedures. Frequently the desired radioactivity must be isolated and identified and checked for purity with regard to contamination by other radioactive substances. Occasions are recalled when undiscovered isotopes have been sought for use as chemical tracers (I^{131} and C^{14} were found in this way). The typical laboratory may have G–M counters arranged for γ counting, thin-window counters for most β counting, perhaps a Lauritsen electroscope, and possibly an ionization chamber with d-c amplifier, useful because of its convenience and its linear response over a very wide range of sample strengths. It is very convenient to have a standard arrangement for holding standard-size samples

in various positions, the same arrangement being used for as many of the instruments as possible.

In one of our laboratories, that at the Department of Chemistry, Washington University, St. Louis, all instruments, G–M counter sets, ionization chamber, and Lauritsen electroscopes, except special-purpose instruments for C^{14} counting and for γ counting

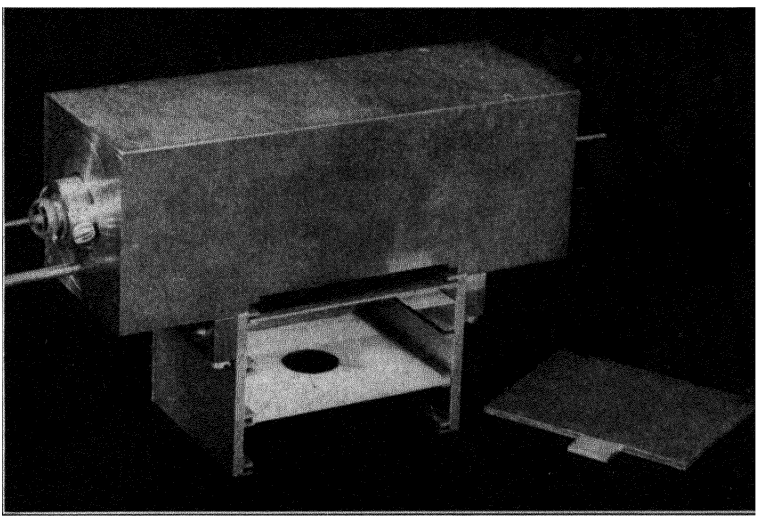


FIGURE X–1. A side-window Geiger–Müller tube with sample holder. A radioactive sample mounted on a standard card is in place in the third step. The first step is being used to hold a thin aluminum absorber, and space is available for insertion of additional absorbers between the sample and the counter window (not shown).

with samples in solution, are fitted with a standardized sample-holder arrangement with milled slots to hold samples at various distances below the detector; the top step is 0.50 cm below the instrument window, and three additional steps are spaced below at successive 1.50-cm intervals. The samples are mounted on pieces of cardboard 2 inches by 2.5 inches, supplied precut by a stationer; for the highest reproducibility the cardboard may be replaced by pieces of aluminum or plastic of the same size. These sample cards slide broadwise into the milled slots, the 2-inch edges being held by the slots, and stops are provided at the back of the slots for convenience in locating the samples in the proper position.

If the detector is a G–M counter, it and the sample-holding arrangement are usually enclosed in a lead shield 1 to 1½ inch thick to reduce background, including background effects from strong samples in the laboratory.

The spaces between the milled sample slots are also milled out to take absorbers cut to the same 2 × 2.5-inch size as the sample cards. Sets of aluminum absorbers are the most useful. They should be available in an assortment of thicknesses (nearly integral values are convenient), so that absorption curves may include points from about a milligram per square centimeter up to several grams per square centimeter. Our thicker absorbers are cut from thick aluminum stock, with a small tab left on one of the long sides which is marked with the thickness in milligrams per square centimeter. The thinnest absorbers are compounded of thin aluminum foils, held in rectangular 2 x 2.5-inch aluminum frames, again provided with tabs. These tabs are spaced along the side like index tabs and serve as convenient handles as well as labels. A set of lead absorbers is more convenient for determining γ-ray absorption curves, but it need not include very thin pieces. A few beryllium, paraffin, or polyethylene, $(CH_2)_n$, plastic absorbers of the same shape can be useful in that they are effective in stopping β rays but are almost completely transparent to γ rays.

All the measuring instruments should be checked occasionally—Geiger counters daily—with standard samples; it is best if a standard is chosen to have a radiation similar to the activity to be measured. An intercalibration of all the instruments for activities of interest can be useful but ordinarily should be depended on only for semiquantitative results. Each instrument must have its response to samples of different strengths determined; outside the linear-response region it should be used cautiously, with calibrated corrections. This calibration can be made in several ways: (1) with samples of different activity carefully prepared from aliquot portions of an active solution; (2) by comparison of the decay curve of a very pure short-lived activity of known half-life with the exponential decay to be expected; and (3) by measurements of the separate and combined effects of samples located in reproducible assigned positions (see chapter IX, page 214). With counters the failure of linearity at high counting rates is attributed to coincidence losses; the correction is known as a

coincidence correction.[1] Ordinarily the necessity for corrections amounting to more than a few per cent should be avoided.

B. CRITICAL ABSORPTION OF X RAYS

Absorption curves in aluminum and lead absorbers are used to determine the energies of β, γ, and X rays, as discussed in chapter VII, but the values obtained are rarely as accurate as those determined by the use of the electron spectrograph and the curved-crystal spectrograph mentioned in chapter VIII, section D. In radiochemical work accurate measurements of X-ray energies are often wanted to establish the identity of the chemical element. The X radiation is characteristic of the particular value of Z at the time of emission of the ray; X rays following β^- decay of a nucleus of charge Z correspond to atomic number $Z + 1$; those following β^+ or K-capture decay correspond to atomic number $Z - 1$; those following converted isomeric transitions correspond to atomic number Z. The ordinary absorption study does not establish the X-ray energy with sufficient accuracy for this purpose, and if the special instruments are not available a technique based on critical absorption edges may be employed.

To understand this method we have to recall that the emission of an X ray from an atom is due to the transition of an electron from one of the outer shells to a vacancy in a shell further in, say, from the L to the K shell. In X-ray terminology, X rays due to transitions from the L to the K shell are called K_α X rays ($K_{\alpha 1}$ and $K_{\alpha 2}$ corresponding to the electron originating in different sublevels of the L shell); X rays due to transitions from the M to the K shell are called K_β, etc. Similarly there are L_α, L_β, etc., X rays.

An X ray can be absorbed by an atom only if its energy corresponds at least to the energy difference between one of the filled shells and the lowest lying shell with a vacancy (the latter being very close in energy to the continuum). It is therefore found that the absorption coefficient (or the half-thickness) in a given element has discontinuities called absorption edges at those

[1] If the dead time of the counter tube is known to be much larger than the time constants of any of the other components the coincidence correction for any counting rate can be calculated from the dead time; see chapter IX, page 213.

X-ray energies corresponding to the binding energies of the K, L, etc., electrons. Thus X rays of energy insufficient for the removal of a K electron (essentially to the continuum) are poorly absorbed and have large half-thickness values, whereas those of energy just exceeding that of the K absorption edge are absorbed much more strongly (perhaps ten times more strongly).

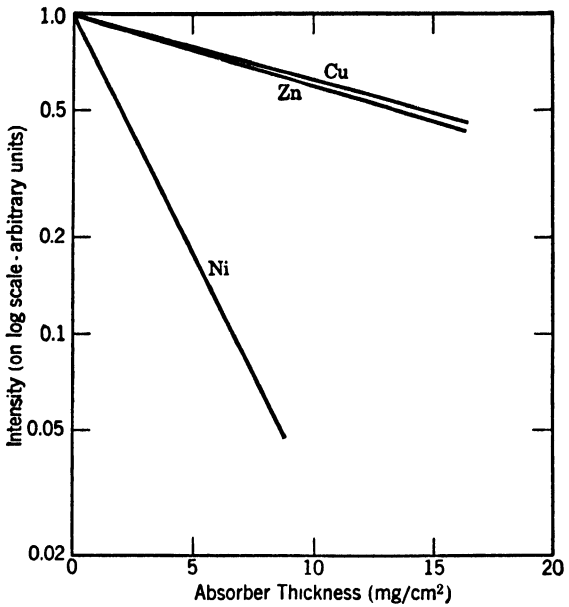


FIGURE X–2. Absorption of zinc K_α X rays in zinc, copper, and nickel. (These absorption curves were calculated from data given in A. H. Compton and S. K. Allison, *X-rays in Theory and Experiment*, New York, D. Van Nostrand Co., 1935.)

It is clear from this that an element is a poor absorber for its own characteristic X rays. The K_α X rays of an element have an energy corresponding to the difference between the K and L shells and can, therefore, not lift a K electron to one of the outer vacant shells in the same element. However, the binding energy of electrons decreases with decreasing Z; therefore, the K_α emission line of an element Z has an energy rather close to but slightly greater than the K absorption edge of an element of somewhat lower Z and is, therefore, strongly absorbed by that element, but not by the next higher one.

For example, the K_α X rays of zinc ($Z = 30$) have a wave length of 1.43 A (energy 8.7 kev). The K absorption edges of $_{29}$Cu and $_{28}$Ni are at 1.38 A (9.0 kev) and 1.48 A (8.4 kev), respectively. Therefore, nickel is a good absorber for zinc K_α X rays, and copper is not (figure X–2). The K_α X rays of gallium ($Z = 31$), on the other hand, are strongly absorbed both in nickel and copper because their wave length is 1.34 A (9.3 kev), but they are not absorbed well in zinc whose K absorption edge is at 1.28 A (9.7 kev).

Both the X-ray emission lines and the absorption edges of the elements can be found in tables, and suitable elements can be chosen as absorbers to decide the origin of a set of X rays accompanying a nuclear decay process. The K_α X rays are usually the most prominent lines, but occasionally, especially with very heavy elements, the absorption of other lines (K_β and L) also must be taken into account. Absorption curves are taken with each of two or three neighboring elements, and their comparison usually brackets the energy of the emission line sufficiently to determine the corresponding atomic number. It should be emphasized that it is entirely unnecessary to use pure elements as absorbers (this would be difficult or impossible for some elements). Compounds of the desired element can be used, provided other elements in the compounds do not appreciably absorb the X rays under investigation. Light elements such as hydrogen, oxygen, nitrogen, carbon are very poor absorbers for energetic X rays, and oxides, hydroxides, or carbonates are, therefore, usually suitable as absorbers. A very convenient method of preparing such absorbers is to suspend weighed amounts of the compound in water (or in an organic liquid) and to filter the suspension through filter paper on a sintered glass or Buchner funnel; the addition of a small amount of binder to the suspension is sometimes useful. Even solutions in shallow plastic dishes are used.

C. BETA-PARTICLE SIGN DETERMINATION

Magnetic Deflection. It is occasionally necessary to distinguish between β^- and β^+ particles. In the electron spectrograph this distinction is made easily according to the polarity of the magnetic or electrostatic field. Without such an instrument a crude determination may be made to distinguish a predominantly positron

from a predominantly negatron emitter. If the sample and detector are separated by a few inches and shielded from each other a magnetic field from an electromagnet or sizable permanent magnet [2] may be arranged to bend particles of one sign around the shield toward the detector.

Annihilation Radiation. Another method for the identification of positrons is based on the presence of the annihilation radiation (two γ rays of 0.51 Mev for each positron annihilated at the end of its range). For this procedure the sample on its card is placed several centimeters from the detector and facing away from the detector. Then enough beryllium, paraffin, or plastic absorber is inserted between sample and detector to stop all β particles. In this arrangement the detector should give a response to γ rays plus possible annihilation radiation from about 50 per cent of the positrons. The other 50 per cent of the positrons leave the sample in the direction opposite to the detector and end their ranges far away from it. Now with the addition of beryllium, paraffin, or plastic absorber on the face of the sample away from the detector, an increase in detector response would indicate positrons stopping in this new absorber and giving rise there to annihilation radiation. For a sample which emits positrons and no nuclear γ rays, and with the absorber–sample–absorber sandwich located at a large distance (compared to its own thickness) from the detector, the increase in response should amount to almost a factor of two.

D. ABSOLUTE DISINTEGRATION RATES

Alpha-disintegration Rates. The determination of absolute disintegration rates with precision is difficult with α emitters, more difficult with β emitters, and still more difficult in cases in which only γ rays may be measured. It is fortunate that absolute disintegration rates are not often needed; for most radiochemical work relative rates for various samples of the same substance or for the same sample at various times are sufficient. The methods for determination of absolute rates for long-lived α emitters (for the purpose of half-life determination) have already been mentioned in chapter V, section E. The absolute counting of α particles requires: (1) a detector of known efficiency, usually an

[2] We have used a permanent magnet designed for radar magnetrons: it provides a field of 1300 gauss over a volume of about 1 cubic inch.

ionization chamber with linear amplifier adjusted to count 100 per cent of the particles crossing the chamber; (2) a sample of known weight spread so uniformly thin that corrections for α particles stopped in the sample are small; (3) a known geometrical factor, usually about 50 per cent, including the correction for back-scattering of α rays from the sample support (which may amount to several per cent). With reasonable care values reliable to about 10 per cent can be obtained, and with great care the errors may be reduced to about 1 per cent. The calorimetric method mentioned in chapter V, section E, for the measurement of absolute α-disintegration rates is capable of at least comparable accuracy. It requires a larger sample activity but makes no demands on the sample's geometrical arrangement, thinness, and the like. It does require a knowledge of the α-particle energy; this is conveniently obtained from the range.

Known Geometry and Detector Efficiency. The remainder of this section will deal with the problem of determining absolute disintegration rates for samples of β emitters. (Because γ rays usually are found accompanying other nuclear changes determination of their absolute intensities ordinarily may be avoided; if not, comparison with a similar radiation is probably the best that can be done.) We will consider three methods; the first method involves estimates of the geometrical factor and detector efficiency as in the α-particle case. For this procedure the detector is almost always a Geiger counter because it counts individual β rays with an efficiency that may be close to 100 per cent. However, it is not easy to arrange the sample and counter so that the geometrical factor is known, and so that absorption of the β particles in the counter wall or window is known or negligible. The mica end window counter minimizes window absorption (windows as thin as 1.5 mg per cm^2 are available), but even with the sample made small and placed close to the window the geometrical factor will not approach 50 per cent so closely as would be desired, and a factor nearer 30 per cent is common. Also, the back-scattering for β particles is more important than for α particles (see table VII–4). Some of these difficulties are minimized in a "low-geometry" arrangement, with the sample placed several centimeters from the counter window and with a shield and aperture to define a cone of rays directed toward the sensitive region of the counter. The sample may be mounted on a very thin support

(mica is often convenient) to reduce back-scattering. Inversion of the sample will eliminate this back-scattering effect (except for the very small effect of the air backing), but then the effect of the support as an absorber must be taken into account.

Although the Geiger-counter efficiency for β particles at rates below those involving appreciable coincidence error is ordinarily taken very close to 100 per cent, the ideal situation in which even a single ion pair produces a count may not be realized for some counters or for some gas-filling mixtures. The efficiency of a counter for a given β-ray spectrum can be estimated by a coincidence counting experiment. The counter must have very thin windows on opposite sides or at opposite ends, or, better, be designed to require no windows at all, so that a collimated beam of the β rays may be passed through the counter and into a second counter without appreciable loss of lower-energy rays. Then each count in the second counter (rate = $\mathbf{R}_2$) shows the presence of a β particle that must have passed through the first counter (counting rate = $\mathbf{R}_1$), and the efficiency of the first counter in recording these particles is given by $\mathbf{R}_{12}/\mathbf{R}_2$, where $\mathbf{R}_{12}$ is the real net coincidence counting rate between the two counters. This determination can be made with three counters in a row, and then the efficiency of the second counter is given by $\mathbf{R}_{123}/\mathbf{R}_{13}$, the ratio of triple coincidences to double coincidences between the first and third counters. (Coincidence methods have been used also for approximate efficiencies for γ rays; in this case beryllium, paraffin, or plastic absorbers thick enough to stop any recoil electrons should be placed between counters.)

Calibrated Detector. In the second method for absolute disintegration rates a calibrated detector is used; here the absorption of β rays in the window must be small, but the only requirements on the geometry and counter efficiency are that these be reproducible and relatively insensitive to the β-ray spectrum. The β source used in calibrating these factors must have a known disintegration rate; ordinarily a β emitter in secular equilibrium with an α activity is used, and the absolute disintegration rate evaluated for the α emitter. Also, the energy spectrum of the β rays should be as nearly as possible the same for the standard and for the unknown activity. The rather energetic β particles from UX_2 in secular equilibrium with U_I, or possibly in transient equilibrium with freshly isolated UX_1, are often used as standard, with a thin

absorber to exclude U_I α particles and soft UX_1 (and UY) β particles. (Absorption of UX_2 β rays in this absorber and the counter window is corrected for by extrapolation of a measured absorption curve back to zero absorber.) [3] The calibration of such a standard could be made by α-particle counting, but since the disintegration constant for U_I is already known it is necessary only to weigh the uranium. The National Bureau of Standards supplies (among other radioactivity standards) carefully standardized RaD-plus-RaE samples; the RaD β particles and RaF α particles may be absorbed in a 0.001-inch aluminum foil, and the medium-energy RaE β particles used as a standard for calibration. As supplied these preparations are deposited electrolytically (in PbO_2) on palladium-clad silver disks, and considerable back-scattering is to be expected. Identical blank disks are available so that the activity to be measured may be mounted on the same type of surface.

Coincidence Method. A different method for absolute disintegration rates of β sources whose spectra include γ rays has recently come into use. This method is easily understood for a simple case in which one γ quantum follows each β decay, and the spectrum is simple; in this case the method is free of all the sources of error already discussed. Consider two counters arranged to count β rays and γ rays, respectively, with measured counting rates $\mathbf{R}_\beta$ and $\mathbf{R}_\gamma$, and with $\beta-\gamma$ coincidences also measured with rate $\mathbf{R}_{\beta\gamma}$; then $\mathbf{R}_\beta = -\frac{dN}{dt}c_\beta$, $\mathbf{R}_\gamma = -\frac{dN}{dt}c_\gamma$, where the coefficients c_β and c_γ may be thought of as defined by these equations, and $\mathbf{R}_{\beta\gamma} = -\frac{dN}{dt}c_\beta c_\gamma$. Now $\frac{\mathbf{R}_\beta \mathbf{R}_\gamma}{\mathbf{R}_{\beta\gamma}} = -\frac{dN}{dt}$, and the absolute counting rate is determined with few assumptions. If the β spectrum

[3] For such an extrapolation to zero absorber the relative positions of sample, absorber, and sensitive region of the counter are very important. If the sample is placed at some distance, say 2 or 3 cm, from the counter, the measured counting rate with an absorber directly above the sample will be appreciably (perhaps as much as 10 or 15 per cent) higher than with the same absorber placed directly under the window. In fact, the addition of a thin absorber near the sample may cause an increase in the counting rate. This effect results from the scattering of β particles into the counter by the absorbers and is clearly related to the "self-scattering" effect discussed below (on p. 231). When a straight-line extrapolation to zero absorber thickness is wanted the absorbers should be placed close to the counter window.

is complex this ratio will give the correct disintegration rate only if the β counter window is sufficiently thin, and that counter does not discriminate appreciably against any component group of the spectrum. However, other sources of error must be considered. It is possible, although not likely, that strong angular correlation between the directions of emission of coincident β and γ rays might exist; this effect could be detected through variation of the angle between the two counters. A source of error of uncertain magnitude results from detection in the γ counter of bremsstrahlung produced by β particles stopping in the absorber used to shield the γ counter from β rays. This effect will increase $\mathbf{R}_\gamma$ without a corresponding increase in $\mathbf{R}_{\beta\gamma}$ because the particular β particles giving rise to this bremsstrahlung have little chance of being counted in the β counter. The magnitude of this error is not easy to estimate but should be fairly small; it will be in such a direction as to make values found for $-dN/dt$ too large. In most cases it would be reduced by a lead filter interposed between the aluminum absorber and the γ counter. A very appreciable error of the same sort would be expected for any sample emitting positrons because of the annihilation radiation. Also, any delayed γ rays which might trip the γ counter too late to be recorded in coincidence would lead to too large values of $-dN/dt$.

The National Bureau of Standards is currently encouraging the intercomparison of absolute β-disintegration rates between various radiochemical laboratories. The intercomparisons of I^{131} samples which have been reported show that uncertainties of the order of at least ± 20 per cent exist in most of the measurements. This may be seen in determinations made by all three methods in our laboratory (at St. Louis) on one of the I^{131} preparations:

(1) Calculated low-geometry counter (average of 3 samples each measured in 2 geometries)	108 ± 9 mrd/ml
(2) Comparison with RaD + RaE standard	125 ± 7 mrd/ml
(3) Coincidence counting:	
(*a*) with Al shield in front of γ counter	168 ± 9 mrd/ml
(*b*) with Al shield plus $\frac{1}{16}$ in. Pb filter	131 ± 11 mrd/ml

E. PREPARATION OF SAMPLES FOR COUNTING

Gamma versus Beta Counting. The determination of relative disintegration rates for several samples of the same kind, or for

the same sample at several times, is the measuring problem most often met in radiochemical work. This problem is much simpler than the determination of absolute disintegration rates, but some points of technique arise even here. If an active sample emits β and γ rays the relative advantages of arranging the detector to respond principally to β rays or principally to γ rays must be considered; ordinarily it is not advisable to have the detector response divided almost equally between the two radiations. For a given thin sample the β-ray effect will offer a greater sensitivity by a factor of very roughly 100, and this may be decisive. However, lack of reproducibility of absorption and self-absorption effects can be troublesome. Because the γ rays are almost always much less strongly absorbed, these effects are not so large when γ rays are counted, even for thicker counter walls and for much thicker samples; in fact, usually no uncertainties need be introduced if the various samples are prepared and mounted in some simple reproducible way—for example, in similar solutions of the same volume in test tubes placed in a fixed holder near the detector. In experiments where the specific activity (activity per gram or per milligram) rather than the total activity is a limiting factor, the sensitivity for γ counting may approach that for β counting because in γ-ray counting the sample can be larger.

Self-absorption Correction. For the measurement of soft radiation from an appreciably thick sample it is theoretically possible to calculate the effect of absorption of the radiation in the sample (self-absorption); however, no rigorous calculation is practical because it would require that the absorption curve for the radiation, the thickness of the sample, the solid angle subtended by the counter and the back-scattering effect be taken into account. If possible the samples should be made thin compared to the half-thickness value for the radiation. When thicker samples must be used it is advisable either to standardize the thickness at a fixed value or to prepare an empirical calibration curve for different thicknesses; in either case careful attention must be given to a reproducible mechanical form for the sample, and reproducibility should be tested by experiment.

Work with appreciably thick samples, in this sense, is most frequently necessary for the low-energy β emitters, especially C^{14} and S^{35}. Some approximate equations have been proposed for normal β-ray spectra with upper limits below 200 kev. The range R in milligrams per square centimeter is given very closely

by $R = E^{5/4}/150$, where E is the upper limit in kiloelectron volts. The absorption coefficient μ is roughly $5/R$. The half-thickness is then $d_{1/2} = \ln 2/\mu \approx 0.14R$. The detection coefficient c for a sample of thickness d is given by the approximate relation:

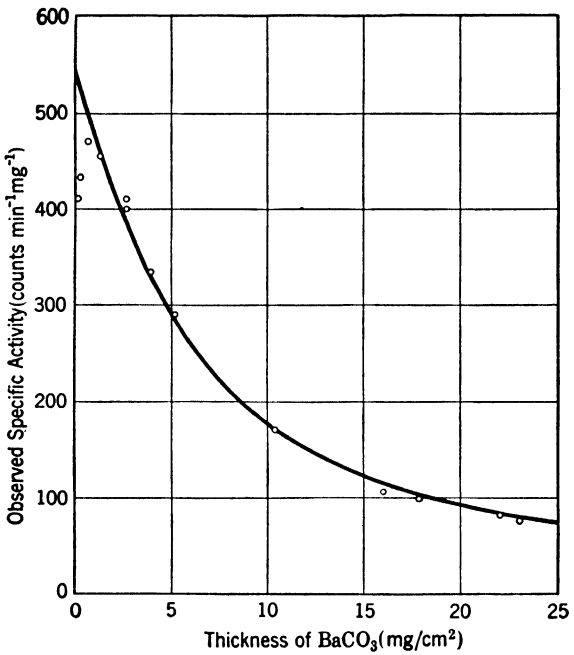


FIGURE X-3. Self-absorption of C^{14} β rays in $BaCO_3$. [Data are taken from A. K. Solomon, R. G. Gould, and C. B. Anfinsen, *Phys. Rev.* **72**, 1097 (1947).] o Experimental points. The solid curve is calculated from $c = c_0 \dfrac{1 - e^{-\mu d}}{\mu d}$, with $\mu = 0.29$ cm^{2}mg^{-1}.

$c = c_0 \dfrac{1 - e^{-\mu d}}{\mu d}$ where c_0 is an arbitrary coefficient. These equations are intended for the geometry which is customary with end window counters in which a flat sample is placed close to the window.

When thicker and thicker samples are prepared from an active material, say, for example, $BaCO_3$ containing C^{14}, the measured counting rate at first increases because of the greater total activity in the sample and then approaches a constant value. This "satu-

ration value" is clearly not a measure of the total activity of the sample, but rather is related to the activity of the amount of sample material in an upper layer no thicker than the particle range R, and is thus a measure of the specific activity of the sample material. This fact is sometimes used to advantage in the measurement of the low-energy tracers; no correction for self-absorption is applied, and it is only necessary to measure the activities of thick samples of the same uniform area and the same chemical composition. Indeed in many tracer experiments the specific activity is more directly significant than the total activity. The minimum sample thickness required for this type of measurement is clearly not more than the range R, and for most practical purposes $0.75R$ is adequate because of the very small relative contribution of the lowest layers and the additional absorption due to the counter window.

In measurements of very thin samples an effect known as "self-scattering" has recently been noticed. A slightly thicker sample may give a counting efficiency higher by several per cent than the infinitely thin sample; this has been attributed to scattering of rays into the detector by the material of the sample, although the observed effects may in part have other causes, such as mechanical loss of sample. Figure X–3 shows the apparent specific activity of $BaCO_3$ precipitates, containing C^{14}, as a function of the sample thickness. The experimental values fit the relation $c = c_0 \dfrac{1 - e^{-\mu d}}{\mu d}$ rather well, except that at very low thicknesses the measured activities fall off (perhaps because the samples become too thin to exhibit self-scattering).

Useful Sample-mounting Techniques. If thin samples are to be used, so that the self-absorption correction is small, it is usually necessary to arrange that they be spread uniformly over the sample-mounting area. If a solution is merely evaporated to dryness in a very shallow cup [4] of the right size the deposit left will probably show very obvious lack of uniformity; most of the

[4] Some workers use shallow flat-bottom porcelain ashing dishes. Very shallow cups stamped from a suitably inert metal foil are better for some purposes. Actually cups are not necessary since a flat disk with a smooth (and possibly greased) edge will hold water. Most often we use microscope cover glasses, support them on smaller bits of cardboard, and carry out the evaporations under an ordinary infrared lamp.

residue will ordinarily be found near the edge. If the active substance is first precipitated and the slurry evaporated, preferably with stirring, a much improved deposit usually results. In another procedure the slurry is added and dried a little at a time. It is sometimes helpful to place a circular piece of cigarette paper, slightly smaller than the dish or disk, on the flat surface; the solution or slurry is allowed to spread over the paper and then to dry; when dry this type of paper weighs about 1 mg per cm^2.

Other methods of preparing samples are sometimes more convenient, especially if the bulk of the deposit or the volume of solution or suspension is large. Filtration on a small Buchner or Gooch filter can give reasonably uniform and very nearly quantitative deposits of precipitates on the filter paper. If the precipitate and filter are washed finally with alcohol [5] the bits of precipitate creeping up the sides of the filter are likely to be washed down; also the paper is then more easily dried, either with or without an ether wash. A glass chimney held tightly against the paper in a Buchner funnel with perforated surface ground flat will help to confine the precipitate to a definite area. Sedimentation of a precipitate followed by washing and drying can be used to give uniform deposits. Special cells with demountable bottoms are employed for this purpose so that the sample may be removed on its mounting for measurement. Sedimentation cells of this type fitted into laboratory centrifuge cups may be used to give harder deposits in very much less time.

Samples prepared in any of these ways should be thoroughly dry before measurement, otherwise the observed activity will grow with time as water evaporates with a consequent reduction in self-absorption of the softer rays. Samples may be found subject to loss of precipitate through powdering when dry; this can be especially troublesome if the active dust should contaminate a measuring instrument. To avoid this effect a few drops of a

[5] The use of alcohol (presumably to lower surface tension) has other applications in the manipulation of samples. For example, when a precipitate is centrifuged down in a semimicro cone some is almost always trapped in the liquid surface (meniscus); addition of a few drops of alcohol on top of the solution followed by another centrifugation will usually bring down most of this precipitate. In the mounting of slurries as described previously a few drops of alcohol as a wash will often clean the residual slurry from the transfer micropipet (a glass tube drawn down to have a long tip less than 1 mm in diameter and fitted with a rubber bulb, like an "eye dropper").

solution of Duco cement in acetone may be used to wet the sample when it is first dried; the concentration of the cement in the acetone solution should be so small that only about 0.1 mg per cm^2 of its dry residue will be left on the sample.

Dry powders may be compacted into disks with a piston-and-cylinder arrangement, or simply pressed smoothly into shallow depressions cut in aluminum or other suitable sample-mounting cards. These techniques cannot be recommended for the preparation of very thin samples for measurements of soft radiations. If the radiation is penetrating, solutions of active materials may be counted directly in a thin-walled glass jacket slipped over a tubular counter, or better in an outer jacket built as a part of a glass counter tube. A dipping counter is arranged to permit immersion of the whole counter (except an end for electric connections) in the solution to be measured. Radiations from relatively thick layers of solutions may be counted reproducibly only if the solution densities are comparable, and if the relative composition of the solutions in terms of elements of various atomic numbers is kept approximately constant, especially for γ-ray measurements (see chapter VII, section C).

If the radioactive element is a metal like copper excellent samples for measurement may be prepared by electrodeposition. Other types of ions may often be deposited by different electrode reactions under suitable conditions. For example, lead may be deposited on an anode as PbO_2 from alkali plumbite solutions. Insoluble hydroxides may be deposited from neutral solutions on cathodes because of the liberation of hydroxide ions there: $H_2O + e^- = \frac{1}{2}H_2 + OH^-$. Insoluble ferrocyanides may form on a cathode from the reduction of some metal ferricyanide solutions. Metal fluorides may be precipitated in adherent form for some metals that can be oxidized or reduced at an electrode from a fluoride-soluble to a fluoride-insoluble state; for example, UF_4 may be deposited in this way and then ignited in air to U_3O_8.

Some specialized sample-spreading techniques have been developed to a high state of the art for the preparation of standard foils, especially of uranium. In one of these a wetting agent, tetraethylene glycol (TEG), is used to improve spreading on evaporation. In this procedure a platinum foil of the right dimension is prepared with a Zapon lacquer border, then a few microliters of TEG per square centimeter are applied, and the uranium

as chloride in dilute hydrochloric acid solution is added. Evaporation is carried out under an infrared lamp, with some rotation of the foil to insure mixing. Finally, the plate is ignited to red heat for a few minutes to convert the deposit to an adherent highly colored layer of U_3O_8. This is rubbed gently with lens paper and then may be weighed to establish the uranium content. This procedure is suitable for the preparation of uranium foils of the order of 0.1 mg per cm^2 and with larger amounts of TEG can be used up to about 1 mg per cm^2. Another technique uses uranyl nitrate dissolved in alcohol (about 50 mg per ml), with about 1 per cent of Zapon lacquer added; this mixture is painted on the platinum or aluminum foil with a brush; each coat is dried, ignited to U_3O_8, and rubbed with lens paper. Thick foils, weighable for uranium content, may be built up by this method. Other elements with alcohol-soluble salts may give satisfactory plates with the same technique. Thin standard boron films are sometimes wanted. These can be prepared by the decomposition of B_2H_6 on hot tungsten at about 900°C under carefully controlled conditions.

EXERCISES

1. The γ-ray activities of two portions of a solution of Co^{60} were found under identical conditions to be: 350 counts per min for a 0.10-cc portion, and 9900 counts per min for a 3.00-cc portion.

(*a*) What would you expect for the counting rate of a 2.00-cc portion?

(*b*) What is the approximate dead time of the Geiger counter? Assume all other resolving times are negligibly small. Neglect self-absorption effects. *Answer:* (*b*) 346 μs.

2. From data on X-ray spectra and X-ray absorption coefficients (in the Chemical Rubber Company Handbook, for example) locate the critical absorbers for the identification of the X rays following K capture (*a*) in A^{37}, (*b*) in Pd^{101}.

3. Calculate the field strength necessary to give a 2.00-cm radius of curvature for the path of a β particle of 0.40 Mev.

4. (*a*) Estimate the half-thickness $d_{1/2}$ for a 1-Mev γ ray in (1) lead, (2) aluminum, (3) beryllium. (*b*) Make the same estimates for a 50-kev γ ray in aluminum and beryllium.

5. A sample of I^{131}, contained in a 6.0-mg precipitate of AgI uniformly distributed over a circular area 1.00 cm in diameter, was placed on the

fourth (lowest) step of the sample holder described on page 219 and counted with an end-window (mica 3 mg per cm^2) Geiger counter with cathode inner diameter 1⅛ inch. It gave a net counting rate of 2000 counts per min. Addition of a few milligrams per square centimeter of aluminum absorber gave a reduction in measured activity corresponding to a mass absorption coefficient $\mu/\rho = 25\ cm^2\ g^{-1}$. Estimate as well as you can the absolute strength of this sample (in rutherfords).

Answer: We estimate 2.4×10^{-3} rd.

6. A sample of 6.6-min Cb^{94} is counted in identical geometrical positions, first with a neon-filled thin-window counter, then with an otherwise identical krypton-filled counter. When corrected for the decay between measurements, the counting rate in the second counter is about 30 per cent higher than that in the first. How can you explain this fact?

7. If you wished to count the Rb^{88} β rays without interference from the Rb^{86} or Rb^{87} β rays you might interpose an aluminum absorber. (*a*) What thickness (in grams per square centimeter or milligrams per square centimeter) of aluminum would be just sufficient to stop the Rb^{86} β's? (*b*) By what factor would this absorber reduce the intensity of the Rb^{88} β's? (The mass absorption coefficient for this radiation you may estimate from the very rough rule that the range is about seven times the half-thickness.)

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XI. IDENTIFICATION, CONCENTRATION, AND ISOLATION OF RADIOACTIVE SPECIES

A. GENERAL REMARKS

In previous chapters we have discussed the production of radioactive species, the radiations emitted by them, and the instruments and techniques for their detection and measurement. Before we can proceed to consider some of the chemical applications of radioactive materials we should give some attention to the problem of identifying, purifying, and isolating a given radioactive species following its production in a nuclear reaction.

In this connection the radiochemist may be confronted with one of two tasks. He may need to prepare a known reaction product free from other radioactive contaminants and sometimes free from certain inactive impurities and in a specified chemical form for use in subsequent experimentation. Or he may wish to identify a hitherto unknown or unidentified radioactive species as to its atomic number, mass number, half-life, and radiation characteristics. In both cases chemical separations are usually required for two reasons: (1) In almost any nuclear bombardment more than one type of reaction occurs, and, therefore, the reaction products have to be separated; (2) impurities present in the target material usually give rise to radioactive products. Apart from the ordinary chemical impurities, further contamination is often introduced in the bombardment procedure; particularly in cyclotron bombardments, where for cooling purposes the target material is usually soldered, pressed, or bolted onto a copper or brass backing plate, transmutation products of the sample container, backing material, solder, and fluxes must be considered.

In a slow-neutron bombardment the only type of reaction produced in almost any target element is the n, γ reaction. Therefore, if an element of sufficient purity is bombarded with slow neutrons, chemical separations are often not required. However, in this case the radioactive product is isotopic with the target, and it is sometimes desirable to free the product isotope from the

bulk of the target material in order to obtain high specific activities. Special techniques developed for this purpose are discussed in section D.

The chemical techniques used for the separation and isolation of radioactive species are essentially the same whether an unknown nuclide is to be identified or a known product to be prepared. In the former case chemical identification is usually not sufficient, and we shall defer a discussion of the chemical separation techniques to section C. First we take up the question of how to obtain the additional information necessary for the unambiguous identification of radioactive species.

B. IDENTIFICATION OF NUCLEAR-REACTION PRODUCTS

Cross Bombardments. For the purposes of this section we assume that the atomic number of a given radioactive species can be determined by chemical analysis and turn our attention to methods for the assignment of the correct mass number. The only case in which the mass number of a reaction product is practically uniquely determined in a single bombardment is the slow-neutron activation of a single nuclear species. For example the slow-neutron bombardment of arsenic (with the single stable species As^{75}) produces a 26.8-hr β-emitting arsenic isotope which is, therefore, readily assigned to As^{76}.

One can frequently make a mass assignment by investigating whether or not the radioactive species being studied is formed in a number of different types of bombardments; this is called the method of cross bombardments. For this purpose target elements as well as projectiles may be varied. In each bombardment the possible products are limited by the stable isotopes of the target element and by the types of reactions possible with the projectile and energy used. As an illustration consider some radioactive isotopes of strontium; figure XI–1 displays the stable nuclides in the region of strontium. Slow-neutron activation of strontium produces strontium isotopes with 2.7-hr and 54-day half-lives; either or both of these activities might be assigned to any of the isotopes Sr^{85}, Sr^{87}, Sr^{88}, Sr^{89}, since they are produced by neutron capture from the stable strontium isotopes. The fact that the same two activities are produced by fast neutrons (say 15 Mev) in zirconium, presumably by n, α reactions, elimi-

nates Sr^{85}. The fast-neutron bombardment of yttrium (say with 10- or 15-Mev neutrons) produces only the 54-day strontium, presumably by n, p reaction, and this activity is therefore assigned to Sr^{89}. The 2.7-hr activity is then to be assigned to an isomeric state of Sr^{87} or Sr^{88}. The fact that 2.7-hr strontium is also produced by proton bombardment of rubidium by p, n reaction leads to its assignment to Sr^{87}*.

In the interpretation of the results of cross bombardments the relative abundances of isotopes in the target elements often have to be considered. For example, failure to observe a certain reac-

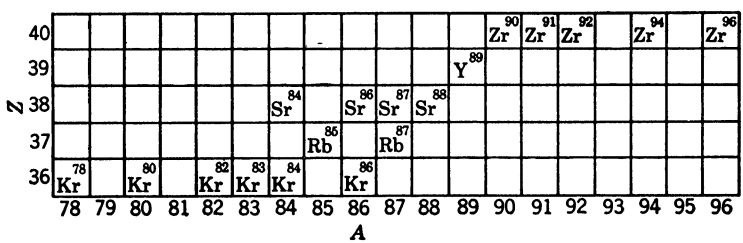


FIGURE XI–1. Naturally occurring nuclear species in the region of strontium.

tion product may simply be due to low abundance of the isotope from which that product could have been made.

Effects of Bombarding Energies and Bombardment Times. The energies of the bombarding particles used in cross bombardments are very important in the analysis of the results. An excitation function may help very much to determine the type of reaction involved, through comparison with similar excitation functions for known reactions (see, for example, the α-particle reactions of silver discussed in chapter III, page 64). Even a knowledge of the energy of the bombarding particle together with an approximate yield of the product can usually be of value for the mass assignment. For example, if a certain activity of element $Z + 1$ is produced in good yield in the bombardment of element Z with 5-Mev deuterons, the reaction can hardly be anything but a d, n reaction; if the deuteron energy had been 20 Mev, a $d, 2n$ reaction or perhaps a $d, 3n$ reaction might more likely have been responsible. Referring again to figure XI–1, we see that the 2.7-hr strontium cannot be expected to result from the bombardment of rubidium with 5-Mev deuterons if it is Sr^{87}, but it could be formed in such a bombardment if it were Sr^{88}.

In the case of neutron-induced reactions the effect of neutron energy on the possible reactions was discussed in chapter III, section C. Thermal neutrons produce, in general, only n, γ reactions. For neutrons of 1 or 2 Mev, capture cross sections are usually much smaller than for thermal neutrons, but formation of excited states by n, n reactions becomes more important at those energies. At 8 or 10 Mev $n, 2n$ reactions may set in. Thus in our example the yield of 54-day strontium in neutron bombardments of strontium would be expected to drop off more with an increase in neutron energy from say 1 ev to 15 Mev than that of the 2.7-hr Sr^{87}, because the former is made by n, γ reaction only, whereas the latter is produced in the reactions Sr^{86} (n, γ), Sr^{87} (n, n), and Sr^{88} $(n, 2n)$.[1] It is evident that with increasing bombardment energies the mass assignment problems become increasingly difficult because of the greater variety of possible reactions.

If several radioactive isotopes of the same element are produced in one bombardment the study of the properties of one is often greatly hindered by the presence of others. When the half-lives are sufficiently different, this difficulty can be minimized by proper choice of the bombardment and "cooling" times. In our example of the strontium isotopes it may be desirable to bombard one sample with slow neutrons for many weeks to build up sufficient 54-day activity; the 2.7-hr activity can then be allowed to decay practically completely in a day or two before the 54-day activity is studied. Another sample may be bombarded for about 2 or 3 hr which builds up the 2.7-hr activity to about 50 per cent of its saturation value while producing only a very small fraction of the saturation amount of the long-lived isotope (see chapter V, section B, for the quantitative treatment).

Beam Contamination and Secondary Reactions. In connection with the subject of cross bombardments it should be noted that extraneous bombarding particles or secondary particles produced

[1] It should be noted that no fast-neutron irradiation can be completely free of slow neutrons because in traversing any matter some neutrons are always slowed down. Cadmium shielding is usually employed to prevent thermal neutrons from reaching a sample. The difference between the activities produced with and without cadmium is often taken as an approximate measure of the thermal-neutron effect. The cadmium shield does not completely eliminate n, γ reactions because they may occur with appreciable cross sections at energies above the cadmium resonance.

by bombardment of the targets may sometimes give rise to nuclear reactions to an extent which can interfere with the recognition of the primary-reaction products. For example, α-particle beams in cyclotrons are commonly contaminated with small amounts of deuterons (from residual deuterium in the ion source); because deuteron cross sections are often much higher than α-particle cross sections the deuteron-produced activities may actually exceed in intensity those from α particles.

The neutrons produced in the bombardment of targets with charged particles or γ rays usually give rise to nuclear reactions in the target material. The products of these reactions may be confused with those that are found or expected to be produced by the primary bombarding particle. For example, in the deuteron bombardment of a thick sodium target to make Na^{24} by the d, p reaction, the Na^{24} activity is found at depths beyond the range of the deuterons, because it can be produced there by n, γ reaction. Even protons and probably α particles of sufficient energy to cause secondary reactions may be produced in bombardments with high-energy projectiles.

Use of Isotope Separations. In many cases the mass number of a radioactive species is uncertain because it is not known from which isotope of the target element the activity has been produced. The bombardment of separated isotopes, or of isotopic mixtures sufficiently enriched in some isotope, is, therefore, of great advantage. Enrichment or impoverishment of an isotope by a factor of two or even less may be sufficient for this purpose; by comparing the yield of the activity of interest from samples of normal and altered isotopic composition one can deduce the origin of the activity. For a long time the assignment of the 37-min chlorine activity produced by slow neutrons or by deuterons in chlorine (stable isotopes Cl^{35} and Cl^{37}) was uncertain and could not be readily determined by cross bombardments; but when it was shown that this activity was not produced by slow-neutron bombardment of a sample of almost pure Cl^{35} the 37-min period could be assigned to Cl^{38} rather than Cl^{36}. Enriched lead isotopes have recently been used to aid in the assignments of a number of radioactive isotopes of lead, bismuth, and polonium.

Until recently, separated or enriched stable isotopes of most elements were not available in appreciable quantities, and they are only now coming into extensive use for the identification of

radioactive products. Some fifty different enriched stable isotopes have now been made available in milligram or gram quantities through the United States Atomic Energy Commission; some of these have already been used in assignment studies, and the technique will undoubtedly be employed much more in the future.

Isotope separation can be used in an even more direct way for the identification of radioactive species: the reaction products themselves may be subjected to an isotope separation process. This procedure was used after the bombardment of separated nickel isotopes to establish the assignment of a 1.75-hr period to Co^{61} by calutron analysis. The mass numbers of quite a number of long-lived activities, particularly fission products and rare earths, have been established by an interesting modification of mass-spectrographic analysis. The material containing the active isotope of interest is sent through an ordinary mass-spectrograph, and the ions are collected on a photographic plate. After development that plate is placed face to face with another unexposed plate called the transfer plate; the β particles from a radioactive isotope deposited along a narrow line on the first plate will blacken the transfer plate, and from the position of the exposed line on the transfer plate the mass number of the active isotope can be deduced. By exposing successive transfer plates to the first plate at various times and in each case finding the exposure time necessary to get the same degree of blackening, one can obtain a rough check on the half-life of the activity. To establish the half-life more accurately the area of the original plate on which the activity is located can be counted over a period of time. The mass-spectrographic technique is limited to half-lives longer than about 1 hr.

Target Material and Effect of Impurities. When the product of a particular nuclear reaction is to be studied, it is important that the target not contain other elements from which the same product might result. As an obvious example, in a study of the production of 34-hr bromine (Br^{82}) by neutron bombardment of rubidium one would not wish to use rubidium bromide as a target. For the same reason impurities of neighboring elements, such as iridium impurity in an osmium target, may lead to misinterpretation of results. In general, it is advisable to use free elements as targets; however, sometimes the bombardment of a compound is indicated, for example, if the element is too reactive, or in a physical

state unsuitable for bombardment, or if the dissolving of the element would make a slow step in the subsequent chemical-separation procedure.

As was pointed out at the beginning of this chapter, the presence of some impurities in a target is in most cases unavoidable. If the impurities present are known, then the chemical separation procedure can be designed to separate the products of interest from the products likely to result from the bombardment of the impurities. The purity requirements for the target material may become very exacting if a reaction of low cross section is studied, for then the expected product yield is small, and care must be taken that comparable amounts of the same species are not produced from an impurity by a much more probable reaction. For example, the formation of Mg^{23} from Al^{27} by $\gamma, p3n$ reaction could be studied only with aluminum very free of magnesium, because the Mg^{24} (γ, n) Mg^{23} reaction has a much larger cross section.

Importance of Decay Relationships and Radiations. Both in the study of new species and in the isolation of known tracer materials the half-life is generally considered the chief characteristic of a radioactive isotope. Complete resolution of the experimentally found decay curve into its components is, therefore, usually desirable, and this may require reproducible activity measurements over long periods of time (months or even years). Some special techniques used in the determination of half-lives have already been discussed in chapter V, section E.

If a radioactive product of a nuclear reaction decays into another radioactive product, the genetic relationship may be investigated by studies of the growth and decay curves of fractions chemically separated at successive times. An understanding of genetic relationships often helps in the assignment of the activities. This is particularly important in the fission-product decay chains. For example the fission product chain of mass 89 was identified with that mass number because its last active member was shown to be the 54-day Sr^{89} already discussed. The decay chain containing 275-day cerium was assigned the mass number 144 by mass-spectrographic determination of the mass of that long-lived cerium.

Half-life and atomic number are frequently not sufficient to characterize a radioactive species. It happens rather often that

two isotopes of the same element have not very different half-lives. In that case the isotopes can usually be distinguished by the types and energies of the radiations they emit. We may use again our example of the strontium isotopes; an investigation of the radiations emitted by the slow-neutron-bombarded strontium sample after the 2.7-hr period has decayed reveals, in addition to the 1.5-Mev β^- particles of the 54-day Sr^{89}, some γ rays and characteristic rubidium X rays. On closer examination these last two radiations are found to follow a 65-day half-life. The 54-day and 65-day periods could certainly not be resolved in a gross decay curve of the neutron-bombarded sample. The presence of rubidium X rays shows that the 65-day isotope decays to rubidium by β^+ emission or K capture; no positrons are observed, and the process must be an electron capture. From the evidence the assignment is probably to Sr^{85} or Sr^{87}; the latter is ruled out by the fact that the 65-day species is not formed by n, α reaction from Zr^{90}. Proton bombardment of rubidium produces the 65-day period (but not the 54-day Sr^{89}) as well as the 2.7-hr Sr^{87}* already discussed, and in addition a 70-min strontium which also fails to appear in the fast-neutron bombardment of zirconium.

Using these four strontium activities as examples we shall illustrate how a study of the mode of decay of an isotope helps in its assignment. The fact that the 54-day isotope emits β^- particles rules out its assignment to any mass number less than 89: a strontium nucleus of mass 88 or less would by β^- decay move away from rather than toward the stability region.[2] A study of the radiations from the 70-min and 2.7-hr activities reveals that both emit strontium X rays, indicating that these periods are associated with isomeric transitions. The fact that the 65-day isotope decays by electron capture does not allow its assignment to any mass number greater than 87. From the facts listed in this and in the previous paragraph the 65-day and 70-min periods can both be assigned to Sr^{85}; the 70-min period is associated with an isomeric transition to the lower state, which in turn decays by K capture to Rb^{85} with a 65-day half-life. Figure XI–2 is identical with figure XI–1 except that the strontium activities discussed are now entered at their proper places.

[2] This type of restriction is without known exception; however, it is not absolute—a nucleus in an excited state could transform to an unstable nucleus which then returns to the ground state of the first nucleus.

On the basis of arguments similar to the ones just presented the modes of decay of active isotopes can generally be used as an aid in their assignment. Some attempts have been made to correlate not only modes of decay but also half-lives and decay energies with the positions of isotopes with respect to the stability region. Semiquantitative predictions based on binding-energy formulas like equation II–4 can be useful in the interpretation of experimental data.

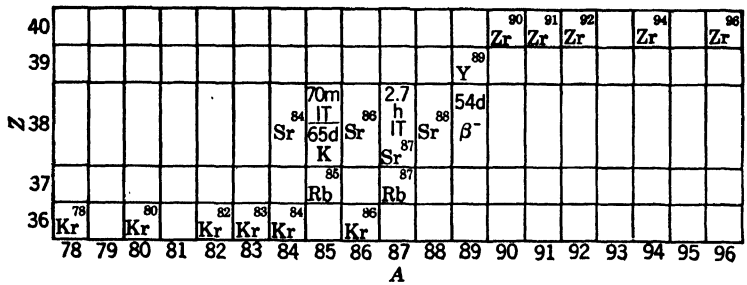


FIGURE XI–2. The four strontium activities discussed in the text are shown at their proper places (with half-lives and modes of decay) in the chart of naturally occurring nuclear species.

C. CHEMICAL ISOLATION OF NONISOTOPIC SPECIES

Comparison with Ordinary Analytical Practice. In many respects the chemical separations which the radiochemist carries out on irradiated targets are very similar to ordinary analytical procedures. However, there are a number of important differences. One of these is the time factor which is often introduced by the short half-lives of the species involved. An otherwise very simple procedure such as the separation of two common cations may become quite difficult when it is to be performed, and the final precipitates are to be dried and mounted, in a few minutes. Where the usual procedures involve long digestions, slow filtrations, or other slow steps, completely different separation procedures must be worked out for use with short-lived activities. Ingenious chemical isolation procedures taking as little as 30 sec have been developed.

In radiochemical separations, at least those subsequent to bombardments with projectiles of moderate energies, we are usu-

ally concerned with several elements of neighboring atomic numbers. Thus the procedures given in complete schemes of qualitative analysis can often be modified and shortened. On the other hand, the separation of neighboring elements often presents considerable difficulties as can readily be seen by considering such groups as Ru, Rh, Pd or Hf, Ta, W or any sequence of neighboring rare earths. In the cases of very high-energy reactions and of fission the products are spread over a wide range of atomic numbers. In these cases the separation procedures either become more akin to general schemes of analysis or, more frequently, are designed for the isolation of a single element away from all the others; the latter type of procedure is required particularly when a short-lived substance is to be isolated, and for such cases many specialized techniques have been developed.

High yields in radiochemical separations are not always of great importance, provided the yields can be evaluated. It may be more valuable to get 50 per cent (or perhaps even 10 per cent) yield of a radioactive element separated in 10 min than to get 99 per cent yield in 1 hr (this is certainly so if the activity has a half-life of 10 or 20 min). High *chemical* purity may or may not be required for radioactive preparations, depending on their use; for identification and study of radioactive species and for many chemical tracer applications it is not important; for most biological work it is. On the other hand *radioactive* purity is usually required and often has to be extremely good.

Some specific effects of the radiations from radioactive substances on the separation procedures may be noted. In case of very high activity levels (say 10^5 rd or more of β particles per milliliter of solution) the chemical effects of the radiations, such as decomposition of water and other solvents, and heat effects, may affect the procedures. However, this is generally not so important as the fact that even at much lower activity levels, especially in the case of γ-ray emitters, the person carrying out the separation receives dangerous doses of radiation unless protected by shielding or distance.[3] For this reason it is often necessary to carry out separations behind lead shields and to

[3] Remember that radiation dosage falls off as the inverse square of the distance from the source. The following are the dosages in milliroentgens per hour received at 10 cm distance from 1 rd of each of several typical γ emitters: Na^{24} 5.2; Mn^{54} 1.3; Fe^{59} 1.8; Co^{60} (5.3 year) 3.5; Br^{82} 4.1; I^{128} 0.05; I^{130} 3.4.

perform operations with the use of tongs and other tools; for very high activity levels (say in excess of 10^5 or 10^6 rd of γ activity) more elaborate remote-control methods are necessary. It is obvious that separation procedures are more difficult under these conditions and in many cases have to be modified considerably to adapt them for remote-control operation. References to discussions of the safe handling of radioactive materials and of appropriate health-protection measures may be found at the end of this chapter.

Carriers. The mass of radioactive material produced in a nuclear reaction is generally very small. Notice, for example, that 1 rd of 37-min Cl^{38} weighs about 2×10^{-13} g, 1 rd of 54-day Sr^{89} weighs about 10^{-9} g, and 1 rd of 6000-year C^{14} weighs about 6×10^{-6} g. Thus the substance to be isolated in a radiochemical separation may often be present in a completely impalpable quantity.[4] It is clear that ordinary analytical procedures involving precipitation and filtration or centrifugation may fail for such minute quantities. In fact, solutions containing the very minute concentrations of solutes which can be investigated with radioactive tracers behave in many ways quite differently from solutions in ordinarily accessible concentration ranges; this subject is treated in the next chapter. Usually some inactive material isotopic with the radioactive transmutation product is deliberately added to act as a carrier for the active material in all subsequent chemical reactions. Most often it is not sufficient to add carrier only for the particular transmutation product to be isolated; frequently it is necessary to add carriers also for other activities which are known or assumed to be formed, including those which derive from target impurities. It is worth noting also that certain contaminating activities such as P^{32} (14.3 days) and Na^{24} (14.8 hr) appear almost inevitably on all targets in cyclotrons which are often used for phosphorus and sodium bombardments. A special step for the removal of P^{32} such as the precipitation of $BiPO_4$ in 1 *N* nitric acid is therefore often necessary.

It is in many cases not necessary to add carriers for all active species present, because several elements may behave sufficiently alike under given conditions so that traces of one will be carried

[4] Actually the mass of an element formed in a nuclear reaction is often exceeded by that of the inactive isotopes of the same element present as an impurity in the target and in the reagents used in the separation procedure.

by macroscopic quantities of another. For example, an acid-insoluble sulfide such as CuS can usually be counted on to carry traces of ions such as Hg^{++}, Bi^{+++}, Pb^{++}, which also form acid-insoluble sulfides. On the other hand, since many precipitates (such as $BaSO_4$ or $Fe(OH)_3$) tend to occlude or adsorb many foreign substances, it is usually necessary to add carriers not only for ions to be precipitated but also for ions to be held in solution when other ions are precipitated. For example, if a zinc activity is to be separated from a ferric solution by ferric hydroxide precipitation with excess ammonia, all the zinc will not be left in solution unless zinc carrier is present. The carrier in such cases is sometimes referred to as hold-back carrier. We shall later discuss cases where carriers are unnecessary.

We have mentioned before that extreme radioactive purity is often very important. Frequently the desired product has an activity that constitutes only a very small fraction of the total target activity; yet this product may be required completely free of the other activities. Such extreme purification is usually quite readily attained by repeated removal of the impurities with successive fresh portions of carrier, until the fractions removed are sufficiently inactive. This so-called "washing-out" principle may be illustrated by the separation of a weak cobalt activity from radioactive copper contamination. Cobalt and copper carriers are added to a 0.3 *N* HCl solution of the activities, CuS is precipitated, and filtered or centrifuged off, excess H_2S removed by boiling, then fresh copper carrier added to the filtrate and the procedure repeated until a final CuS precipitate no longer shows an objectionable amount of activity. The same principle can be applied to other than precipitation reactions. Radioactive iron impurity might be removed by repeated extraction of ferric chloride from 6 *N* HCl into ether, with fresh portions of $FeCl_3$ carrier added after each extraction. In applying the washing-out method one must, of course, make sure that the desired product is not partially removed along with the impurity in each cycle. If the washing out works properly, the activities of successive impurity fractions should decrease by large and approximately constant factors, provided the conditions in each step are about the same.

In order that an added inactive material serve as a carrier for an active substance, the two must generally be in the same chemical form. For example, inactive iodide can hardly be expected

to be a carrier for active iodine in the form of iodate ion; sodium phosphate would not carry radioactive phosphorus in elementary form. The chemical form in which a transmutation product emerges from a nuclear reaction is usually very hard to predict and has been investigated so far only in a very small number of cases. However, it is often possible to treat a target in such a way that the active material of interest is transformed to a certain chemical form. For example, if a zinc target is dissolved in a strongly oxidizing medium (say, HNO_3, or $HCl + H_2O_2$), any copper present as a transmutation product is found afterwards in the Cu^{++} form. If there is any uncertainty about the chemical form of the transmutation product—as to its oxidation state or presence in some complex or undissociated compound, for example—the only method which can be relied on to avoid difficulties is the addition of carrier in the various possible forms and a subsequent procedure for the conversion of all of these into one form. To go through such a procedure prior to the addition of carrier may not be adequate. In fact, it appears that it may not always be sufficient to add the carrier element (say iodine) in its highest oxidation state (IO_4^-) and carry through a reduction to a low oxidation state (I_2); in the case of the iodine compounds this procedure does not seem to reduce all the active atoms originally present in intermediate oxidation states.

So far we have not spoken of the amounts of carriers used. For manipulative reasons it is often convenient to use about 10 or 20 mg of each carrier, and less than about 1 mg is used only rarely; in separations from large bulks of target material larger amounts of carrier (perhaps 100 to 500 mg) are sometimes useful.[5] The amount of carrier frequently has to be measured at various stages of a chemical procedure to determine the chemical yield in the different steps, or at least in the over-all process. In such cases very small amounts of carrier are inconvenient. On the other hand, it is necessary to keep the quantities of carrier small in the preparation of sources of high specific activity. The specific activity of a sample of an element is sometimes expressed as the ratio of the number of radioactive atoms to the total number of atoms of the element in the sample; more conveniently it is often expressed in terms of the disintegration rate (say in ruther-

[5] In a radiochemical laboratory it is convenient to have carrier solutions for a large number of elements on hand. These may, for example, be made up to contain 1 or 10 mg of carrier element per milliliter, or possibly 1 mg per drop.

fords) per unit weight (gram or milligram). High specific activities are essential particularly in many biological and medical applications of radioactive isotopes, and also are often very desirable in samples to be used in physical measurements or chemical tracer studies, to insure small self-absorption or to permit high dilution factors.

Sometimes it is possible to prepare samples of very high specific activities by the use of a nonisotopic carrier in the first stages of the separation; this may later be separated from the active material. In the isolation of radioyttrium (105-day Y^{88}) from deuteron-bombarded strontium targets, ferric ion can be used as a carrier for the active Y^{+++}; ferric hydroxide is then precipitated, centrifuged, washed, redissolved and, after the addition of more strontium as hold-back carrier, it is reprecipitated several more times to free it of strontium activity. Finally the ferric hydroxide which carries the yttrium activity is dissolved in 6 *N* HCl, and ferric chloride is extracted into ether, leaving the active yttrium in the aqueous phase almost carrier-free. The use of nonisotopic carriers is particularly important in cases where no stable isotopes of the active material have been found in nature; these cases are treated in some detail in the next chapter. Criteria for the choice of nonisotopic carriers are also discussed there in connection with the behavior of substances at very small concentrations.

Not all chemical procedures require the use of carriers. Particularly procedures which do not involve solid phases may sometimes be carried out at tracer concentrations without the addition of carriers. Because of the great importance of high specific activities, considerable work has been done on the preparation of carrier-free sources of many radioactive species. In the course of the following brief discussion of the various types of separation techniques we shall, therefore, point out those which lend themselves to the production of carrier-free preparations.

Chemical Separation Techniques. In most radiochemical separations, as in conventional analytical schemes, precipitation reactions play a predominant role. The chief difficulties with precipitations arise from the carrying down of other materials. Some precipitates such as manganese dioxide and ferric hydroxide are so effective as "scavengers" that they are sometimes used deliberately to carry down foreign substances in trace amounts. Some other precipitates, such as rare-earth fluorides precipitated in acid solution, cupric sulfide precipitated in acid solution, or

elementary tellurium brought down by reduction with SO_2, have little tendency to carry substances not actually insoluble under the same conditions, and, therefore, can sometimes be brought down without the addition of hold-back carriers for activities that are to be left in solution. Most precipitates have an intermediate behavior in this regard.

As mentioned before, adsorption on precipitates has been used to effect separations of tracer quantities. Adsorptions on the walls of glass vessels and on filter paper, which are sometimes bothersome, have also been put to successful use in special cases. Carrier-free yttrium activity has been quantitatively adsorbed on filter paper from an alkaline strontium solution at yttrium concentrations at which the solubility product of yttrium hydroxide could not have been exceeded.

Ion-exchange Separations. An exceedingly useful separation technique closely related to adsorption chromatography has recently been developed for use both with and without carriers. This technique involves the adsorption of a mixture of ions on an ion-exchange resin followed by selective elution from the resin. The resin (such as the Amberlite IR-1 or Dowex-50) is usually a synthetic polymer containing free sulfonic acid groups; cations are adsorbed on it by replacing the hydrogen ions of these acid groups. The strength of the resin-ion bond increases with increasing ionic charge and decreasing (hydrated!) ionic radius. In practice a solution containing the ions to be separated is run through a column of the finely divided resin, and conditions (column dimensions, solution volume, concentration, and flow rate) are chosen such that the ions are adsorbed in a narrow region near the top of the column. Then an eluting solution, usually containing a complexing agent (such as oxalic acid or citric acid) which forms complexes of different stability with the various ions, is run through the column. There exists then a competition between the resin and the complexing agent for each ion, and if the column is run close to equilibrium conditions each ion will be exchanged between resin and complex form many times as it moves down the column.[6] The number of times an ion is ad-

[6] Slow flow rate, high resin-to-ion ratio, and fine resin particle size favor close approach to equilibrium. In practice, a compromise has to be made between high separation efficiency on the one hand and good yield and speed on the other.

sorbed and desorbed on the resin in such a column is analogous to the number of theoretical plates in a distillation column. The rates with which different ionic species move down the column under identical conditions are different, because the stabilities of both the resin compounds and the complexes vary from ion to ion; separations are particularly efficient if both these factors work in the same direction, that is, if the complex stability increases as the metal-resin bond strength decreases. As the various adsorption bands move down the column their separations increase, until finally the ion from the lowest band appears in the effluent. The various ions can then be collected separately in successive fractions of the effluent.

The ion-exchange column technique was developed and has proved particularly useful for fission-product separations. Many of the carrier-free fission-product activities sold by the United States Atomic Energy Commission are isolated by this method. The most striking application of ion-exchange columns is in the separation of rare earths from each other, both on a tracer scale and in gram or hundred-gram lots. The eluting solution in this case may be 5 per cent citric acid solution buffered with ammonia to a *p*H somewhere between 2.5 and 3.0. The rare earths are eluted in reverse order of their atomic numbers, and yttrium falls between dysprosium and holmium. Very clean separations can be obtained, with impurities in some cases reduced to less than one part per million. By continuously recording the specific activity of the effluent solution as a function of time one obtains separate sharp peaks for the activities of the various rare earths when a mixture of rare-earth radioactivities is run through the column. This method led to the definite assignment of several decay periods to isotopes of element 61.

Volatilization. Other separation methods avoiding the difficulties inherent in precipitations have frequently been used in radiochemical work. Among these are volatilization, solvent extraction, electrodeposition, and leaching. In special cases all these techniques lend themselves to the preparation of carrier-free tracers (as illustrated in chapter XII). Many volatile substances have been separated from less volatile ones by distillation. For example, after the solution of deuteron-bombarded tellurium in nitric acid, active iodine can be distilled out of the nitric acid solution at the boiling point. Radioactive noble gases can be swept out

of aqueous solutions with some inert gas. The volatility of such compounds as $GeCl_4$, $AsCl_3$, $SeCl_4$ can be used to effect separations from other chlorides by distillation from HCl solutions. Similarly OsO_4 can be distilled from concentrated HNO_3, and RuO_4 from $HClO_4$ solutions.

Solvent Extraction. Distribution between two immiscible solvents has been used to effect a number of radiochemical separations. Ferric chloride and gallium chloride, for example, are extracted quite efficiently (>95 per cent) into ether out of 6 *N* HCl, whereas most other chlorides stay almost quantitatively in the aqueous phase. Similarly gold nitrate and mercuric nitrate can be extracted into ethyl acetate from nitric acid solutions. The extraction of uranyl nitrate into ether often serves as a convenient method for the separation of fission products from the bulk of uranium. The basic acetate of beryllium is very soluble in chloroform, and a chloroform solution of this compound may be shaken with water to separate beryllium from many impurities.

Electrodeposition. Electrolysis or electrochemical deposition may be used either to plate out the active material of interest or to plate out other substances leaving the active material in solution. For example, it is possible to separate radioactive copper from a dissolved zinc target by an electroplating process. Carrier-free radioactive zinc may be obtained from a deuteron-bombarded copper target by solution of the target and electrolysis to remove all the copper. Sometimes care must be taken to avoid loss of active material due to electrochemical displacement; in the example of a deuteron-bombarded zinc target from which copper is to be separated after solution in HCl, it is essential that all the zinc be dissolved; as long as solid zinc is present the copper deposits on the zinc surface.

Occasionally it may be possible to leach an active product out of the target material. This has been done successfully in the case of neutron- and deuteron-bombarded magnesium oxide targets; radioactive sodium is separated rather efficiently from the bulk of such a target by leaching with hot water.

D. CHEMICAL CONCENTRATION OF ISOTOPIC SPECIES: THE SZILARD-CHALMERS PROCESS

Principle of the Method. The methods already discussed for preparation of sources of high specific activity apply only if the

radioactive product is not isotopic with the target material. If target and product are isotopic the problem is much more difficult; isotope separation by one of the usual physical methods is possible in principle but at present usually not practical for the preparation of tracer activities. However, a chemical separation is sometimes possible. In 1934 L. Szilard and T. A. Chalmers showed that after the neutron irradiation of ethyl iodide most of the iodine activity formed could be extracted from the ethyl iodide with water; they used a small amount of iodine carrier, reduced it to I^- and finally precipitated AgI. Evidently the iodine–carbon bond was broken when an I^{127} nucleus was transformed by neutron capture to I^{128}. This type of process has since been used to concentrate the products of a number of n, γ reactions, and of some γ, n and $n, 2n$ reactions, and is referred to as the Szilard–Chalmers process. Three conditions have to be fulfilled to make a Szilard–Chalmers separation possible. The radioactive atom in the process of its formation must be broken loose from its molecule; it must not recombine with the molecular fragment from which it separated, nor rapidly interchange with inactive atoms in other target molecules; and a chemical method for the separation of the target compound from the radioactive material in its new chemical form must be available.

Most chemical bond energies are in the range of 1 to 5 ev (20,000 to 100,000 cal per mole). In any nuclear reaction involving heavy particles either entering or leaving the nucleus with energies in excess of 10 or 100 kev the kinetic energy imparted to the residual nucleus far exceeds the magnitude of bond energies.[7] In the case of thermal-neutron capture, where the Szilard–Chalmers method has its most important applications, the incident neutron does not impart nearly enough energy to the nucleus to cause any bond rupture. But neutron capture is always followed by γ-ray emission, and the nucleus receives some recoil energy in this process. A γ ray of energy E_γ has a momentum $p_\gamma = E_\gamma/c$; to conserve momentum the recoiling atom must have an identical momentum, and, therefore, the recoil energy $E_r = p_\gamma^2/2M = E_\gamma^2/2Mc^2$, where M is the mass of the atom. For M in atomic mass units

[7] For reactions other than n, γ, particularly for d, p reactions, the Szilard–Chalmers technique is not very useful because the energy dissipated by the incident radiation in the target is so great that many inactive molecules are also disrupted. The chemical effect of radiation—the field of radiation chemistry—is not treated in this book.

and the energies in millions of electron volts we have

$$E_r = \frac{E_\gamma^2}{1862\,M}. \qquad \text{(XI-1)}$$

Table XI-1 shows values of E_r for a few values of E_γ and M. Neutron capture usually excites a nucleus to about 6 or 8 Mev,

TABLE XI-1

RECOIL ENERGIES IN ELECTRON VOLTS IMPARTED TO NUCLEI BY γ RAYS OF VARIOUS ENERGIES

M	E_γ = 2 Mev	E_γ = 4 Mev	E_γ = 6 Mev
20	107	430	967
50	43	172	387
100	21	86	193
150	14	57	129
200	11	43	97

and a large fraction of this excitation energy is dissipated by the emission of one or more γ rays; unless all the successive γ rays emitted in a given capture process have low energies (say below 1 or 2 Mev), which is a relatively rare occurrence, the recoiling nucleus receives more than sufficient energy for the rupture of one or more bonds. Of course, it is not the entire recoil energy but something more like its component in the direction of a bond that should be compared with the bond energy; furthermore, the momenta of several γ rays emitted in cascade and in different directions may partially cancel each other. There is no evidence that two capture γ rays in cascade are preferentially emitted in opposite directions, and momentum cancelation is, therefore, hardly expected to reduce the probability of bond rupture by a very large factor. In most n, γ processes the probability of rupture is certainly very high.

The second condition for the operation of the Szilard–Chalmers method requires at least that *thermal* exchange be slow between the radioactive atoms in their new chemical state and the inactive

atoms in the target compound. The energetic recoil atoms may undergo exchange more readily than atoms of ordinary thermal energies. These exchange reactions and other reactions of the high-energy recoil atoms (often called "hot atoms") determine to a large extent the separation efficiencies obtainable in Szilard–Chalmers processes. Hot-atom reactions are considered briefly after a discussion of some examples of Szilard–Chalmers separations.

Illustrations. The largest amount of work in the field of Szilard–Chalmers separations has been done on halogen compounds. Many different organic halides (including CCl_4, $C_2H_4Cl_2$, C_2H_5Br, $C_2H_2Br_2$, C_6H_5Br, CH_3I) have been irradiated, and the products of neutron capture reactions (Cl^{38}, Br^{80}, Br^{82}, I^{128}) removed by various techniques. Extraction with water, either with or without added halogen or halide carrier, results in rather efficient separations. Yields are often improved, especially in the case of iodine, by extraction with an aqueous solution of a reducing agent such as HSO_3^-. Nearly complete extraction of activity has been reported in some cases when carrier was present. In the absence of carrier, yields of 50 per cent and large concentration factors have been found. Other methods which have been used for the removal of the active halogen from irradiated organic halides include adsorption on activated charcoal (with 30 or 40 per cent yields of halogen without added carrier) and collection on charged plates (with up to 70 per cent yields of halogen without added carrier).

Szilard–Chalmers separations of halogens with 70 to 100 per cent yields have also been obtained in neutron irradiations of solid or dissolved chlorates, bromates, iodates, perchlorates, and periodates; from these the active halogen can be removed as silver halide after addition of halide ion carrier. Szilard–Chalmers separations based on differences in oxidation state before and after the neutron capture have been successful for a number of other elements. About half the P^{32} activity formed in neutron irradiation of phosphates (solid or in solution) is found in +3 phosphorus. Most of the Mn^{56} activity can be removed from neutron-irradiated neutral or acid permanganate solutions in the form of MnO_2. Tellurium and selenium activities can be concentrated through the separation of tellurite or selenite carrier from irradiated tellurate or selenate solution by reduction of the lower

oxidation state to the element with SO_2 (reduction of the +6 state proceeds much more slowly than of the +4 state). Similarly a Szilard–Chalmers separation for arsenic has been reported by the addition of arsenite carrier to irradiated arsenate solution and precipitation of As_2S_3. Reduction to the metal has been found to occur in the neutron capture of gold in various compounds. Whether or not an active element can be successfully isolated in a different oxidation state from the bombarded compound depends not only on the relative stabilities of the two oxidation states but also on the speed of exchange between them under the conditions of the experiment. (Exchange reactions are discussed in chapter XIII, section B.)

Collection of charged fragments on electrodes has been used successfully for a number of Szilard–Chalmers separations. Arsenic activity has been separated by this method from arsine gas with yields up to 34 per cent. Deposition occurs on both positive and negative electrodes in this case.

The bombardment of metal-organic compounds and complex salts is often useful for Szilard–Chalmers separations if the free metal ion does not exchange with the compound, and if the two are separable. Some of the compounds which have been used successfully are: cacodylic acid, $(CH_3)_2AsOOH$, from which As^{76} can be separated as silver arsenite in 95 per cent yield; copper salicylaldehyde *o*-phenylene diamine, from which as much as 97 per cent of the Cu^{64} activity can be removed as Cu^{++} ion; uranyl benzoylacetonate, $UO_2(C_6H_5COCHCOCH_3)_2$, from which U^{239} activity has been extracted in about 10 per cent yield. It has been suggested that metal ion complexes which exist in optically active forms and do not racemize rapidly may be generally suitable for Szilard–Chalmers processes because the metal ion in such a complex is not expected to exchange rapidly with free metal ion in solution. Some complexes of this type have been used successfully, for example the triethylenediamine nitrates of iridium, platinum, rhodium, and cobalt, $Ir(NH_2CH_2CH_2NH_2)_3(NO_3)_3$ etc.

Hot-atom Chemistry. The highly excited recoil atoms resulting from neutron-capture reactions have been shown to undergo various types of chemical reactions. One of these is recombination with the fragment from which the "hot" atom had broken away. Insofar as it occurs, such recombination increases the

retention of the activity, retention being defined as the fraction of the active atoms not separable from the target compound. By means of retention studies recombination reactions have been shown to be more probable in the liquid than in the gas phase, and often more probable in the solid than in the liquid phase; for example, in liquid ethyl bromide the retention of bromine activity was found to be 75 per cent, and in ethyl bromide vapor (390 mm partial pressure with 370 mm of air) only 4.5 per cent. Retention has been shown to decrease markedly when the target substance is diluted. For example, the retention of bromine activity is about 60 per cent in solid carbon tetrabromide, about 28 per cent in an alcohol solution containing 1.15 mol per cent CBr_4, and 0 ± 2 per cent in an alcohol solution containing 0.064 mol per cent CBr_4. These results have been interpreted as indicating that retention cannot be caused entirely by recombination of fragments (in a so-called reaction cage) but is at least in part brought about through replacement by recoil atoms of isotopic atoms in other molecules of the target substance.

It has been shown that in reacting with a molecule a "hot" atom may replace another atom or group. For example, after the slow-neutron irradiation of CH_3I, 11 per cent of the I^{128} activity was found in the form of CH_2I_2; furthermore, this result was shown to be temperature-independent between -195°C and 15°C, which proves that the substitution is not an ordinary thermal reaction. The formation of labeled CH_2Br_2 in the irradiation of $CH_2BrCOOH$, of labeled CH_3I and C_2H_5I in the irradiation of iodine dissolved in ethyl alcohol, and of labeled C_6H_5Br in the irradiation of aniline hydrobromide show that the excited halogen atoms can replace such groups as —COOH, —OH, $—CH_2OH$, $—NH_2$, and probably many others. The yield of active atoms in one of these substitution products is usually less than about 10 per cent. Reactions of this type might conceivably be used to synthesize labeled compounds of high specific activity.

Some hot-atom reactions have been studied in inorganic systems. The retention of activity in permanganate, phosphate, and arsenate in thermal-neutron irradiations has been determined under varying conditions of *p*H and concentration. The results have been interpreted by W. F. Libby on the basis of competition between hydration reactions and oxidation–reduction reactions

for the hot atoms. In the case of permanganate, for example, Libby found the retention to be practically independent of permanganate concentration in neutral and alkaline solution and concluded from this that the manganese in the primary recoil fragment is in the +7 oxidation state (MnO_3^+, MnO_2^{+3}, MnO^{+5}, or Mn^{+7}). These species are then considered to undergo either hydration reactions, such as $MnO_3^+ + 2OH^- = MnO_4^- + H_2O$, or reduction by water, such as $4MnO_3^+ + 2H_2O = 4\,MnO_2 + 3O_2 + 4H^+$. At pH $\geq$ 12 the retention is nearly 100 per cent, presumably because the hydration reactions strongly predominate. Below pH 12 the retention falls rapidly with decreasing pH and reaches a constant value of about 7 per cent for pH values between 8 and 2. According to Libby's interpretation the reduction by water is faster than hydration in neutral and acid solutions. At still higher acid concentrations the retention again rises slightly, perhaps because exchange reactions between the active MnO_3^+ and inactive MnO_4^- can then compete with reduction by water; in support of this Libby showed that in acid solutions the retention increases with permanganate concentration. It is interesting to note that in arsenate bombardments the retention is nearly 100 per cent over a wide pH range; here the hydration reactions apparently far outweigh the oxidation–reduction reactions with water. The phosphate experiments showed retentions of about 50 per cent under all conditions tried, which might possibly be taken to indicate that only about 50 per cent of the primary recoil fragments contain phosphorus in the +5 state.

Isomer Separations. "Hot" atoms may result not only from nuclear reactions but also from radioactive-decay processes. The chemistry of hot atoms formed as a result of β-decay processes has been studied in a number of cases; for example, reactions such as

$$TeO_3^= \rightarrow IO_3^- + \beta^- \quad \text{and} \quad MnO_4^- \rightarrow CrO_4^= + \beta^+$$

can occur in addition to molecular disruption leading to other forms. Of course, for these studies the nucleus resulting from the β decay must itself be radioactive if its fate is to be investigated.

Up to the present time most of the published studies of the chemistry of recoil atoms following radioactive decay have been

confined to one type of decay process, isomeric transition. It is perhaps not immediately clear why isomeric transitions may lead to bond rupture. The γ-ray energies in isomeric transitions are much lower than in neutron-capture processes, often below 100 kev and rarely above 500 kev. According to equation XI–1, a 100-kev γ ray would give a nucleus of mass 100 a recoil energy of only about 0.05 ev, which is not nearly enough to break a chemical bond. Although internal-conversion electron emission gives rise to roughly 10 times greater recoil energy than γ emission at the same energy,[8] even this is not sufficient for bond rupture in most cases. However, the vacancy left in an inner electron shell by the internal conversion leads to electronic rearrangements and emission of Auger electrons; the atom is, therefore, in a highly excited state (and positively charged), and molecular dissociation may take place if the atom is bound in a molecule.

Separations of nuclear isomers analogous to Szilard–Chalmers separations have been performed in a number of cases where the isomeric transition proceeds largely by conversion-electron emission. The 18-min Br^{80} has been separated from its parent, the 4.4-hr Br^{80}, by a number of different methods analogous to the Szilard–Chalmers methods used for bromine. The lower states of Te^{121} (17 days), Te^{127} (9.3 hr), Te^{129} (72 min), and Te^{131} (25 min) have been separated as tellurite in good yield from tellurate solutions containing the corresponding upper isomeric states. Isomer separations are sometimes useful for the assignment of isomer activities and for the elucidation of genetic relationships. That the possibility of obtaining isomer separations depends on internal conversion was shown in experiments using the gaseous compounds $Te(C_2H_5)_2$ containing 90-day Te^{127} and 32-day Te^{129}, and $Zn(C_2H_5)_2$ containing the 13.8-hr upper isomeric state of Zn^{69}. The lower isomeric states of the tellurium isomers could be separated on the walls of the vessels or on charged plates, but under identical conditions no separation of the zinc isomers was obtained; the isomeric transition in Zn^{69} proceeds by an unconverted 440-kev γ ray, whereas the tellurium transitions have energies of only about 100 kev but are almost completely converted.

[8] In nonrelativistic approximation an atom of mass M receives a recoil energy $E_r = E_e(m/M)$ from a conversion electron of energy E_e.

EXERCISES

1. Suggest hopeful easily prepared compounds for use in Szilard–Chalmers processes of (*a*) iron, (*b*) mercury, (*c*) technetium.

2. A certain activity chemically proved to be associated with technetium (element 43) is produced in the bombardment of molybdenum with 12-Mev deuterons and in the bombardment of ruthenium with 12-Mev deuterons, but not in the bombardment of ruthenium with fast (up to 15 Mev) neutrons. To what isotope of technetium should the activity be assigned? What mode of decay would you expect?

3. A sample of sodium iodide is irradiated with fast neutrons to produce 90-day Te^{127}. Suggest a chemical procedure for the isolation of the tellurium. How would you modify this procedure if you knew that the sodium iodide contained some sodium bromide impurity?

4. What is the recoil energy imparted to a Te^{129} atom by the emission of a 70-kev conversion electron? (Use the relativistic expression for the electron energy.) *Answer*: 0.318 ev.

5. Element Z has a single stable nuclide of mass number A. In the bombardment of element Z with 28-Mev deuterons the following activities chemically identified with element $Z + 1$ were found:

a moderately strong 3-hr positron emitter,
a strong 2.6-day activity emitting mostly γ and X rays,
a weak 30-min positron emitter.

The last activity was not produced when the deuteron energy was lowered to 20 Mev.

Critical-absorption measurements showed the X rays of the 2.6-day activity to be those of element Z. An 11-day isotope of element Z was shown to grow from the 2.6-day activity, whereas the 3-hr activity decayed to a 50-min X-ray emitter chemically identified with element Z. The X rays from this latter activity were characteristic of element Z.

One-million-electron-volt neutrons produced in a target of element Z the 50-min X-ray emitter, in addition to a 14-hr β^--emitting isotope of Z which had also been identified in the deuteron bombarded Z samples. With 15-Mev neutrons the 11-day isotope of Z was also produced.

Make mass assignments for the various radioactive isotopes of elements Z and $Z + 1$, and indicate the most likely mode of decay for each. The stable nuclides of $Z + 1$ have mass numbers $A + 1$, $A + 2$, and $A + 3$; those of $Z - 1$ have mass numbers $A - 3$, $A - 2$, and $A - 1$.

6. Suggest methods for the chemical identification of

(*a*) V^{52} produced in the fast-neutron bombardment of a chromate solution,

(*b*) Mn^{52} produced in the deuteron bombardment of iron,

(*c*) O^{14} produced in the proton bombardment of nitrogen gas.

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XII. CHEMISTRY OF LOW CONCENTRATIONS AND THE STUDY OF NEW ELEMENTS

A. THE LOW-CONCENTRATION REGION

Limits of Detection. The working region of concentrations in ordinary chemical studies is limited by the sensitivity of available analytical methods. The lower detection limits for different substances vary widely. Gravimetric procedures rarely are useful for concentrations as low as one part per million (1 ppm); spectroscopic elementary analysis in favorable cases offers about the best sensitivity, and detection of a number of elements at 0.01 ppm and less may be practical. It is true that some compounds of very pronounced odor may be noticed at much lower concentrations; for example, at 0.01 ppm in air the odor of mercaptan is very strong, and it may be recognized at 0.00001 ppm. The number of mercaptan molecules sufficient for recognition in this way is estimated to be about 3×10^{10}, corresponding to 2×10^{-12} g in about 100 ml of air. Other detection methods, especially biological assays, may approach and exceed even this sensitivity. But for the most part these methods have not offered practical means for extending knowledge of chemical behavior to such extremely low concentrations. The property of radioactivity does offer a rather convenient analytical method for concentrations so low and even much lower. As few as several thousand radioactive molecules may be detected, even when contained in sizable samples. The practical working limits are fixed by the half-life and by the nature of the radiation. A polonium solution at 10^{-12} mole per liter has an easily detectable activity of 35 μrd per ml (35 microrutherfords, or 35 disintegrations per sec, per milliliter). The shorter-lived La^{140} (40-hr half-life) may be studied in 10^{-14} molar solution. A new concentration region is opened for study by the technique of radioactive tracers.

Radiocolloids. It was observed many years ago that radioactive elements in some solutions where they existed at extremely low concentrations showed unusual physical properties in that they

behaved more like colloids than true solutes. That the active atoms or molecules were clustered together in these solutions was shown in suitable cases by photographic registration of the spotty distribution of disintegration α rays. The size of the colloidal particles has been estimated from observed sedimentation rates on centrifugation, and it has been established that in some cases the phenomenon is one of adsorption of many of the active solute molecules or ions on particles of dust, silica, or the like, inevitably present even in "pure" water. The general adsorption phenomena are discussed in the next section. It should be remembered that effects of this sort, including adsorptions on container surfaces, filter paper, and so on, may be quite important in work at these very low "tracer" concentrations. The same effects, no doubt, occur in work at ordinary concentrations, but then the amount of material involved represents such a small fraction of the total amount that these effects are not noticed.

B. COPRECIPITATION AND ADSORPTION

Fajans' Precipitation Rule. Many of the manipulations of ordinary chemistry and also of radiochemistry require precipitation reactions. How are substances at tracer concentrations to be separated by precipitation, when often the concentration is too small to exceed the solubility-product condition, and when the amount of precipitate even if formed would be quite imperceptible? Of course, if the radioactivity is isotopic with an element available in quantity, more of the element may be added in a suitable chemical form as a carrier, and then the chemical problems become ordinary ones. If necessary or if desirable, nonisotopic carriers may be tried. Microscopic amounts of radium in solution are brought down with barium in the precipitation of $BaSO_4$. Strontium ions have been carried by calcium salts, iodide ions by chloride precipitates, and so on. In some cases precipitates carry down active substances where the chemical similarities are not so obvious; for example, tracer lead (ThB) is well carried by ammonium dichromate crystals, and many active bodies are carried by $Fe(OH)_3$ precipitates. On the basis of a number of such observations K. Fajans in 1913 formulated this principle: the lower the solubility of the compound formed by the radioelement (as cation) with the anion of the precipitate, the greater

the amount of radioelement carried with the precipitate. As an illustration, bismuth tracer is carried by $BaCO_3$ and $Fe(OH)_3$ but not by $BaSO_4$ or $PbSO_4$ from acid solutions. Lead tracer may be carried by all these, but is carried less well by AgCl, and is not carried by a nitron nitrate precipitate. Exceptions to this rule are not uncommon; ThB (a lead isotope) is not precipitated with HgI_2 or with cupric fumarate although both PbI_2 and lead fumarate are rather insoluble. The occurrence of carrying not predicted by this rule is widely observed even on the macroscale in analytical chemistry; for example KNO_3 is appreciably coprecipitated with $BaSO_4$. Clearly factors in addition to that expressed in the Fajans rule are important in these phenomena.

It should be noticed that precipitates previously formed in the absence of tracer may take up a tracer when added in suspension to the tracer solution. This is the method of preformed precipitates. This procedure and an intermediate case between it and ordinary coprecipitation find convenient uses in radiochemical separations. For example, consider the carrying of radioactive yttrium by lanthanum fluoride; after a single precipitation of LaF_3 with excess HF a small fraction of the yttrium activity may remain in solution. Now, when more lanthanum is added to excess, a new precipitate of LaF_3 forms immediately, probably before the La^{+++} and Y^{+++} ions are well mixed. This precipitate may carry yttrium with it, but probably not so well as the first precipitate, or the third precipitate which may now be formed by addition of excess HF.

Hahn's Classification. O. Hahn in 1926 proposed a classification of carrying phenomena, distinguishing cases of true coprecipitation from cases of surface adsorption. The four principal types of carrying which he described are (1) isomorphous replacement, (2) surface adsorption, (3) anomalous mixed crystals, and (4) internal adsorption. In discussing these we will give most emphasis to the first two; the others are not so well understood.

1. *Isomorphous Replacement.* If the carrying ion and the ion carried form with the precipitating ion isomorphous crystalline compounds, coprecipitation of this type is to be expected. The radioelement is distributed throughout the precipitate crystals as may be shown by a radioautograph technique, and the mechanism is simply one of replacement at normal ion sites in the crystal lattice. This true coprecipitation is not much affected

by conditions during precipitation such as acidity, order of addition of reagents, rate of crystallization, and temperature; and repeated washing of the precipitate cannot remove the coprecipitated substance. The precipitation of radium with barium salts is an example of this class of carrying.

2. *Surface Adsorption.* Freshly formed precipitate crystals with large surface areas may be capable of adsorbing radioelements effectively. For this type of carrying the Fajans rule is significant, but also another important factor is the surface charge on the precipitate relative to the ionic charge of the tracer substance. Because important adsorption occurs only when these charges are of opposite sign, experimental factors affecting the surface charge of the precipitate strongly influence the carrying, and this type is recognized by sensitivity to such factors as acidity, order of addition of reagents, and physical state of subdivision of the precipitate. In many instances an appreciable part of the adsorbed activity may be washed off, or displaced by another ion of similar charge. The carrying of ThB (a lead isotope) at tracer concentrations by $CaSO_4$ or AgBr, and of Ra by Ag_2CrO_4 are examples. Table XII–1 shows the importance of excess of the anion, neces-

TABLE XII–1

CARRYING OF ThB BY $CaSO_4$

Excess Ca^{++}	Excess $SO_4^{=}$	ThB carried
600%		1.7%
10%		5.2%
	5%	88 %
	900%	98 %

sary to produce a negative surface charge on the precipitate crystals, for the carrying of cations.

3. *Anomalous Mixed Crystals.* A type of carrying which at least superficially closely resembles isomorphous replacement is observed in a number of cases where true isomorphism is unexpected and even unlikely. An example is the carrying, in a manner hardly affected by precipitate surface charges, of RaB or RaD (lead isotopes) by $BaCl_2 \cdot 2H_2O$; these crystals are monoclinic, but $PbCl_2$ in macroscopic amounts crystallizes in the rhombic system. The capacity of the barium chloride for lead ions has

been found to be limited to about 0.1 mol per cent lead in the crystals.

4. *Internal Adsorption.* There are some cases of carrying that do not fit into any of the three types already discussed and are characterized by a spotty distribution of tracer within the precipitate crystals as shown by radioautographs. These cases may not be very numerous and seem to be associated with very poor carrying. For example, although lead tracer is carried in an anomalous mixed crystal by barium chloride, it is carried only very slightly by barium bromide; the small fraction of ThB that is carried is distributed in spots and patches scattered through the barium bromide crystals.

In addition to these four types Hahn considers the mechanical inclusion in precipitate crystal masses of radiocolloids that might exist in the solution and inclusion of portions of the mother liquor itself.

Doerner–Hoskins and Berthelot–Nernst Distributions. For true coprecipitation of the isomorphous-replacement type, and apparently also of the anomalous-mixed-crystal type, the progress of crystal separation and the detailed distribution of the active tracer within the precipitate crystals may tend to approach either of two limiting laws. For the assumption that the precipitate crystals grow progressively, with equilibrium conditions maintained between the solution and the crystallizing layer, and with both re-solution and solid-diffusion effects negligible, a quantitative treatment is easily made. Let x and y be the amounts of the two substances X and Y precipitated before a given instant, and let a and b be the total amounts of X and Y; then:

$$\frac{dx}{dy} = \lambda \frac{a - x}{b - y},$$

where λ is a constant characteristic of the system. In words, the ratio of X to Y in the forming surface layer is proportional to the ratio of the respective concentrations still remaining in the solution. On integration,

$$\log \frac{a}{a - x} = \lambda \log \frac{b}{b - y};$$

this is the logarithmic distribution law derived by H. A. Doerner and W. M. Hoskins. There is evidence that it is closely

approached in many actual coprecipitations; it is found especially for precipitations produced by gradual evaporation with care to avoid any supersaturation, and for precipitations from supersaturated solutions that are vigorously stirred and quickly separated (filtered).

If for any reason the entire crystal rather than just the surface layer is brought into equilibrium with the solution, then the differential equation just given should be replaced by a similar nondifferential expression:

$$\frac{x}{y} = D\frac{a-x}{b-y}.$$

Here the ratio of X to Y in the crystals is proportional to the ratio of X to Y left in solution; this is the Berthelot–Nernst homogeneous distribution law applicable to partition of a solute between liquid phases. Coprecipitations made from strongly supersaturated solutions and coprecipitations in which a finely divided precipitate is left standing in contact with the solution are likely to approach this distribution law. In the first case failure to maintain equilibrium between the growing crystals, the immediately surrounding solution, and the bulk of the solution seems to be involved; in the other re-solution and recrystallization probably play an important role.

C. OTHER CHEMICAL PROPERTIES

Partition between Solvents. The partition of active solutes at tracer concentrations between two immiscible liquid phases one would suppose might be simpler than the distributions in precipitations. Actually not much information on this subject has been reported, but such results as have been obtained do not give evidence of any abnormal effects at very low concentrations. The distribution of $GaCl_3$ between aqueous HCl and ether phases has been studied at about 10^{-12} molar, and the distribution coefficient has been found to be the same as at a higher concentration (0.0016 molar). However, this cannot be expected to be true for all substances; for example, if the molecule extracted should be a dimeric form, then at sufficiently low concentration the distribution would surely change.

Volatility. The volatility of unweighable amounts of tracer substances has been often investigated, although not for a wide variety of elements. This volatility is probably related to the macroscopic volatility of the substances but is, no doubt, modified by the nature of the surface attachment to the necessary supporting foil. Bismuth isotopes of the active deposit from emanation are volatilized from platinum in air at 800° to 900°C, and the active lead isotopes are volatilized at 700° to 800°C. These temperatures may be reduced somewhat by previous exposure of the substance to a free halogen, presumably because bismuth and lead halides are formed. Radioactive element 85 (astatine) formed in metallic bismuth by α-particle bombardment volatilizes almost completely in vacuum from liquid bismuth at about 300°C. Cadmium formed by transmutation from silver is separated in the same way at 900°C. Information is not available to permit quantitative comparisons of macrovolatilities with tracer volatilities from the interiors of solid substances. Volatility properties of unknown substances available only as tracers have been sought in experiments in which a solid containing the tracer (possibly by coprecipitation) is volatilized and the tracer either volatilized or left behind; these experiments too can be difficult to interpret because the tracer may be swept away with the carrier somewhat below its normal volatilization temperature.

Electrochemistry. The electrochemistry of ions at tracer concentrations has been the subject of many investigations, and a number of convenient radiochemical separation procedures are based on electrochemical methods. For example, RaF (polonium) is separated from RaD (lead) and RaE (bismuth) by its spontaneous deposition from dilute HCl solution on a silver foil; RaD and RaE are more electropositive and are not displaced from the solution by silver. Replacement by nickel with deposition on a nickel foil can be used to remove RaE and leave RaD in solution. The deposition potentials of some tracers have been measured for the extremely low concentrations; the cathode potential (measured with respect to a suitable auxiliary reference electrode) necessary for the deposition of the tracer is interpreted as analogous to the decomposition potential. All these experiments give means of locating the trace substance in the electromotive series and permit approximate evaluation of the standard electrode potential, provided proper account is taken of the large shifts in emf caused by the extremely low ion concentrations.

Tracer ions in a solution between electrodes move in a direction determined by their charge, and the rough average sign of charge is revealed by observation of the net average transport of the tracer. In simple cases some information on the magnitude of the charge can be obtained by a careful quantitative study.

D. DISCOVERIES OF NEW ELEMENTS BY TRACER METHODS

The Role of New Physical Methods. More than half a century ago the methods of chemistry conventional at that time had already reached a limit in the search for new and missing elements; discoveries since that time have depended on the introduction of new physical methods. Through studies of optical spectra the elements rubidium, cesium, indium, helium, and gallium were found. The first evidence for hafnium and rhenium came from X-ray spectra. Early investigations of the natural radioelements revealed the existence (often in extremely small amounts) of polonium (number 84), radon (86), radium (88), actinium (89), and protactinium (91), and recently the missing element number 87 has been found in the same way. Through studies of nuclear reactions and artificially induced radioactivities the elements 43, 61, and 85 have been identified, and more recently elements 93, 94, 95, and 96 have been added to the periodic chart. In this section we will discuss briefly the discoveries of the last eight elements mentioned.

Technetium. In 1925 W. Noddack, I. Tacke, and O. Berg, who had previously found the new element rhenium through its X-ray spectrum, reported observation of a faint X-ray line in a concentrated rhenium sample that would correspond to the lighter homolog of rhenium, the missing element 43; they proposed for it the name masurium, symbol Ma. The element could not be concentrated, and the work has not been verified; present knowledge of the isotopes in this region of atomic weights ($\sim$100) makes it appear unlikely that this element exists in nature in stable form. (See chapter VI, page 140.) C. Perrier and E. Segrè working in Italy isolated and studied radioactive isotopes of element 43 from an old molybdenum deflector plate of the Berkeley 37-inch cyclotron. These were long-lived K-capture activities, produced through d, n reactions by the deuteron beam. Perrier and Segrè have recently offered for this element the name technetium, symbol Tc, derived from the Greek word meaning "artificial."

In their early studies, Perrier and Segrè compared the chemical behavior of the tracer activity with that of several carrier substances which might be guessed to be chemically similar, particularly manganese and rhenium because technetium falls between these in subgroup VII of the periodic table. They found the stability of the +7 oxidation state to be greater than for manganese and less than for rhenium, as expected. Technetium was carried by a precipitate of Re_2S_7 on addition of H_2S to HCl solutions up to 6 normal; the technetium seemed to concentrate somewhat in the solution on partial precipitation, which suggests a greater solubility for Tc_2S_7, although this evidence taken alone is subject to other interpretations. Precipitates of MnO_2 and $Mn(OH)_2$ did not carry technetium unless strong reducing agents were present. $KReO_4$ and $CsReO_4$ did carry the tracer, and the insolubility of the technetium compound seemed to be less than that of the perrhenate for the potassium salts and greater for the cesium salts. Nitron perrhenate was found to carry technetium quantitatively. The technetium tracer volatilized completely in air at about 300°C, probably as an oxide; $TcCl_7$ from Tc_2S_7 plus Cl_2 volatilized at 100°C. Technetium may be separated from molybdenum by precipitation of the latter with 8-hydroxyquinoline. It may be separated from ruthenium by volatility of $TcCl_7$ or by distillation of RuO_4 from perchloric acid solution. It may be freed of rhenium by volatilization of the rhenium at about 180°C in dry hydrogen chloride, by fractional crystallization of $KReO_4$, or by precipitation of Re_2S_7 from 10 *N* HCl. It may be volatilized from columbium in oxygen at a high temperature.

At this time twenty-one activities have been reported for technetium isotopes, and most of them are well established. Perhaps most interesting is the lower isomeric state of Tc^{99}; it has a half-life of about 10^6 years and may be produced in quantity in a chain reactor. Its chemical properties have now been investigated at macroconcentrations and found to agree insofar as reported to date with the indications given by the tracer experiments. The sulfide Tc_2S_7 precipitated from 4 *M* H_2SO_4 is dark brown and highly insoluble. The pertechnetate ion TcO_4^- is probably pink. Technetium metal has been prepared and found by X-ray diffraction to be in the hexagonal close-packed arrangement, isomorphous with rhenium, with density 11.49 g per cc.

Astatine. Isotopes of element 85 have been claimed and fairly well identified by their radiations as very short-lived branch products in the radium and thorium series, formed from RaA and ThA by β decay in a very small fraction of the disintegrations (the normal mode is α decay for both). F. Allison claimed in 1931 that this element had been observed as a natural substance by the magneto-optic technique. However, the first quite definite demonstration of element 85 was given by D. R. Corson, K. R. Mackenzie, and Segrè; they produced a radioactive isotope 85^{211} by the $\alpha, 2n$ reaction on Bi^{209} using 30-Mev helium ions from the then newly completed 60-inch cyclotron in Berkeley. The half-life is 7.5 hr, and the decay is 40 per cent by α emission and 60 per cent by K capture. Several other isotopes are now known, but the half-lives are all short. Corson, Mackenzie, and Segrè have recently proposed the name astatine, symbol At, derived from the Greek word meaning "unstable."

Astatine is the halogen just heavier than iodine, and its chemical properties make a very interesting study. Because of the short half-life macroscopic quantities may not be accumulated, but tracer studies by G. L. Johnson, R. F. Leininger, and Segrè have given information which we may summarize. The element is separated from bismuth, from which it is formed, and from simultaneously produced radioactive polonium by its volatility from molten bismuth in vacuum. The free element is quite volatile, particularly from a glass surface, even at room temperature. It has a rather specific affinity for metallic silver surfaces even at 325°C. The free element exists in aqueous solution (presumably as At_2 or possibly At) and usually is partly lost on evaporation of acidic solutions. The free element is readily extracted from water solutions into benzene or carbon tetrachloride very much like iodine. It may not be extracted from alkaline solution, again like iodine.

Astatine may be reduced by SO_2 or by zinc, but not by ferrous ion, almost certainly to the -1 oxidation state. This ion is precipitated with AgI or TlI from acidic or basic solutions. Cold concentrated nitric acid seems to oxidize elemental astatine only slowly. It is oxidized by bromine, and to some extent by ferric ion, to some positive oxidation state (possibly AtO^-). This state is shown by migration experiments to be an anion; it is hardly AtO_3^- since it is not carried by a precipitate of $AgIO_3$. With

HClO or hot $S_2O_8^{=}$ as oxidizing agent a different anion results; this is carried by $AgIO_3$ and may be AtO_3^-.

Francium. In 1914 F. Paneth observed some α-particle emission from $_{89}Ac^{227}$, which decays mostly by β emission to RdAc. In 1939 M. Perey observed the daughter produced in the 1 per cent α branch and called the isotope of element 87 so produced AcK. This body decays by β emission (to AcX); the half-life is 21 min. Mlle. Perey has proposed the name francium, symbol Fr, for the new element. Its chemical properties appear to be as expected from its position in the periodic table in group I below cesium. It is carried along with $CsClO_4$ or Cs_2PtCl_6 and also by the analogous rubidium salts. Although the corresponding sodium and potassium salts apparently crystallize in the same crystal systems (rhombic for the perchlorates and cubic for the chloroplatinates), these do not carry francium effectively, presumably because of the great differences in ionic radii. Another isotope of francium may be formed in a rare α-particle branching of $MsTh_2$ (Ac^{228}), but this product has not been observed. Recently Fr^{221} has been described as an α emitter with a 4.8-min half-life in the $4n + 1$ series, and other very short-lived α-emitting francium isotopes have been produced artificially.

Element 61. The history of element 61 has been and remains confused. In 1926 several groups of researchers reported evidence based on optical and X-ray spectral lines for the existence of the element in various minerals and rare-earth concentrates; the names of these workers included J. A. Harris, B. S. Hopkins, and L. F. Yntema; L. Rolla and L. Fernandes; and J. M. Cork, C. James, and H. C. Fogg. Names for the element proposed in this period were illinium, Il, by Hopkins, and florentium by Rolla. If the element actually exists in nature in stable form and is detectable by methods then used, it is rather surprising that higher concentrations have not been prepared. About 1941 and shortly thereafter workers at Ohio State University including H. B. Law, M. L. Pool, J. D. Kurbatov, and L. L. Quill and later C. S. Wu and Segrè in Berkeley obtained from cyclotron bombardments several activities which were attributed to isotopes of the missing element; however, the certainty of this interpretation was not positively established. Pool and Quill have recently proposed for the element the name cyclonium, symbol Cy. The fission of uranium produces several radioactive isotopes of ele-

ment 61, and these have been investigated and definitely characterized by workers at Oak Ridge including C. D. Coryell, J. A. Marinsky, and L. E. Glendenin. They were able to concentrate the tracer activities by the ion-exchange resin adsorption and elution technique. Their proposal for the name of the element is promethium, symbol Pm. Recently, visible amounts of 61^{147} have been exhibited.

Transuranium Elements. When Fermi and his group in Rome first exposed uranium to slow neutrons they observed a number of activities, and in the following few years many more active species were found to be produced; most of these were at that time assigned to transuranium elements. The assignments were made because the substances were transformed by successive β emissions which led to higher Z values, and because they could be shown by chemical tests to be different from all the known elements in the neighborhood of uranium in the periodic chart. This situation was resolved in the discovery by Hahn and F. Strassman that these activities could be identified with known elements much lighter than uranium and that, therefore, the neutrons produce fission of the uranium nuclei. Further investigation of the fission process and products led to the proof by E. M. McMillan and P. Abelson that one of the activities, the one with 2.3 days half-life, could not be a product of fission and was actually the daughter of the 23-min β-particle-emitting U^{239} which resulted from U^{238} (n, γ) U^{239}. Also, they devised a procedure for separating chemically the element 93 tracer from all known elements through an oxidation–reduction cycle, with bromate as the oxidizing agent in acid solution, and with a rare-earth fluoride precipitate as carrier for the reduced state. They gave the name neptunium, symbol Np, to the new element, taking the name from Neptune, the planet next beyond Uranus in the solar system.

At the present time seven isotopes of neptunium are known, with half-lives from 53 min for K- and α-active Np^{231} to 2.25×10^6 years for α-active Np^{237}. Both Np^{239} discovered by McMillan and Abelson and Np^{238} from α, $p3n$ or d, $2n$ reactions on U^{238} emit β particles and lead to known isotopes of element 94, very naturally named plutonium after Pluto (a planet beyond Neptune), with symbol Pu. These isotopes, Pu^{238} and Pu^{239}, are moderately long-lived α emitters first studied by McMillan, G. T. Seaborg, Segrè, A. C. Wahl, and Kennedy; Pu^{239} is distinguished for its

practical usefulness in slow- and fast-neutron fission. Another isotope Pu^{241} has been reported; it decays with a half-life of about 10 years by β^- emission to produce Am^{241}, an isotope of the element 95, named americium with symbol Am. This americium isotope has a half-life of 500 years and emits α particles. By an n, γ reaction Am^{241} is converted to Am^{242}, a β-emitting 17-hr isotope; this decays to the isotope of curium, Cm^{242}. This last substance emits α particles and has a half-life of 5 months; it is formed also by the reaction Pu^{239} (α, n) Cm^{242}. In addition Cm^{240}, an α emitter with 1 month half-life, has been prepared by Pu^{239} $(\alpha, 3n)$. Other isotopes of these elements are listed in table A in the appendix.

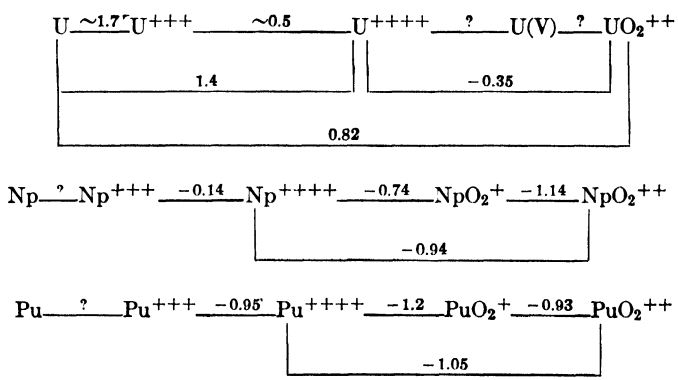
The chemical properties of all these transuranium elements—neptunium, plutonium, americium, and curium—have been studied first as tracers and later by ultramicrochemical techniques. At the present time Pu^{239} exists in some quantity, Np^{237} has been isolated to the extent of hundreds of milligrams, Am^{241} has been isolated on a microscale, and a very small amount of Cm^{242} has been isolated. Most of the work has been done in Seaborg's laboratory, at the University of Chicago Metallurgical Project and at the University of California Radiation Laboratory; the elements americium and curium were discovered by Seaborg, R. A. James, L. O. Morgan, and A. Ghiorso. The transuranium elements and uranium and thorium all have similar precipitation properties when in the same oxidation state; they differ principally in the ease of formation and in the existence of the various oxidation states. Seaborg has advanced the hypothesis, and with considerable evidence, that a new rare-earth series begins with actinium (number 89), with the $5f$ electron orbitals being filled in subsequent elements. This would be analogous to the lanthanide rare-earth series beginning with lanthanum (number 57), with the $4f$ orbitals filling in the next fourteen elements. Some of the evidence for this actinide series may be seen in these facts: (1) lanthanum is chemically similar to actinium; (2) thorium is similar to cerium in the +4 state; (3) the ease of removal of more than three electrons decreases from uranium to curium. (Approximate oxidation potentials for uranium, neptunium, and plutonium are given in table XII-2.) There is additional evidence for the second rare-earth series from spectroscopic and crystal-structure data.

The interpretation of such magnetic data as are available is not at all clear.

It does seem evident that this new series differs from the familiar rare-earth series in that the resemblance of successive elements is less than for the lanthanide series. The lanthanide earths are for the most part separable only by multiple fractionation processes, or better by adsorption and elution from ion-exchange resins; the elements from 89 to 95 are separable by oxidation–

TABLE XII–2

OXIDATION POTENTIALS OF URANIUM, NEPTUNIUM, AND PLUTONIUM IONS



reduction processes, but the separation of 95 from 96, both of which are stable in the +3 state, may require an ion-exchange column or a fractionation method. On the actinide hypothesis, curium, by analogy to gadolinium, would be expected to resist oxidation or reduction in the +3 state, because the $5f^7$ and $4f^7$ structures, with one electron in each of the seven f orbitals, are particularly stable. Then americium, by analogy to europium, might possibly be reducible to a +2 state. (The oxide AmO has been reported, also AmO_2.) Actually it is on the basis of these analogies that the names for 95 and 96 were suggested; curium after Pierre and Marie Curie and americium after North and South America. On the basis of an actinide series we might speculate on the properties of element 97, which likely will be found in time; by analogy to terbium it might be capable of oxidation to a +4 state.

Some of the difficulties in work with substances like Cm^{242} may be mentioned here, difficulties in addition to those naturally associated with work on the ultramicrochemical scale. The heavy short-lived α emitters are extremely dangerous as radioactive poisons, and amounts of the order of a few micrograms taken into the body may produce harmful effects. Also, the high level of α radiation in concentrated samples can be expected to have some effect on chemical reactions; notice that a curium preparation glows in the dark. In fact the rate of energy release is so great that if cooling effects are neglected it may be estimated that a 0.1 molar Cm^{242} solution would begin to boil in about 15 sec and reach dryness in about 2 min. The question of possible existence of longer-lived isotopes of curium, of course, arises. The stability curve in this region would suggest that less α-active isotopes might occur at slightly higher masses, nearer Cm^{244} or Cm^{245}. No very practical way of preparing these isotopes comes easily to mind.

EXERCISES

1. Calculate the electrode potentials corresponding to these half-reactions at the specified concentrations:

(*a*) $Ag = Ag^+ + e^-$, with $(Ag^+) = 10^{-13}$ molar;

(*b*) $Al = Al^{+++} + 3e^-$, with $(Al^{+++}) = 10^{-15}$ molar;

(*c*) $2Hg = Hg_2^{++} + 2e^-$, with $(Hg_2^{++}) = 10^{-8}$ molar.

(*d*) Would the shift in emf in (*c*) continue to be proportional to the logarithm of the concentration of mercury ions in solution as that concentration was indefinitely reduced? *Answer:* (*a*) −0.031 v.

2. In analytical chemistry Fe^{+++} is used as an oxidizing agent to convert I^- to I_2. Would Fe^{+++} be suitable to oxidize a solution of (pure) I^{128}, concentration 150 μrd per ml?

3. On the basis of data given in table XII–2, estimate the concentrations of Pu^{+++}, PuO_2^+, and PuO_2^{++} at equilibrium in a solution which has $(Pu^{++++}) = 1$ molar and $(H^+) = 1$ molar.

Answer: (Pu^{+++}) = about 0.1 molar.

4. What is the electrode potential for $Cu = Cu^{++} + 2e^-$ at a Cu^{++} concentration equal to exactly zero? What do you suppose would be the result if a pure copper electrode were immersed in absolutely pure water?

5. In an experiment on the crystallization of mixed radium–barium chlorides from supersaturated solutions the following data were obtained:

Percentage of Radium Remaining in Solution	Percentage of Barium Remaining in Solution
87.41	97.48
60.30	89.21
59.01	88.45
54.72	86.58
47.61	83.53
43.15	80.24

Do these fractional crystallizations obey the Doerner–Hoskins or the Berthelot–Nernst equations? Find λ or D.

6. Take the partition coefficient for the distribution of astatine between carbon tetrachloride and water as 100 at 10^{-10} molar for the nonaqueous phase. Further, assume that the substance in carbon tetrachloride has the molecular formula At_2. What might you expect for this partition coefficient at 10^{-11} molar (nonaqueous phase) if the zero state in water at these concentrations is predominantly (*a*) At_2, (*b*) At, (*c*) $At^- + HAtO$, (*d*) $At_3^- + HAtO$, (*e*) $3\,At^- + AtO_2^-$. *Answer:* (*b*) 32.

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XIII. TRACERS IN CHEMICAL APPLICATIONS

A. THE TRACER METHOD; DIFFUSION STUDIES

Isotopic Tracers. Most of the ordinary chemical elements are composed of mixtures of isotopes, and each mixture remains essentially invariant in composition through the course of physical, chemical, and biological processes. That this is so is shown by the constant isotopic ratios found for elements from widely scattered sources [1] and by the fact that atomic weights reliable to many significant figures may be determined by chemical means. It is true that isotopic fractionation may be appreciable for the lightest elements where the percentage mass difference between isotopes is greatest, and this effect must always be considered in the use of hydrogen tracer isotopes. However, apart from H^3 (tritium) there is no practical radioactive tracer lighter than Be^7, which differs in mass from stable Be^9 by only about 25 per cent, and the next heavier tracer is in carbon where already the specific isotope effect may be neglected in most tracer work of ordinary precision.

The fact that a given nuclide may be radioactive does not in any way affect its chemical (or biological) properties; at least this is true for each atom until its nucleus actually undergoes the spontaneous radioactive change. Because the tracer-isotope atoms are detected by their radioactivity, this means that they behave normally up to the moment of detection; after that moment they are not detected, and their fate is of no consequence. Of course, if the resulting atoms after the nuclear transformation also should be radioactive and capable of a further nuclear change, the detection method must be arranged to give a response which measures the proper (in this case the first) radioactive species only. For example, if RaE (Bi^{210}) is used as a tracer for bismuth the α particles from its daughter Po^{210} should not be allowed to

[1] Some exceptions to the constancy of isotopic ratios were mentioned in chapter I, section C, and chapter II, section B.

enter the detection instrument but should be absorbed by a suitable absorber or by the counter wall. As a tracer for thorium UX_1 is suitable in spite of the fact that most of the detectable radiation will be from its daughter UX_2; the reason is that the short half-life, 1.14 min, of UX_2 insures that it will be in transient equilibrium with the UX_1 by the time the sample is mounted and ready for counting, so that the total activity will be proportional to the UX_1 content. Many multiple decays are found in the fission-product activities. If Ba^{140} ($t_{1/2}$ = 12.8 days) is to be used as a tracer for barium, the isolated samples either should be freed chemically of the daughter La^{140} ($t_{1/2}$ = 40 hr) or should be kept before counting for a week or two until the transient equilibrium is achieved. The isomeric transition activities present interesting cases; if the 4.4-hr Br^{80} is chosen as a tracer for bromine the 18-min lower isomeric state of Br^{80} will always be present, and, because of the hot-atom effects accompanying the isomeric transition as discussed in chapter XI, the two isomers may be present in different chemical forms. However one may use the 4.4-hr Br^{80} with confidence, provided only that one measures isolated samples for the 4.4-hr period by analysis of the decay curves or simply by holding the prepared samples for a time long compared to 18 min before counting. These special cases do not arise in the use of the great majority of popular tracer isotopes. Another source of interference with the tracer principle is a possible chemical (or biological) effect produced by the ionizing rays; this radiation chemistry effect is not often encountered at the usual tracer activity levels and may always be checked by experiments with a much higher or lower level of radioactivity.

A definite limitation of the radioactive tracer method is the absence of known active isotopes of suitable half-life for a few elements, especially oxygen and nitrogen. There are radioactive oxygen isotopes, O^{15} and O^{19}, but these have half-lives of 126 sec and 31 sec, respectively. The 7-sec N^{16} and 4-sec N^{17} are useless as tracers, but some applications of the 10-min N^{13} β^+ activity have been made. Also, helium, lithium, and boron do not have periods longer than 1 sec. The use of separated stable isotopes as tracers is a very valuable technique, and availability of the necessary concentrated isotopes is increasing. Enriched O^{18} and N^{15} are essential for many interesting and important purposes, and C^{13} offers significant advantages for some carbon tracer

experiments. Deuterium has found many applications as a hydrogen tracer, and the use of tritium (H^3) is not entirely equivalent because its properties are even more different from those of protium (H^1).

The uses of isotopic tracers may be classified into two groups: (1) applications in which the tracer is necessary in principle, and (2) applications in which a tracer not necessary in principle may be a great practical convenience. Those applications which depend uniquely on the tracer principle—although the tracers may be either radioactive or separated stable isotopes—may be illustrated by studies of self-diffusion of an element or other substance into itself; no other investigative techniques can give information on such matters. On the other hand, the studies on coprecipitation already mentioned in chapter XII might be done, at least at the higher concentrations, by careful application of conventional chemical methods, or perhaps by spectroscopic or other means of analysis. In some of the more involved applications, particularly in biology, both of these aspects of tracer usefulness appear together.

Self-diffusion. To illustrate the unique tracer method we will discuss first studies that have been made of self-diffusion. By the use of sensitive spectroscopic analyses the rates of diffusion of various metals (including gold, silver, bismuth, thallium, and tin) in solid lead at elevated temperatures have been investigated, but the first attempt (by G. Hevesy and his collaborators) to observe the diffusion of radioactive lead into ordinary lead failed, showing that the diffusion rate must be at least one hundred times smaller than that for gold in lead (which is the fastest of those just named, the others showing decreasing rates in the order listed). The method first used was a rather gross mechanical one, and the workers evolved a much more sensitive method based on the short range of the α particles from ThC in transient equilibrium with ThB. The lead containing ThB isotopic tracer was pressed into contact with a thin foil of inactive lead which was chosen just thick enough to stop all the α rays, and then as diffusion progressed an α activity appeared and increased as measured through this foil. The diffusion coefficient **D** is obtained through a suitable integration of Fick's diffusion law, $\frac{\partial c}{\partial t} = \mathbf{D}\frac{\partial^2 c}{\partial x^2}$, where c is concentration of the diffusing tracer, t is time, and x is the coordinate

along which the diffusion is measured; some typical values for **D** were 0.6×10^{-6} cm^2 per day at 260°C, 2.5×10^{-6} at 300°C, and 47×10^{-6} at 320°C. A similar but even more sensitive technique than the α-range method was based on the very much shorter ranges (a few millionths of a centimeter in lead) of the nuclei recoiling from α emission, with the radioactivity of the resulting ThC″ as an indicator of the emergence of recoil nuclei from the very thin lead foils. At 200°C the diffusion of lead in lead is about ten times slower than that of tin in lead, and roughly 10^5 times slower than that of gold in lead.

The diffusion of bismuth in bismuth has been studied with ThC as tracer; an interesting result is that the diffusion parallel to the c axis of the crystal is very different in magnitude ($\sim 10^5$ times slower at 250°C) and in temperature dependence from that perpendicular to the c axis. Self-diffusion of gold was studied by H. A. C. McKay; he induced activity principally on one surface only of a thin gold disk by an ingenious activation with resonance neutrons, for which the capture cross section in gold is so great that few neutrons could penetrate far below the exposed surface. Self-diffusion of copper has been studied by several investigators, with the use of copper disks either activated on one surface by deuteron irradiation or plated on one side with active copper. Self-diffusions of zinc and of silver have been measured from electroplated surfaces. Diffusion of some ions in crystals has been investigated, for example, of Pb^{++} in $PbCl_2$ and in PbI_2.

The rates of diffusion of ions through aqueous salt solutions of uniform composition may be determined with radioactive tracers, and this information may be of special significance since each rate is a property of the particular ion in that system, whereas salt-diffusion rates under a concentration gradient as ordinarily observed must necessarily depend on the diffusion tendencies of ions of both charges. Diffusion coefficients have been determined for ions such as Na^+, Cl^-, and I^- in salt solutions.

Other Migration Problems. Radioactive tracers are useful in the study of numerous migration problems other than self-diffusion, particularly where movements of very small amounts of material are involved. In most such applications the tracer is serving only as a very sensitive and relatively convenient analytical tool. Erosion and corrosion of surfaces may be measured with great sensitivity if the surface to be tested can be made intensely

radioactive. Transfer of very minute amounts of bearing-surface materials during friction has been studied in this way. Radioactive gases or vapors may be detected in small concentrations, and leakage, flow, and diffusion rates may therefore be studied by the tracer method.

B. EXCHANGE REACTIONS

Qualitative Observations. In a very early exchange experiment in 1920 Hevesy demonstrated by the use of ThB (Pb^{212}) the rapid interchange of lead atoms between $Pb(NO_3)_2$ and $PbCl_2$ in water solution. The experiment was performed by the addition of an active $Pb(NO_3)_2$ solution to an inactive $PbCl_2$ solution and by the subsequent crystallization of $PbCl_2$ from the mixture. The result is not at all surprising because the well-known process of ionization for these salts leads to chemically identical lead ions, Pb^{++}. Hevesy also showed a rapid exchange of the lead atoms between $Pb(C_2H_3O_2)_2$ and $Pb(C_2H_3O_2)_4$ in acetic acid; we may conclude that the plumbous and plumbic forms enter a reversible oxidation–reduction reaction of some sort. Very many exchange systems have been examined since that time, and for the majority of cases where exchange is rapid at ordinary temperature there are known reversible reactions which would lead to interchange. For the other observed exchanges there either exist such reversible reactions, which are possibly unknown, or the exchange occurs by a simple collision mechanism (which may amount to about the same thing).

It was soon shown when artificially radioactive isotopes became available that aqueous Cl^- and Cl_2, Br^- and Br_2, and I^- and I_2 exchanged at room temperature so quickly that the rates could not be measured by ordinary methods. These exchanges are interpreted as occurring through the reactions illustrated by $I^- + I_2 \rightleftharpoons I_3^-$. It has been found that Br_2 and HBr, either in the gas phase or in solution in dry carbon tetrachloride, exchange rapidly at room temperature, probably through reversible formation of a complex HBr_3, although the life of this intermediate may be very short and thus its concentration very low. Also I_2 and SbI_3 in dry pentane exchange within 20 min at 37°C, very possibly through SbI_5. Rapid exchanges at room temperature in carbon tetrachloride are found between Br_2 and $AsBr_3$ and between Br_2

and $SnBr_4$; we may imagine that these proceed through intermediates like $AsBr_5$ and $SnBr_2$, or bromides of other oxidation states. In aqueous solution $PtBr_4^=$ or $PtBr_6^=$ rapidly exchanges all bromine atoms with Br^- ion; the four iodine atoms in $HgI_4^=$ exchange with I^- ion.

In dilute-acid solution at room temperature there is no rapid exchange of halogen atoms between Cl_2 and ClO_3^- or ClO_4^-, Br_2 and BrO_3^-, I_2 and IO_3^-, ClO_3^- and ClO_4^-, IO_3^- and IO_4^-; however, some of these do exchange at measurable rates. No exchange was found in alkaline solution between Cl^- and ClO_4^-, Br^- and BrO_3^-, I^- and IO_3^-.

Interesting exchange studies have been made with the tracer S^{35}. Sulfur and sulfide ions exchange in polysulfide solution. Even at 100°C $S^=$ and $SO_4^=$, $SO_3^=$ and $SO_4^=$, H_2SO_3 and HSO_4^- do not exchange appreciably. If active sulfur is reacted with inactive $SO_3^=$ to form $S_2O_3^=$, and then the sulfur removed with acid, the H_2SO_3 is regenerated inactive; therefore, the two sulfur atoms in thiosulfate are not equivalent. The ions $S_2O_3^=$ and $SO_3^=$ exchange only very slowly at room temperature, but exchange one sulfur fairly rapidly at 100°C. (Notice that this result can be found only by labeling the proper sulfur atom, the one attached directly to the oxygen atoms.) Sulfide ion and $S_2O_3^=$ slowly exchange (probably only one sulfur) at 100°C. Sulfur does not exchange with CS_2 at 100°C; SO_2 and SO_3 do not exchange appreciably even at 280°C. The short-lived C^{11} has been used to show that CO and CO_2 do not exchange in 1 hr at 200°C.

Phosphoric and phosphorous acids, H_3PO_4 and $H_2(HPO_3)$, and also phosphoric and hypophosphorous acids, H_3PO_4 and $H(H_2PO_2)$, do not exchange phosphorus atoms even at 100°C, although the first of these exchanges might be expected to proceed (at some unknown rate) through the formation of hypophosphoric acid, $H_4P_2O_6$. Arsenate and arsenite ions, and H_3AsO_4 and $HAsO_2$ do not exchange appreciably even at 100°C.

The exchange reactions of various manganese compounds have been surveyed. The following do not exchange readily: MnO_4^- and Mn^{++}, MnO_4^- and $Mn(C_2O_4)_3^=$, MnO_4^- and MnO_2, Mn^{++} and MnO_2. Rapid exchanges at room temperature were found for the pairs Mn^{++} and $Mn(C_2O_4)_3^=$ and MnO_4^- and $MnO_4^=$; in these cases oxidation–reduction by electron transfer only is involved. Ferrous and ferric ions transfer an electron (that is,

they exchange) readily in 6 normal HCl, but apparently according to one recent report [1a] they exchange only very slowly in $HClO_4$; this may mean that Fe^{++} and Fe^{+++} ions have difficulty in approaching sufficiently close for the oxidation-reduction reaction because of coulombic repulsion, but in HCl the neutral $FeCl_3$ may react. The two ions, $Fe(CN)_6^{\equiv}$ and $Fe(CN)_6^{\equiv}$, probably exchange rapidly, although the method (precipitation) which has been used for separation might possibly be responsible for the exchange. As an example of this effect, Tl^{+} and Tl^{+++} ions were first reported to exchange rapidly, but recent investigations show that the exchange is a moderately slow bimolecular process inhibited by high concentrations of strong acids; a correction is applied to the experimental data for a partial exchange induced by the precipitation used as a separation method. Again in this case the slowness of the exchange may be due to coulombic repulsion, and the effect of strong acid may be due to the reduction in number of the less-charged partially hydrolyzed forms of Tl(III); the induced exchange during precipitation may proceed through neutralized molecules of transient existence. The mercury ions Hg^{++} and Hg_2^{++} have been reported to exchange readily.

Some metal surfaces exchange rather well with the corresponding metal ions in solution, as observed in the case of silver, zinc, and lead. With silver the exchange reaches to a depth of 10 to 100 atomic layers in about 1 hr at room temperature. In part these effects may be due to local electrolysis caused by imperfections in the metal surface. Precipitates such as AgBr exchange rather effectively with component ions such as Br^{-} when freshly formed, but only much more slowly when aged. An exchange of over 100 per cent completion is simulated in some cases; that is, initially inactive crystals may reach a higher specific activity than that of the final halide solution, because the inner and outer parts of grown crystals may not be in equilibrium.

Alkyl halides of all types ordinarily do not exchange readily at room temperature with either free halogens or halide ions. However, the nature of the solvent (particularly ethyl alcohol, acetone, and amyl alcohol in some reported experiments) can

[1a] *Note added in proof:* We now believe that this exchange is fast even in $HClO_4$. Moreover it appears that Ce(III) and Ce(IV) also exchange rapidly in $HClO_4$, so that the role of coulombic repulsion in electron-transfer exchanges is not clear.

exert a marked influence in producing exchange in these systems. In the solvents named, I^- exchanges with ethyl, *n*-propyl, isopropyl, and methylene iodides and iodoform in about 15 min or less at 100°C. Most remarkable, in these solvents I^- and CH_3I are reported to exchange in 1 min at room temperature; the exchange of CH_3I with I_2 is much less rapid. Iodide ion exchanges rapidly with iodoacetic acid, but rapidly only at elevated temperatures with β-iodopropionic acid. The phenyl halides (including *p*-nitro and *p*-amino derivatives) exchange less readily than the alkyl halides. Halogen exchanges with alkyl halides have been produced photochemically. On the other hand, gaseous HBr and C_2H_5Br did not exchange photochemically but did exchange thermally at 300°C.

The behavior of aluminum bromide in exchange reactions is remarkable, paralleling its extraordinary character as a catalyst. It exchanges bromine atoms readily at room temperature with many alkyl bromides, with benzyl bromide, and with many aliphatic polybromides; it exchanges also, but more slowly, with aryl bromides. Aluminum iodide appears to behave in a similar way. Because these aluminum halides also exchange readily with gaseous halogen or hydrogen halide, a convenient synthesis of labeled organic halides is provided. Obviously the presence of aluminum bromide will catalyze an exchange between two organic bromides.

Knowledge of the occurrence or nonoccurrence of rapid exchange has been used in the study of bond character. It is obvious that exchange data give information on the degree of stability of particular bonds, but the relation of this information to the bond type and to other aspects of bond character is not established at the present time.

Quantitative Exchange Law. Consider a schematic exchange-producing reaction,

$$AX + BX^0 = AX^0 + BX,$$

where X^0 represents a radioactive atom of X. The radioactive decay of this species will be neglected; in practice if the decay is appreciable correction of all measured activities to some common time must be used to avoid error from this condition. The rate of the reaction between AX and BX in the dynamic equilibrium we call **R**, in units of moles liter^{-1} sec^{-1}; notice that **R** is quite

independent of the concentration and even of the existence of the active tracer X^0. We indicate mole-per-liter concentrations as follows: $(AX) + (AX^0) = a$, $(BX) + (BX^0) = b$, $(AX^0) = x$, $(BX^0) = y$ and $x + y = z$. The rate of increase $\left(\frac{dx}{dt}\right)$ of (AX^0) is given by the rate of its formation minus the rate of its destruction. The rate of formation of AX^0 is given by $\mathbf{R}$ times the factor y/b, which is the fraction of reactions that occur with an active molecule BX^0, and times the factor $(a - x)/a$, which is the fraction of reactions with the molecule AX initially inactive. The rate of destruction of AX^0 is given by $\mathbf{R}$ times the factor x/a, which is the fraction of reactions in the reverse direction that occur with an active molecule AX^0, and times the factor $(b - y)/b$, which is the fraction of reverse reactions with the molecule BX initially inactive. The differential equation is then

$$\frac{dx}{dt} = \mathbf{R}\frac{y}{b}\cdot\frac{(a-x)}{a} - \mathbf{R}\frac{x}{a}\frac{(b-y)}{b}$$

$$= \frac{\mathbf{R}}{ab}(ay - bx) = \frac{\mathbf{R}}{ab}(az - ax - bx);$$

$$\frac{dx}{dt} = -\frac{a+b}{ab}\mathbf{R}x + \frac{z}{b}\mathbf{R}.$$

The solution of this first-order linear equation is found by standard methods:

$$x = Ce^{-\frac{a+b}{ab}\mathbf{R}t} + \frac{a}{a+b}z.$$

After a very long time, that is at $t = \infty$, let $x = x_\infty$ and $y = y_\infty$; substituting these values in the previous solution we have then

$$x_\infty = \frac{a}{a+b}z,$$

and, since $y_\infty = z - x_\infty$,

$$y_\infty = \frac{b}{a+b}z.$$

These two relations constitute an algebraic expression for the reasonable and well-known rule that when exchange is complete the specific activity (activity per mole or per gram of X) is the same in both fractions; at that time the specific activity of AX is $\frac{x_\infty}{a} = \frac{z}{a+b}$, and the specific activity of BX is $\frac{y_\infty}{b} = \frac{z}{a+b}$.

The solution may now be rewritten:

$$x = Ce^{-\frac{a+b}{ab}\mathbf{R}t} + x_\infty.$$

If at $t = 0$ we have $x = 0$, that is, if AX is inactive at the start, we find the constant $C = -x_\infty$, and some useful (final) forms may be obtained:

$$1 - \frac{x}{x_\infty} = e^{-\frac{a+b}{ab}\mathbf{R}t},$$

$$2.303 \log\left(1 - \frac{x}{x_\infty}\right) = -\frac{a+b}{ab}\mathbf{R}t; \qquad \text{(XIII-1)}$$

and, by differentiation with respect to t,

$$\mathbf{R} = -2.303\frac{ab}{a+b}\frac{d}{dt}\log\left(1 - \frac{x}{x_\infty}\right).$$

The last result shows that **R** may be evaluated from the slope of a plot of $\log [1 - (x/x_\infty)]$ vs. t. Probably the most convenient procedure is to plot $[1 - (x/x_\infty)]$ on semilog paper against t, read off the half-time $T_{1/2}$ at which the fraction exchanged, x/x_∞, is $1/2$, and find **R** from an equation derived immediately from equation (XIII-1):

$$\mathbf{R} = \frac{ab}{a+b} \cdot \frac{0.69315}{T_{1/2}}.$$

It is important to notice that if a or b or both should be varied the variation in half-time for the exchange would not directly reflect the variation in **R**, because of the factor $\frac{ab}{a+b}$.

For a number of practical exchange studies the simple formulas AX and BX may not represent the reacting molecules; for example, AX_2 or BX_n might be involved. So long as the several atoms of X are entirely equivalent (or at least indistinguishable in exchange experiments) in each of these molecules, the equations just derived may be applied without modification, provided only that we redefine all the concentrations in gram atoms of X per liter rather than moles of AX or AX_2, etc., per liter. This is equivalent to considering (for this purpose only) one molecule of AX_2 as replaceable by two molecules of $A_{1/2}X$, etc., in the derivation. If in a molecule like AX_2 the two X atoms are not equivalent, and if they exchange through two different reactions with rates $\mathbf{R}_1$ and $\mathbf{R}_2$, it may be seen that the resulting semilog plot will be not a straight line but a complex curve. The differential equations for the exchanges to the several positions may be set up and solved simultaneously, so that the curve may, at least in principle, be resolved to give values for the several $\mathbf{R}$'s; however, this becomes very difficult for more than about two rates. A simplification may be made if $a \ll b$, with the several nonequivalent positions in the molecule AX_n; here the value of y is very nearly a constant, and in this limit the complex semilog curve is resolvable in the same way as a radioactive decay curve into straight lines measuring $\mathbf{R}_1$, $\mathbf{R}_2$, etc. No example of a complex exchange curve has been reported except the limiting case with $\mathbf{R}_1$ measurable, but $\mathbf{R}_2 = 0$ within experimental accuracy (like the sulfur exchange between $S_2O_3^=$ and $SO_3^=$). In this limiting case no unusual feature appears if x_∞ is used in an experimental sense, although after a much longer time x_∞ may be expected to reach a higher value.

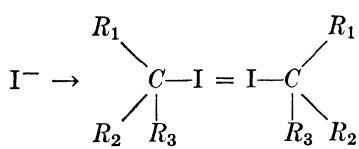
Reaction Kinetics and Mechanisms. Radioactive tracers are sure to find an important place in the investigations of reaction kinetics and mechanisms. We will discuss several examples to illustrate the kinds of information in this field that can be obtained with tracers but hardly in any other way. Consider the reversible reaction:

$$HAsO_2 + I_3^- + 2H_2O \rightleftharpoons H_3AsO_4 + 3I^- + 2H^+.$$

The familiar theory of dynamic equilibrium takes $K = k_f/k_r$, where K is the equilibrium constant and k_f and k_r are the specific

rate constants of the forward and reverse reactions. Ordinarily K may be measured only at equilibrium and k_f or k_r far from equilibrium. Using radioactive arsenic to measure the rate of exchange between arsenious and arsenic acids induced by an iodine catalyst in accordance with the foregoing equilibrium reaction, J. N. Wilson and R. G. Dickinson were able to find the rate law and specific rate constant at equilibrium. For the reverse direction as written they found $\mathbf{R} = k_r\,(H_3AsO_4)(H^+)(I^-)$, with $k_r = 0.057$ liter^2 mole^{-2} min^{-1}, which is in satisfactory agreement with the information from ordinary rate studies made far from equilibrium, $\mathbf{R} = 0.071\,(H_3AsO_4)(H^+)(I^-)$.

A theory of the Walden inversion calls for inversion at each substitution by the schematic mechanism:



As shown here the substitution is by a like group, and if the initial molecules are optically active the final product will be the racemic mixture. It has been shown that for sec-octyl iodide (or for α-phenyl ethyl bromide) the rate of exchange with radioactive iodide ion (or bromide ion) is identical with the rate of racemization, which is a verification of the mechanism.

A different type of racemization is that of chromioxalate ion, $Cr(C_2O_4)_3^{\equiv}$, which may be optically active through different linkings of the six octahedral bonds of the chromium with the carbon–oxygen chains. This racemization in aqueous solution is fairly rapid and apparently first order; it had been proposed that the mechanism involved an ionization as the rate-determining step:

$$Cr(C_2O_4)_3^{\equiv} = Cr(C_2O_4)_2^{-} + C_2O_4^{=}.$$

Another theory favored an intramolecular rearrangement instead. The racemization has been allowed to proceed in a solution containing radioactive $C_2O_4^{=}$ (prepared with C^{11}). The activity did not enter the chromium complex compound; therefore, the ionization is disproved, and the intramolecular rearrangement hypothesis supported.

C. APPLICATIONS TO ANALYTICAL CHEMISTRY

Test of Separations. Radioactive tracers can be very conveniently used to follow the progress and test the completeness of chemical separation procedures. If one component of a mixture is radioactive, frequently it can be followed satisfactorily through successive operations if beakers containing filtrates, funnels with precipitates, and so on, are merely held near a counter or ionization chamber. We have seen good chemical isolations made by these methods in the almost complete absence of knowledge of specific chemical properties. The crude qualitative procedure may be refined as far as desired, and valuable tests of analytical separation methods have been made with tracers. As the first of a few examples we consider a procedure that has been used for the determination of gold–platinum–iridium compositions. The three elements in solution are precipitated by reduction with hot alkaline sodium formate; the residue after ignition is treated with aqua regia to dissolve gold and platinum but leave iridium; the resulting solution on treatment with hydrogen peroxide gives the gold as a precipitate of the metal, and the platinum is finally precipitated from the filtrate with sodium formate. By simple gravimetric studies on known compositions the iridium fraction was found to be too heavy, the gold fraction too heavy, and the platinum fraction too light. With radioactive tracer it was shown that the gold fraction actually contained only 97 per cent of the original gold, but with more than enough platinum to mask this; the remainder of the gold was mostly in the platinum fraction.

The coprecipitation of cobalt with SnS_2 has been investigated in relation to various experimental conditions, with active cobalt as a tracer. The amount of coprecipitation was smaller at higher hydrogen ion concentrations; the cobalt contamination could be made negligible provided acrolein was present as a flocculating agent. Radioactive beryllium has been used to find that aluminum precipitated by 8-hydroxyquinoline carries some beryllium at *p*H values greater than 6; below *p*H 6 the coprecipitation is absent. The carrying down of tellurium by antimony oxide precipitated from boiling concentrated nitric acid has been studied with tracers. Also a number of solvent extraction procedures have been tested in this way for interference effects.

The solubility of quite insoluble precipitates may be judged by the use of radioactivity. This has been done in the precipitation of tin as $Sn_3[Fe(CN)_6]_2$. The approach to equilibrium between solid phase and solution can be followed very conveniently by repeated measurements of the specific activity of the supernatant solution. A somewhat analogous tracer method is applicable to the determination of small vapor pressures.

Analysis by Isotope Dilution. It may frequently occur that quantitative analysis for a component of a mixture is wanted where no quantitative isolation procedure is known. Particularly for complex organic mixtures it may be possible to isolate from the unknown the desired compound with satisfactory purity but only in low and uncertain yield. In such a case the analysis may be made by the technique of isotope dilution. To the unknown mixture is added a known weight of the compound to be determined containing a known amount (activity) of radioactively tagged molecules. Then the specific activity of that pure compound isolated from the mixture is determined and compared with that of the added material; the extent of dilution of the tracer shows the amount of inactive compound present in the original unknown. (You may think of the tracer as serving to measure the chemical yield of the isolation procedure. Obviously exchange reactions which would reduce the specific activity of the compound must be absent.) To date this powerful method has found uses principally in biochemistry and biology.

Analysis by Activation. Throughout most of the tracer work discussed radioactive isotopes are assayed by measurement of their activities; this is actually an analytical procedure, but we have not emphasized that aspect because the samples are subject to analysis only if the tracer was provided earlier in the experiment. Of course, the naturally radioactive elements, and this includes besides uranium, thorium, radium, other elements, particularly potassium and rubidium, may be assayed by radioactive measurement; a very practical although not very sensitive procedure for potassium assay by means of its radioactivity has been reported.

A somewhat different technique can be useful, in which an unknown sample is subjected to activation by neutrons, deuterons, or other irradiation for appropriately chosen lengths of time, and chemical elements are identified and assayed by analysis of the

probably complex decay curve resulting. Some practical uses of this technique of analysis by activation have been made. By deuteron irradiation very small impurities of gallium in iron, copper in nickel, and iron in cobalt oxide have been found. With neutron activation small amounts of phosphorus and sulfur have been detected, and also hafnium in zirconium, europium in gadolinium, and dysprosium in yttrium. Analyses for sodium by neutron activation can be very convenient when there are not many other elements present which give strong activities of comparable half-life. In all these procedures the method may be standardized by the use of known samples, and in some cases where the cross sections are known or may be guessed with confidence absolute orders of magnitude may be obtained without standardization.

Radiometric Analysis. Analytical procedures by tracer methods for elements which are not themselves radioactive have been introduced and given the name radiometric analysis. For example, silver ion may be determined by the precipitation of AgI with radioactive iodide ion. A recent report describes the collection by adsorption on $Fe(OH)_3$ of very small amounts of AgI formed, so that as little as 10 ppm of silver could be detected. In another procedure almost invisible amounts of TlI are centrifuged onto a plate for counting; either Tl^+ or I^- might be determined by the use of the other in concentrated radioactive form.

D. RADIOCARBON TRACER STUDIES

The Special Importance of Tracer Carbon. The special importance of carbon compounds in chemistry and biochemistry give a special importance to radioactive carbon tracer. When only the 20-min C^{11} was available, serious limitation was obviously imposed on the location and duration of tracer studies with this element. However, in spite of this limitation some remarkable experiments were carried out; for example, S. Ruben, M. D. Kamen, and W. Z. Hassid studied photosynthesis by preparing C^{11} in a Berkeley cyclotron, carrying the boron target to another building, removing the active carbon and forming $C^{11}O_2$, allowing growing plants to assimilate this and photosynthesize compounds from it, then isolating dozens of compounds in attempts

to identify the active intermediates, and even taking the active plant extracts to Palo Alto for determinations of the molecular weight range (~1000) of the active substances in a Stanford University ultracentrifuge.

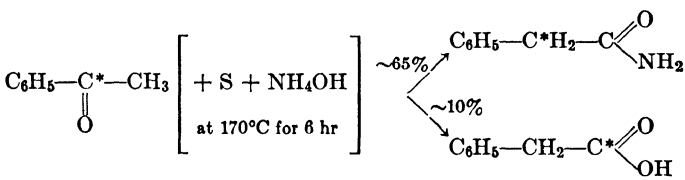
In 1940 Ruben and Kamen discovered the long-lived C^{14} from d, p reaction on the rare isotope C^{13}; however, the very small activities obtainable even at great cost prevented any widespread use of the material at that time. Today this important isotope is made from nitrogen by N^{14} (n, p) C^{14} in the Oak Ridge, Tenn., chain reactor, and is available for purchase from the U. S. Atomic Energy Commission at the price of $50 per millicurie. One millicurie, or 37 rd, of pure C^{14} would weigh very nearly 0.25 mg, if the half-life is taken as 6400 years. As ordinarily supplied this amount of active carbon is mixed with several milligrams of ordinary carbon. In many ways C^{14} is a very excellent tracer and because it emits no γ rays and only soft β rays is easily shielded during chemical operations; however, the small penetration of the radiation does necessitate some special techniques of activity measurement.

Some Typical Results. P. Nahinsky, C. N. Rice, Ruben, and Kamen studied the oxidation by alkaline permanganate of propionate to the products carbonate and oxalate (1 mole of each from 1 mole of propionate). It might have been a plausible guess that the $CO_3^=$ is formed from the carboxyl group; however, with the carboxyl carbon labeled they found that only about 25 per cent of the $CO_3^=$ was from that part of the molecule. In acid solution in the oxidation of propionic acid by dichromate they found that all the CO_2 did originate from the —COOH, demonstrating different mechanisms in the two instances. The oxidation of fumaric acid,[(2)] HOOC*CHCHC*OOH, by acid permanganate has been

[2] By common usage these asterisks indicate labeled positions (the two carboxyl carbons in this case) in the molecules; of course, because the active tracers are almost always very highly diluted with ordinary atoms it is very improbable for any given molecule actually to contain two radioactive atoms. Thus the asterisk denotes not a radioactive atom, but an atom taken at random from a sample containing some active atoms; in other words, the position marked with the asterisk will in some of the molecules be labeled with a radioactive atom. Notice that on page 285 we avoided the asterisk and chose a different superscript because there we wished to indicate an actual radioactive atom.

investigated; the product HCOOH (1 mole per mole of fumaric acid) is formed always from one of the secondary carbon atoms, and the CO_2 (3 moles per mole of fumaric acid) is from the carboxyl carbons and the other secondary carbon.

W. G. Dauben, J. C. Reid, P. E. Yankwich, and M. Calvin followed the mechanism of the Willgerodt reaction on acetophenone, and obtained this result:



Notice the positions of the tracer in the two products; clearly the two are not formed through the same mechanism, and one does not result entirely from the other by hydrolysis as had previously been supposed.

At the present moment C^{14} is finding extensive use in the preparation or study of amino acids, carcinogens, sugars, antibiotics, hormones, vitamins, fuel hydrocarbons, and very many other important classes of organic compounds.

EXERCISES

1. A mixture is to be assayed for penicillin. You add 10.0 mg of penicillin of specific activity 15.0 mrd per mg (possibly prepared by biosynthesis). From this mixture you are able to isolate only 0.35 mg of pure crystalline penicillin, and you determine its specific activity to be 1.3 mrd per mg. What was the penicillin content of the original sample?

2. Refer to the information on fumaric acid oxidation on page 293. The authors of that report (M. B. Allen and S. Ruben) in the experiment measured the specific activities of the original fumaric acid, the CO_2 evolved, and the formic acid remaining. If the C^{11} activity of the original acid had been 20,000 counts per min per mg of carbon at 6:00 P.M., what would they have found for the specific activity of the CO_2 if measured at 7:22 P.M., and of the HCOOH if measured at 8:30 P.M.? What must have been the activity per milligram of carbon of the KC*N used to synthesize the fumaric acid from dichloroacetylene, corrected to 6:00 P.M.?

Answer to last part: 40,000 counts min^{-1} mg^{-1}.

3. Approximately what fraction of a gram of copper may be detected by the tracer method if the tracer copper (Cu^{64}) has been prepared by irradiation of copper with 14-Mev deuterons? See table C in the appendix and chapters IV, VII, and VIII.

Answer: We estimate $\sim 10^{-12}$ g for a thick copper target, less for a thin one.

4. Would you expect almost complete exchange, and by what mechanism, in 1 hr at room temperature, between the members of each of the following pairs:

(*a*) Ag^+, Ag^{++}
(*b*) Hg^{++}, very finely divided Hg
(*c*) $Cr^*O_4^=$, CrO_2^-
(*d*) $Cr^*O_4^=$, $Cr_2O_7^=$
(*e*) CH_3I^*, C_6H_5I
(*f*) $Hg^*(CH_3)_2$, $Hg(C_2H_5)_2$

5. A series of 10-mg sodium fluoride samples containing 0.1, 0.01, 0.001, and 0.0001 per cent sodium iodide, respectively, have been mixed up before they were labeled. No fraction of the samples is available for chemical analysis, and it has therefore been suggested that they be identified by slow-neutron activation. Is this feasible with a thermal-neutron flux of 10^{11} neutrons per cm^2 per sec available?

6. The completeness of the separation of 10 mg of Ni from 100 mg of Co by dimethylglyoxime precipitation is to be tested with the use of a sample of 5-year Co^{60} whose specific activity is 2 rd per g of cobalt. Estimate the minimum amount of coprecipitated cobalt that can be detected.

7. The exchange between I_2^* and IO_3^- has been studied under these conditions: $(I_2) = 0.00050$ mole per liter (formal), $(HIO_3) = 0.00100$ formal, $(HClO_4) = 1.00$ formal, at 50°C. At specified times samples were taken and measured for total (I_2 plus IO_3^-) radioactivity by γ counting; these counting rates corrected to the time $t = 0$ on the basis of the 8.0-day half-life of I^{131} are given below in the column "Corrected Total Activity." The I_2 fractions were removed by extraction with CCl_4 and the residual (IO_3^-) radioactivity measured and corrected in the same way; these rates are in the column "Corrected IO_3^- Activity."

Time (hours)	Corrected Total Activity	Corrected IO_3^- Activity
0.9	1680	9.9 ± 3.0
19.1	1672	107 ± 4.1
47.3	1620	246 ± 6.6
92.8	1653	438 ± 9.4
169.2	1683	610 ± 13
"∞"	1640	819 ± 9.8

Find the half-time $T_{1/2}$ for the exchange and the rate **R** of the exchange reaction. *Answer:* $T_{1/2} = 90.6$ hr; $\mathbf{R} = 3.83 \times 10^{-6}$ moles liter^{-1} hr^{-1}.

8. The experiment described in exercise 7 was repeated but with this difference, $(I_2) = 0.00100$ formal. The results are tabulated as before.

Time (hours)	Corrected Total Activity	Corrected IO_3^- Activity
0.9	1717	7.6 ± 2.7
19.1	1483	70.1 ± 3.6
47.3	1548	178 ± 5.2
92.8	1612	305 ± 7.3
169.2	1587	413 ± 9.8
"∞"	1592	534 ± 5.7

For these conditions find $T_{1/2}$ and **R**. What is the apparent order of the exchange reaction with respect to I_2? *Note:* Do not be surprised if the order is not an integer; according to O. E. Meyers and Kennedy, this order is consistent with this rate law for the exchange-producing reaction: $\mathbf{R} = k(I^-)(H^+)^3(IO_3^-)^2$.

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APPENDIX

TABLE A

Radioactive and Stable Isotopes of the Elements

In this table are listed all the nuclear species (nuclides) whose existence has been fairly reliably established, together with the best available published information on some of their characteristics. Publications received before February 1, 1949, have been covered. Data from the following earlier compilations have been used extensively in the preparation of this table:

G. T. Seaborg, "Table of Isotopes," *Rev. Modern Phys.* **16**, 1 (1944).[1]

"Nuclei Formed in Fission: Decay Characteristics, Fission Yields, and Chain Relationships" (issued by the Plutonium Project), *JACS* **68**, 2411 (1946).

H. A. Bethe, "Table of Nuclear Species" in *Elementary Nuclear Theory*, John Wiley & Sons, New York, 1947.

"1947 Summary of Nuclear Data" compiled by Nuclear Data Committee of Clinton National Laboratory, *Nucleonics* **2** no. 5, 81 (May 1948).

Column 1 gives the atomic number and column 2 the chemical symbol of the element with the mass number as a superscript. If two or three mass numbers are listed for one species, this indicates that the activity in question may be assigned to one of these mass numbers. A question mark in the superscript after the mass number(s) indicates uncertainty in the mass assignment. A question mark after the symbol *and* mass number indicates uncertainty in the chemical identification of the activity. Whenever a nuclide is known to be an upper isomeric state this fact is indicated by underlining of the mass number(s). The common designations for the members of the natural radioactive families (such as RaE for Bi^{210}) are given in parentheses after the designation by element and mass number.

Column 3 lists per cent abundances for the stable and long-lived naturally occurring radioactive nuclides. Column 4 gives isotopic masses, that is atomic masses on the physical atomic weight scale; most of these values are taken from Bethe's table quoted previously.

Half-lives are listed in column 5. In a few cases several different values reported in the literature are listed for a given activity. The symbols used

[1] A revised "Table of the Isotopes," by G. T. Seaborg and I. Perlman, is scheduled for publication in *Reviews of Modern Physics* for October, 1948. A copy of this table was kindly made available to us by Professor Seaborg prior to publication, and some previously unpublished data from that source are included here.

in this column are y = years, d = days, h = hours, m = minutes, s = seconds, μs = microseconds.

Column 6 lists the modes of decay and the energies of the radiations emitted. The symbols used are:

α	alpha particles,
β^-	negative beta particles (negatrons),
β^+	positive beta particles (positrons),
γ	gamma rays,
K	electron capture (not necessarily restricted to K-electron capture),
IT	isomeric transition,
e^-	internal-conversion electrons,
n	neutrons.
x	X rays

The numbers following the symbols in this column indicate the measured energies in million electron volts; in case of β^- and β^+ the energies listed are the maximum beta-particle energies for each transition. An attempt has been made to list different modes of decay or different energies in order of decreasing abundance where the relative abundances are known. Radiations found in less than 1 per cent of the disintegrations are given in parentheses.

Column 7 lists some suggested modes of formation. In most cases not all the nuclear reactions by which the given nuclide has been produced are listed. On the other hand, some methods of production whose use has not been reported are sometimes included if they appear obviously suitable for the preparation of the nuclide in question. Different methods listed for the preparation of one radionuclide may be advantageous for different purposes, for example, to give good yields, or high specific activities, or minimum interference from other radionuclides. In the choice of suggested modes of formation, irradiation of natural isotopic mixtures only has been considered; the use of ultrahigh-energy projectiles (say >20 or 30 Mev) has been disregarded except where it offers the only good method of producing the nuclide in question. When one radionuclide can be produced by the radioactive decay of another this has been indicated by such symbols as Zn^{73}-β^-, which means "produced by β^- decay of Zn^{73}." The uranium fission reaction is represented by U (n, f), but this symbol is used only for the first (known) members of chains of fission products. For activities existing in nature by virtue of their own long lives the word "natural" appears in this column.

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
1	H^1	99.98	1.008123			
	H^2	0.02	2.014708			
	H^3		3.01702	12y	β^-0.0179 No γ	Li^6 (n, α) Be^9 $(d, 2\alpha)$
2	He^3	1.3×10^{-4} (atm.) 1.6×10^{-5} (well)	3.01700			
	He^4	100	4.00390			
	He^6		6.0209	0.87s	β^-3.7, No γ	Be^9 (n, α)
3	Li^6	7.30	6.01697			
	Li^7	92.70	7.01822			
	Li^8		8.02502	0.88s	β^-12.7, 15.8	Li^7 (d, p) B^{11} (n, α)
4	Be^7		7.01916	52.9d	K, γ0.478	Li^7 $(d, 2n)$ Li^7 (p, n) Li^6 (d, n)
	Be^8		8.00791	$10^{-15} - 10^{-17}$s	2α0.055 each	Be^9 (γ, n)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Be^{9}	100	9.01503			
	Be^{10}		10.01677	2.7×10^{6}y	β^{-}0.56 No γ	Be^{9} (d, p) B^{10} (n, p)
5	B^{10}	18.83	10.01618			
	B^{11}	81.17	11.01284			
	B^{12}		12.0183	0.022s	β^{-}13.4	B^{11} (d, p)
6	C^{10}		10.0210	19s	β^{+}2.2 $\gamma \sim 1$	B^{10} (p, n) B^{10} $(d, 2n)$
	C^{11}		11.01495	20.5m	β^{+}0.99 No γ	B^{11} $(d, 2n)$ B^{10} (d, n) B^{11} (p, n)
	C^{12}	98.9	12.00382			
	C^{13}	1.1	13.00758			
	C^{14}		14.00767	6400 $\pm$ 200y (5100 $\pm$ 150y)	β^{-}0.156 No γ	N^{14} (n, p)
7	N^{13}		13.00967	10.1m	β^{+}1.24 No γ	C^{12} (d, n)

	N^{14}	99.62	14.00751			
	N^{15}	0.38	15.00493			
	N^{16}		16.010	7.35s	β^-3.8, 4.3, 10.3, 5.2 γ6.2, 6.7	N^{15} (d, p) O^{16} (n, p)
	N^{17}			4.14s	β^-3.7 n0.9 (mean)	F^{19}(d, ?)
8	O^{14}			76.5s	β^+1.8, γ2.3	N^{14} (p, n)
	O^{15}		15.0078	126s	β^+1.7	N^{14} (d, n)
	O^{16}	99.757	16.00000			
	O^{17}	0.039	17.00450			
	O^{18}	0.204	18.0049			
	O^{19}		19.0087	27.0s	β^-2.9, 4.5 γ1.6	F^{19} (n, p)
9	F^{17}		17.0075	70s	β^+2.1	O^{16} (d, n) N^{14} (α, n)
	F^{18}		18.0065	110m	β^+0.60, 0.95 K? γ1.4?	O^{18} (d, $2n$) Ne^{20} (d, α) O^{18} (p, n)
	F^{19}	100	19.00450			
	F^{20}			12s	β^-5.0 γ2.2	F^{19} (d, p) F^{19} (n, γ)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
10	Ne^{19}		19.00781	18.2s	$\beta^+2.20$	F^{19} (p, n) F^{19} $(d, 2n)$
	Ne^{20}	90.51	19.99877			
	Ne^{21}	0.28	20.99963			
	Ne^{22}	9.21	21.99844			
	Ne^{23}		23.00213	40.7s	$\beta^-4.1$	Na^{23} (n, p) Mg^{26} (n, α)
11	Na^{21}		21.0035	23s		Ne^{20} (d, n) Ne^{21} (p, n)
	Na^{22}		22.0015	3.0y (2.6y)	$\beta^+0.575$ $\gamma 1.30$	Mg^{24} (d, α) F^{19} (α, n)
	Na^{23}	100	22.99618			
	Na^{24}		23.9976	14.8h	$\beta^-1.39$ $\gamma 1.38, 2.76$	Na^{23} (d, p) Na^{23} (n, γ) Mg^{24} (n, p)
	Na^{25}		24.9972	59s	$\beta^-3.7, 2.7$ $\gamma 1.0$	Mg^{25} (n, p) Mg^{26} (γ, p)
12	Mg^{23}		23.0002	11.6s	$\beta^+2.82$	Na^{23} $(d, 2n)$ Na^{23} (p, n)

	Mg^{24}	78.60	23.9925			
	Mg^{25}	10.11	24.9938			
	Mg^{26}	11.29	25.9898			
	Mg^{27}		26.9925	10.2m	β^-1.78, 0.79 γ1.02, 0.84, 0.64	Mg^{26} (d, p) Mg^{26} (n, γ) Al^{27} (n, p)
13	Al^{25}		24.9981	7.3s	β^+	Mg^{25} (p, n)
	Al^{26}		25.9929	6.3s	β^+2.99	Mg^{26} (p, n) Na^{23} (α, n)
	Al^{27}	100	26.9899			
	Al^{28}		27.9908	2.4m	β^-3.01 γ1.80	Al^{27} (d, p) Al^{27} (n, γ) P^{31} (n, α)
	Al^{29}		28.9893	6.7m	β^-2.5	Mg^{26} (α, p)
14	Si^{27}		26.9949	4.9s	β^+3.6	Al^{27} $(d, 2n)$ Al^{27} (p, n)
	Si^{28}	92.28	27.9866			
	Si^{29}	4.67	28.9866			
	Si^{30}	3.05	29.9832			
	Si^{31}		30.9862	170m	β^-1.8 No γ	P^{31} (n, p) Si^{30} (d, p) Si^{30} (n, γ)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
15	P^{29}		28.9919	4.6s	$\beta^+3.63$	Si^{28} (d, n) Si^{29} (p, n)
	P^{30}		29.9873	2.55m	$\beta^+3.5$	S^{32} (d, α) Al^{27} (α, n)
	P^{31}	100	30.9843			
	P^{32}		31.98249	14.30d	$\beta^-1.712$ No γ	P^{31} (d, p) P^{31} (n, γ) Cl^{35} (n, α) S^{32} (n, p)
	P^{34}		33.9826	12.4s	$\beta^-5.1, 3.2$ $\gamma 1.9$	Cl^{37} (n, α) S^{34} (n, p)
16	S^{31}		30.9899	3.2s (2.6s)	$\beta^+3.85$	P^{31} $(d, 2n)$ P^{31} (p, n)
	S^{32}	95.1	31.98089			
	S^{33}	0.74	32.9800			
	S^{34}	4.2	33.97710			
	S^{35}		34.97883	87.1d	$\beta^-0.169$	Cl^{35} (n, p) Cl^{37} (d, α) S^{34} (n, γ) S^{34} (d, p)

	S^{36}	0.016	35.978			
	S^{37}		36.9821	5.04m	β^-1.6, 4.3 γ2.7	Cl^{37} (n, p)
17	Cl^{33}		32.9860	2.4s	β^+4.13	S^{32} (d, n)
	Cl^{34}		33.9828	33m	β^+5.1, 2.4 γ3.4	P^{31} (α, n) S^{34} $(d, 2n)$ S^{34} (p, n)
	Cl^{35}	75.4	34.97867			
	Cl^{36}		35.9788	2×10^6y	β^-0.66, (β^+) K	Cl^{35} (n, γ) Cl^{35} (d, p) K^{39} (n, α)
	Cl^{37}	24.6	36.97750			
	Cl^{38}		37.981	38m	β^-4.94, 1.19, 2.79 γ1.60, 2.15	Cl^{37} (d, p) Cl^{37} (n, γ) K^{41} (n, α)
	Cl^{39}			1h	β^-	
18	A^{35}		34.9850	1.88s	β^+4.4	Cl^{35} (p, n) Cl^{35} $(d, 2n)$ S^{32} (α, n)
	A^{36}	0.35	35.9780			
	A^{37}		36.9777	34.1d	K	Cl^{37} $(d, 2n)$ K^{39} (d, α) Cl^{37} (p, n) Ca^{40} (n, α)

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	A^{38}	0.08	37.974			
	A^{40}	99.57	39.9756			
	A^{41}		40.9770	109m	β^-1.18, (2.55) γ1.37	A^{40} (d, p) A^{40} (n, γ) K^{41} (n, p)
19	K^{37}?		36.9830	1.3s		K^{39} $(\gamma, 2n)$?
	K^{38}		37.9795	7.5m	β^+2.53 γ2.1	Cl^{35} (α, n) Ca^{40} (d, α)
	K^{39}	93.18	38.9747			
	K^{40}	0.012	39.9760	4.5×10^8y $(1.3 \times 10^9$y)	β^-1.4 K, γ1.55	Natural
	K^{41}	6.82	40.974			
	K^{42}			12.4h	β^-3.57, 2.05 γ1.51	Sc^{45} (n, α) K^{41} (d, p) K^{41} (n, γ)
	K^{43}			22.4h	β^-0.24, 0.81 γ0.4	A^{40} (α, p)
	$K^{43,44}$			18m (27m)	β^-	Ca (n, p)

20	Ca^{39}			1.06s	β^+	K^{39} $(d, 2n)$ K^{39} (p, n) Ca^{40} $(n, 2n)$
	Ca^{40}	96.96	39.9753			
	Ca^{41}			$\ll$1d or $>$1y		Ca^{40} (d, p) Ca^{40} (n, γ)
	Ca^{42}	0.64	41.9711			
	Ca^{43}	0.15	42.9723			
	Ca^{44}	2.06				
	Ca^{45}			152d (180d)	β^-0.260	Sc^{45} (n, p) Ca^{44} (d, p) Ca^{44} (n, γ)
	Ca^{46}	0.0033				
	Ca^{47}?			5.8d	β^-1.1 γ1.3	Ti^{50} (n, α)?
	Ca^{48}	0.19				
	Ca^{49}			2.5h	β^-2.3 γ0.8	Ca^{48} (d, p) Ca^{48} (n, γ)
	Ca^{49}			30m	β^-	Ca^{48} (d, p) Ca^{48} (n, γ)
21	Sc^{41}			0.87s	β^+4.94	Ca^{40} (d, n)
	Sc^{43}			3.92h	β^+1.13 γ1.65	Ca^{40} (α, p) Ca^{42} (d, n)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Sc^{44}			3.92h	β^+1.33, K? γ1.33	K^{41} (α, n) Sc^{45} (n, $2n$)
	$Sc^{\underline{44}}$			2.44d	IT e^-, γ0.27	K^{41} (α, n) Sc^{45} (n, $2n$)
	Sc^{45}	100	44.9669			
	Sc^{46}			85d	β^-0.36, 1.49 γ0.89, 1.12	Sc^{45} (n, γ) Sc^{45} (d, p) Ti^{48} (d, α)
	$Sc^{\underline{46}}$			20s	IT 0.18 γ0.18, e^-0.165	Sc^{45} (n, γ)
	Sc^{47}			3.4d	β^-0.46	Ca^{44} (α, p)
	Sc^{48}			44h	β^-0.57 γ1.33, 0.98	V^{51} (n, α) Ti^{48} (n, p)
	Sc^{49}			57m	β^-1.8 No γ	Ti^{49} (n, p) Ca^{49}-β^- (2.5h)
22	Ti^{43}?			0.58s		Ca^{40} (α, n)?
	Ti^{45}			3.08h	β^+1.2	Sc^{45} (d, $2n$) Sc^{45} (p, n) Ti^{46} (n, $2n$)
	Ti^{45}?			21d		Sc^{45} (p, n)?

	Ti^{46}	7.95	45.9661			
	Ti^{47}	7.75	46.9647			
	Ti^{48}	73.45	47.9631			
	Ti^{49}	5.51	48.9646			
	Ti^{50}	5.34	49.9621			
	Ti^{51}		50.9587	72d	$\beta^-0.45$ $\gamma 1.02$	Ti^{50} (d, p) Ti^{50} (n, γ) V^{51} (n, p)
	$Ti^{5\underline{1}}$			6m	$\beta^-1.6$ γ	Ti^{50} (d, p) Ti^{50} (n, γ) V^{51} (n, p)
23	V^{47}			33m	$\beta^+1.9$	Ti $(d, ?)$ Ti (p, n)
	V^{48}			16d	$\beta^+0.716$, K $\gamma 1.33$, 0.98	Sc^{45} (α, n) Ti^{48} $(d, 2n)$ Ti^{48} (p, n)
	$V^{49,50?}$			600d	K, no β^+ No γ	Ti $(d, ?)$
	V^{51}	100	50.9577			
	V^{52}			3.74m	$\beta^-2.7$ $\gamma 1.44$ $e^-0.25$, IT?	V^{51} (n, γ) V^{51} (d, p) Mn^{55} (n, α)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
24	Cr^{49}			42m	$\beta^+1.45$ $\gamma 0.18, 1.55$	Ti^{46} (α, n) Cr^{50} $(n, 2n)$
	Cr^{50}	4.31	49.9642			
	Cr^{51}		50.958	26.5d	K, no β^+ $\gamma 0.32$, $e^-0.2$	V^{51} $(d, 2n)$ V^{51} (p, n) Ti^{48} (α, n) Cr^{50} (n, γ)
	Cr^{52}	83.76	51.9557			
	Cr^{53}	9.55	52.9550			
	Cr^{54}	2.38	53.9540			
	Cr^{55}			1.3–2.3h	$\beta^-1.4$	Cr^{54} (n, γ) Cr^{54} (d, p) Mn^{55} (n, p)
25	Mn^{51}			46m	$\beta^+2.0$	Cr^{50} (d, n)
	Mn^{52}			6.5d	K, $\beta^+0.582$ $\gamma 0.73, 0.94, 1.46$	Cr^{52} $(d, 2n)$ Cr^{52} (p, n) Fe^{54} (d, α)
	Mn^{52}			21m	$\beta^+2.66$ $\gamma 1.46$ (IT, $e^-0.39$)	Cr^{52} $(d, 2n)$ Cr^{52} (p, n) Fe^{54} (d, α)

	Mn^{54}			310d	K, γ0.85 ($\beta^- \sim 1$)	Fe^{56} (d, α) Mn^{55} (n, $2n$)
	Mn^{55}	100	54.957			
	Mn^{56}			2.59h	β^-2.81, 1.04, 0.75 γ0.822, 1.77, 2.06	Mn^{55} (n, γ) Mn^{55} (d, p) Co^{59} (n, α)
26	Fe^{52}			7.8h	β^+0.55	Cr^{50} (α, $2n$)
	Fe^{53}			8.9m	β^+	Cr^{50} (α, n) Fe^{54} (n, $2n$)
	Fe^{54}	5.81	53.9575			
	Fe^{55}			4y	K, Mn-x (γ0.07, e^-)	Mn^{55} (d, $2n$) Mn^{55} (p, n) Fe^{54} (n, γ)
	Fe^{56}	91.64	55.9531			
	Fe^{57}	2.21	56.9546			
	Fe^{58}	0.34	57.9506			
	Fe^{59}			47d	β^-0.26, 0.46 γ1.10, 1.30	Co^{59} (n, p) Fe^{58} (n, γ) Fe^{58} (d, p)
27	Co^{55}			18.2h	β^+1.50 γ0.8, 1.2, 0.16, 0.22	Fe^{54} (d, n) Ni^{58} (p, α)

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	Co^{56}			72d	K, $\beta^+1.50$ $\gamma 0.845$, 1.26, 1.74, 2.55, 3.25, 2.01	Fe^{56} $(d, 2n)$
	Co^{57}			270d	K, $\beta^+0.26$ $\gamma 0.117$, 0.130, 0.202, 0.215, e^-	Fe^{56} (d, n)
	Co^{58}			72d	K, $\beta^+0.470$ $\gamma 0.805$	Ni^{58} (n, p) Mn^{55} (α, n) Fe^{57} (d, n)
	Co^{59}	100				
	Co^{60}			5.3y	$\beta^-0.308$ $\gamma 1.115$, 1.317	Co^{59} (n, γ) Co^{59} (d, p) Ni^{60} (n, p)
	$Co^{\underline{60}}$			10.7m	IT 0.056 $e^-0.048$ $\beta^-1.35$, $\gamma 1.5$	Co^{59} (n, γ) Co^{59} (d, p) Ni^{60} (n, p)
	Co^{61}			1.75h	$\beta^-1.1$	Ni^{61} (n, p)
	Co^{62}			13.9m	$\beta^-2.5$, $\gamma 1.3$	Ni^{62} (n, p)
28	Ni^{57}			36h	$\beta^+0.67$	Fe^{54} (α, n) Ni^{58} $(n, 2n)$

	Ni^{58}	67.76	57.9594			
	Ni^{59}			Long	K	Ni^{58} (n, γ) Co^{59} $(d, 2n)$
	Ni^{60}	26.16	59.9495			
	Ni^{61}	1.25	60.9537			
	Ni^{62}	3.66	61.9493			
	Ni^{63}			Long	β^-0.05	Ni^{62} (n, γ) Cu^{63} (n, p)
	Ni^{64}	1.16	63.9471			
	Ni^{65}			2.6h	β^-1.9 γ1.1	Cu^{65} (n, p) Ni^{64} (n, γ) Ni^{64} (d, p)
	Ni^{66}			56h	β^-	As^{75} $(d, ?)$
29	Cu^{58}			10m (7.9m)	β^+	Ni^{58} $(d, 2n)$ Ni^{58} (p, n)
	Cu^{59}			81s	β^+	Ni^{58} (d, n)
	Cu^{60}			24.6m	β^+1.8, 3.3 γ1.5	Ni^{60} $(d, 2n)$ Ni^{60} (p, n)
	Cu^{61}			3.4h	β^+1.205, K No γ	Ni^{60} (d, n)

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	Cu^{62}			10.1m	β^+2.6 γ0.56	Ni^{62} $(d, 2n)$ Co^{59} (α, n) Cu^{63} $(n, 2n)$
	Cu^{63}	68.94	62.957			
	Cu^{64}			12.8h	K (γ1.35) β^-0.571 β^+0.657	Cu^{63} (d, p) Cu^{63} (n, γ) Zn^{64} (n, p) Zn^{66} (d, α)
	Cu^{65}	31.06	64.955			
	Cu^{66}			5m	β^-2.9 γ1.32	Cu^{65} (d, p) Cu^{65} (n, γ) Zn^{66} (n, p)
	$Cu^{67?}$			56h (61h)	β^-0.56	Zn^{67} (n, p)
30	Zn^{62}			9.5h	K	Cu^{63} $(d, 3n)$
	Zn^{63}			38m	β^+2.36, 1.46, 0.47, K γ0.96, 1.89, (2.60)	Cu^{63} $(d, 2n)$ Cu^{63} (p, n) Zn^{64} $(n, 2n)$
	Zn^{64}	48.89	63.9534			

	Zn^{65}			250d	K, γ1.11 β^+0.35	Cu^{65} $(d, 2n)$ Cu^{65} (p, n) Zn^{64} (n, γ) Zn^{64} (d, p)
	Zn^{66}	27.82	65.9465			
	Zn^{67}	4.15	66.9479			
	Zn^{68}	18.51	67.9485			
	Zn^{69}			57m	β^-1.0 No γ	Zn^{68} (d, p) Zn^{68} (n, γ) Ga^{71} (d, α)
	$Zn^{\underline{69}}$			13.8h	IT 0.44 γ0.44, e^- Zn-x	Zn^{68} (d, p) Zn^{68} (n, γ) Ga^{71} (d, α)
	Zn^{70}	0.62	69.946			
	Zn^{71}			2.2m	β^-2.1 γ	Zn^{70} (n, γ) Ga^{71} (n, p) Ge^{74} (n, α)
	Zn^{72}			49h	β^-0.3, 1.6 γ	U (n, f)
	Zn^{73}			<2m	β^-	U (n, f) Ge^{76} (n, α)
31	Ga^{64}			48m	β^+	Zn^{64} (p, n) Zn^{64} $(d, 2n)$

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	Ga^{65}			15m	K, e^-0.045, 0.108 (γ0.054, 0.117)	Zn^{64} (d, n)
	Ga^{66}			9.4h	β^+3.1	Cu^{63} (α, n) Zn^{66} $(d, 2n)$ Zn^{66} (p, n)
	Ga^{67}			79h	K, γ0.093, 0.297, 0.180 e^-	Zn^{66} (d, n)
	Ga^{68}			68m	β^+1.9	Zn^{68} $(d, 2n)$ Cu^{65} (α, n) Ge^{70} (d, α)
	Ga^{69}	60.16	68.952			
	Ga^{70}			20m	β^-1.65 γ	Ga^{69} (n, γ) Ga^{69} (d, p) Ge^{70} (n, p) Ge^{72} (d, α)
	Ga^{71}	39.84	70.952			
	Ga^{72}			14.1h	β^-0.56, 0.74, 1.00, 1.45, 1.74, 2.57, 3.17 γ0.84, 0.63, 2.2, 2.5, 1.05, 1.81, 1.57 e^-0.68	Ga^{71} (d, p) Ga^{71} (n, γ) Ge^{72} (n, p) Zn^{72}-β^-

	Ga^{73}		5h	$\beta^-1.4$ No γ	Ge^{73} (n, p) Zn^{73}-β^-
32	Ge^{66}		~140m		Ge^{70} $(d, p5n)$
	Ge^{67}		23m	β^+	Ge^{70} $(d, p4n)$
	Ge^{68}		250d	K	Zn^{66} $(\alpha, 2n)$
	Ge^{69}		39.6h	K, Ga $- x$, $\beta^+1.09$ $\gamma 1.22$	Ga^{69} $(d, 2n)$ Zn^{66} (α, n)
	Ge^{70}	20.55			
	Ge^{71}		11.4d	K, Ga $- x$ e^-, $\gamma 0.32$	Ge^{70} (n, γ) Ge^{70} (d, p) Ga^{71} $(d, 2n)$ Ga^{71} (p, n)
	Ge^{72}	27.37			
	Ge^{72}		0.5μs	IT, $e^-0.7$	Ga^{72}-β^-
	Ge^{73}	7.61			
	Ge^{74}	36.74			
	Ge^{75}		89m (82m)	$\beta^-1.2$	Ge^{74} (n, γ) Ge^{74} (d, p) As^{75} (n, p)
	Ge^{67}	7.67			

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	Ge^{77}			12h	$\beta^-2.0$ γ	Ge^{76} (n, γ) Ge^{76} (d, p) Se^{80} (n, α) U (n, f)
	Ge^{77}			59s	$\beta^-2.8$	Ge^{76} (n, γ)
	$Ge^{78?}$			2.1h	$\beta^-0.9$ γ	U (n, f)
33	As^{69}			52m	β^+	
	As^{71}			2.08d	K, $\beta^+0.6$?	Ge^{70} (d, n)
	As^{72}			26h	K, x, $\beta^+2.8$ $\gamma 0.60$, 1.40	Ga^{69} (α, n) Ge^{72} (p, n) Ge^{72} $(d, 2n)$
	As^{73}			76d	K, $\epsilon^-0.04$, Ge $- x$ $(\gamma 0.05)$	Ge^{72} (d, n)
	As^{74}			17.5d	$\beta^-1.3$ $\beta^+0.9$ $\gamma 0.58$	Ga^{71} (α, n) Ge^{74} $(d, 2n)$ Ge^{74} (p, n) As^{75} $(n, 2n)$
	As^{75}	100				

	As^{76}		26.8h	β^-3.04, 2.49, 1.29 γ0.55, 1.20, 1.75	As^{75} (n, γ) As^{75} (d, p) Br^{79} (n, α)
	As^{77}		40h	β^-0.7	Ge^{76} (d, n) Ge^{77}-β^-
	As^{78}		90m (65m)	β^-1.4, 4.1 γ0.27	Br^{81} (n, α) Se^{80} (d, α) Ge^{78}-β^-
	As^{81}		<10m	β^-	U (n, f)
34	Se^{69}		44m	β^+	As^{75} $(d, ?)$ Ge^{70} $(\alpha, 5n)$
	Se^{72}		9.5d	K	As^{75} $(d, 5n)$ Ge^{70} $(\alpha, 2n)$
	Se^{73}		7.0h	β^+1.29 K, x	Ge^{70} (α, n)
	Se^{74}	0.87			
	Se^{75}		127d	K, As-x γ0.10, 0.12, 0.14, 0.26, 0.40 e^-	As^{75} $(d, 2n)$ As^{75} (p, n) Se^{74} (n, γ)
	Se^{76}	9.02			
	Se^{77}	7.58			

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	Se^{77}			17.5s	IT, Se-x γ0.15, e^-0.135	Se^{76} (n, γ) Se^{76} (d, p)
	Se^{78}	23.52				
	Se^{79}			<10m or $>7 \times 10^6$y	β^-	Se^{78} (n, γ) U (n, f)
	Se^{80}	49.82				
	Se^{81}			18m	β^-1.5 No γ	Se^{80} (n, γ) Se^{80} (d, p) Br^{81} (n, p) U (n, f)
	Se^{81}			58m	IT 0.099 e^-0.087, 0.096 γ0.099	Se^{80} (n, γ) Se^{80} (d, p) Br^{81} (n, p) U (n, f)
	Se^{82}	9.19				
	Se^{83}			25m	β^-1.5 γ0.17, 0.37, 1.1	Se^{82} (n, γ) Se^{82} (d, p) U (n, f)
	Se^{83}			67s	β^-3.4 γ	Se^{82} (n, γ) Se^{82} (d, p)
	Se^{84}			~2m	β^-	U (n, f)

35	Br^{75}			1.7h	K, $\beta^+1.6$ x	Se^{74} (d, n) Se^{74} (p, γ)
	Br^{77}			2.4d	K, x β^+ 0.36, γ	Se^{76} (d, n) As^{75} $(\alpha, 2n)$
	Br^{78}			6.4m	$\beta^+2.3$ $\gamma 0.046$, 0.108 $e^-0.033$, 0.044, 0.095, 0.105	Se^{78} $(d, 2n)$ Se^{78} (p, n) As^{75} (α, n) Br^{79} $(n, 2n)$
	Br^{79}	50.51	78.9440			
	Br^{80}			18m	$\beta^-2.0$ $\beta^+0.8$ $\gamma < 0.5$	Br^{79} (n, γ) Br^{79} (d, p) Br^{80} $(4.4h)$-IT
	Br^{80}			4.4h	IT 0.086 $e^-0.047$, 0.035, 0.024 $\gamma 0.037$, (0.049)	Br^{79} (d, p) Br^{79} (n, γ) Se^{80} $(d, 2n)$ Se^{80} (p, n)
	Br^{81}	49.49	80.9419			
	Br^{82}			34h	$\beta^-0.465$ $\gamma 0.547$, 0.787, 1.35, 2.9	Br^{81} (n, γ) Br^{81} (d, p) Rb^{85} (n, α) Se^{82} $(d, 2n)$
	Br^{83}			2.4h	$\beta^-1.0$ No γ	Se^{83}-β^- Se^{82} (d, n)

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	Br^{84}			30m	β^-5.3 γ	U (n, f) Rb^{87} (n, α)
	Br^{85}			3.00m	β^-2.5	U (n, f)
	Br^{87}			56s	β^- n 0.3	U (n, f)
	$Br^{>86}$			4.51s	β^-, n 0.43	U (n, f)
	Br^{88}			16s	β^-	U (n, f)
36	Kr^{77}			65m	K, β^+1.7 γ, x	Se^{74} (α, n)
	Kr^{78}	0.34				
	Kr^{79}			34h	K, β^+0.6, 0.9 x, γ0.2	Se^{76} (α, n) Br^{79} $(d, 2n)$ Br^{79} (p, n)
	Kr^{80}	2.23				
	$Kr^{79,81}$			13s	No β^+ e^-0.173, 0.185 γ0.187	Br (p, n)
	$Kr^{79,81}$			55s	No β^+ e^-0.113, 0.125 γ0.127	Br (p, n)

Kr^{82}	11.50			
Kr^{83}	11.48			
$Kr^{\underline{83}}$		113m	IT, Kr-x e^-0.032, 0.045, 0.028	Br^{83}-β^- Kr^{82} (n, γ) Kr^{82} (d, p) Se^{80} (α, n)
Kr^{84}	57.02			
Kr^{85}		~10y	β^-0.74 No γ	U (n, f) Kr^{84} (n, γ)
$Kr^{\underline{85}}$		4.5h	β^-0.94 γ0.17, 0.37	Br^{85}-β^- Kr^{84} (n, γ) Kr^{84} (d, p) Sr^{88} (n, α)
Kr^{86}	17.43			
Kr^{87}		74m	β^-4	U (n, f) Kr^{86} (n, γ) Kr^{86} (d, p) Rb^{87} (n, p)
Kr^{88}		3h	β^-2.5	U (n, f)
Kr^{89}		2.6m	β^-	U (n, f)
Kr^{90}		~33s	β^-	U (n, f)
Kr^{91}		9.8s 5.7s	β^-	U (n, f)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Kr^{92}			3s	β^-	U (n, f)
	Kr^{93}			2.0s	β^-	U (n, f)
	Kr^{94}			1.4s	β^-	U (n, f)
	Kr^{97}			Short	β^-	U (n, f)
37	Rb^{81}			5.0h	K, x, $\beta^+0.9$, $\gamma 0.8$	Br^{79} $(\alpha, 2n)$
	Rb^{82}			20m		Br^{79} (α, n)
	Rb^{82}			6.5h	K, x, $\beta^+0.9$, $\gamma 1.0$	Br^{79} (α, n)
	Rb^{84}			$\sim$40d	β^+	Rb^{85} $(n, 2n)$ Sr^{86} (d, α)
	Rb^{85}	72.8				
	Rb^{86}			19.5d	$\beta^-1.82$, 0.72 $\gamma 1.08$	Rb^{85} (n, γ) Rb^{85} (d, p) Sr^{88} (d, α) Sr^{86} (n, p)
	Rb^{87}	27.2		6.0×10^{10}y	$\beta^-0.13$, $e^-0.56$? $\gamma 0.034$, 0.053, 0.082, 0.102, 0.129	Natural

	Rb^{88}		17.8m	β^-4.6	Rb^{87} (n, γ) Rb^{87} (d, p) Kr^{88}-β^- Sr^{88} (n, p)
	Rb^{89}		15.4m	β^-3.8	Kr^{89}-β^-
	Rb^{90}		Short	β^-	Kr^{90}-β^-
	Rb^{91}		Short	β^-	Kr^{91}-β^-
	Rb^{92}		Short	β^-	Kr^{92}-β^-
	Rb^{93}		Short	β^-	Kr^{93}-β^-
	Rb^{94}		Short	β^-	Kr^{94}-β^-
	Rb^{97}		Short	β^-	Kr^{97}-β^-
	Rb		80s	β^-	U (n, f)
38	Sr^{84}	0.55			
	Sr^{85}		65d	K γ0.8	Rb^{85} (p, n) Rb^{85} $(d, 2n)$
	Sr^{85}		70m	IT 0.17 γ0.17, e^-0.16	Rb^{85} (p, n) Rb^{85} $(d, 2n)$
	Sr^{86}	9.75			
	Sr^{87}	6.96			

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	Sr^{87}			2.7h	IT 0.386 γ0.386 e^-0.370, 0.383	Sr^{87} (n, n) Sr^{86} (n, γ) Rb^{87} $(d, 2n)$ Rb^{87} (p, n)
	Sr^{88}	82.74				
	Sr^{89}			54d	β^-1.50 No γ	Sr^{88} (d, p) Sr^{88} (n, γ) Rb^{89}-β^-
	Sr^{90}			25y	β^-0.61 No γ	Rb^{90}-β^-
	Sr^{91}			9.7h	β^-3.2, 1.3 $\gamma\sim$1.3	Rb^{91}-β^- Zr^{94} (n, α)
	Sr^{92}			2.7h	β^-	Rb^{92}-β^-
	Sr^{93}			7m	β^-	Rb^{93}-β^-
	Sr^{94}			$\sim$2m	β^-	Rb^{94}-β^-
	Sr^{97}			Short	β^-	Rb^{97}-β^-
39	Y^{87}			80h	K	Sr^{86} (d, n) Sr^{87} $(d, 2n)$ Sr^{87} (p, n)

Y^{87}		14h	IT 0.50 γ0.50 (e^-0.49)	Sr^{86} (d, n) Sr^{87} (d, $2n$) Sr^{87} (p, n)
Y^{88}		2.0h	β^+1.65	Sr^{88} (d, $2n$) Sr^{88} (p, n) Y^{89} (n, $2n$)
Y^{88}		104d	K (β^+0.83) γ0.91, 1.85, 2.76	Sr^{88} (d, $2n$) Sr^{88} (p, n) Y^{89} (n, $2n$)
Y^{89}	100			
Y^{90}		65h (60–72h)	β^-2.35 No γ	Sr^{90}-β^- Y^{89} (n, γ) Y^{89} (d, p) Cb^{93} (n, α)
Y^{91}		57d	β^-1.53 No γ	Sr^{91}-β^- Zr^{91} (n, p)
Y^{91}		51m	IT 0.61 γ0.61, e^-0.5	Sr^{91}-β^- Zr^{91} (n, p)
Y^{92}		3.5h	β^-3.4 γ0.6	Sr^{92}-β^- Zr^{92} (n, p) Zr^{94} (d, α)
Y^{93}		10h	β^-3.1 γ0.7	Sr^{93}-β^-

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Y^{94}			16.5m	β^-, γ	Sr^{94}-β^- Zr^{94} (n, p)
	Y^{95}			<3h	β^-	U (n, f)
	Y^{97}			Short	β^-	Sr^{97}-β^-
40	Zr^{89}			78h	β^+1.0 No γ	Y^{89} (p, n) Y^{89} $(d, 2n)$ Mo^{92} (n, α)
	Zr^{89}			4.5m	IT or K γ, e^-0.555	Y^{89} (p, n) Y^{89} $(d, 2n)$
	Zr^{90}	51.46				
	Zr^{91}	11.23				
	Zr^{92}	17.11				
	Zr^{93}?			2.5m	β^-?	Cb (n, p)?
	Zr^{94}	17.40				
	Zr^{95}			65d	β^-0.394, 1.0 γ0.73, 0.92?	Y^{95}-β^- Zr^{94} (n, γ) Zr^{94} (d, p)
	Zr^{96}	2.80				

	Zr^{97}		17.0h	$\beta^-2.1$ $\gamma\sim0.8$	Y^{97}-β^- Zr^{96} (n, γ) Zr^{96} (d, p)
	Zr		6m	$\beta^-\sim1.9$	Zr (n, γ)
	Zr?		18m	β^-	Zr $(n, ?)$
	Zr?		70h	$\beta^-1.17$	Zr $(n, ?)$
41	Cb^{90}		18h (21h)	$\beta^+\sim1$	Zr^{90} (p, n) Zr^{90} $(d, 2n)$
	Cb^{91}		$\sim$55d	K, $e^-0.011$ (γ0.94)	Zr^{90} (d, n)
	Cb^{92}		21.6h	$\beta^-1.2$ $\gamma0.6$	Zr^{92} (p, n) Mo^{92} (n, p) Y^{89} (α, n)
	Cb^{92}		10.1d	$\beta^-1.38$ $\gamma1.0$	Zr^{92} (p, n) Mo^{92} (n, p) Y^{89} (α, n)
	Cb^{93}	100			
	$Cb^{\underline{93}}$		42d	IT	Cb^{93} (n, n)
	Cb^{94}		>100y		
	$Cb^{\underline{94}}$		6.6m	IT 0.05 Cb-x ($\beta^-1.35$)	Cb^{93} (n, γ) Cb^{93} (d, p)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Cb^{95}			35d	$\beta^-0.15$ $\gamma 0.75, 0.92?$ $(e^-0.735)$	Zr^{95}-β^- Mo^{97} (d, α)
	Cb^{95}			90h	IT 0.24 $e^-0.22, 0.24$ Cb-x	Zr^{95}-β^-
	Cb^{96}			3–4d		Zr^{96} (p, n) Zr^{96} $(d, 2n)$
	Cb^{97}			75m	$\beta^-1.4$ $\gamma 0.78$	Zr^{97}-β^- Mo^{97} (n, p)
	Cb?			38m		Zr (p, n)?
	Cb?			12m		Zr (p, n)?
	Cb?			4m		Zr (p, n)?
42	Mo^{92}	15.84				
	Mo^{93}			6.7h	$\beta^+0.3, 0.7$ $\gamma 1.6$	Cb^{93} $(d, 2n)$ Cb^{93} (p, n)
	$Mo^{93?}$			17m	$\beta^+2.65$	Mo^{94} $(n, 2n)$
	Mo^{94}	9.04	93.945			
	Mo^{95}	15.72	94.946			

	Mo^{96}	16.53	95.944			
	Mo^{97}	9.46	96.945			
	Mo^{98}	23.78	97.943			
	Mo^{99}			67h	β^-1.05, 0.21 γ0.75, 0.24	U (n, f) Mo^{98} (n, γ) Mo^{98} (d, p)
	Mo^{100}	9.63				
	Mo^{101}			14.6m	β^-1.0, 2.2 γ0.3, 0.9	U (n, f) Mo^{100} (n, γ) Mo^{100} (d, p)
	$Mo^{102?}$			12m	β^-	U (n, f)
	Mo^{105}			Short	β^-	U (n, f)
43	Tc^{92}			47m	K, Mo $- x$, γ1.5, e^-0.54	Mo^{92} $(d, 2n)$
	Tc^{93}			4.5m	β^+4.3 γ1.3	Mo^{92} (d, n)
	Tc^{93}			2.7h	K, β^+0.83 γ2.00	Mo^{92} (d, n)
	Tc^{94}			<53m	K, β^+2.45 γ0.38, 0.87, 1.48, 1.85, 2.74	Tc^{94}-IT

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Tc^{94}			53m	IT 0.033 e^-0.013, 0.031, 0.033 Tc-x	Mo^{94} (d, $2n$) Mo^{94} (p, n)
	Tc^{95}			20h	K, γ0.762, 0.932, 1.071 e^-0.742, 0.912, 1.051 x	Ru^{95}-β^+ Mo^{92} (α, p) Mo^{95} (p, n) Mo^{95} (d, $2n$)
	Tc^{95}			52d (62d)	K, γ0.201, 0.57, 0.81, 1.01 (β^+0.4) Mo-x, e^-0.18	Mo (p, ?) Mo (d, ?)
	Tc^{96}			4.2d	K, Mo-x (β^-0.8?) γ0.771, 0.842, 0.312, 0.806, 1.119 e^-0.751, 0.822, 0.786, 0.292, 1.099	Mo^{96} (p, n) Cb^{93} (α, n) Mo^{96} (d, $2n$)
	Tc^{97}			93d	IT ?0.108 e^-0.087, γ? Tc-x	Ru^{97}-K Mo^{96} (d, n)
	$Tc^{97,98?}$			40m	β2.0	$Mo^{97,98}$ (d, ?)
	Tc^{98}			2.8d	β^-1.3 γ0.9	Mo^{98} (d, $2n$)

	Tc^{99}			1×10^6y	β^-0.3	Mo^{99}-β^-
	Tc^{99}			5.9h	IT 0.136, x e^-0.116 γ0.136	Mo^{99}-β^-
	$Tc^{100,101}$			80s	β2.3 γ0.6	Mo^{100} (d, ?)
	Tc^{101}			14.0m	β^-1.3 γ0.30	Mo^{100} (d, n) Mo^{101}-β^-
	$Tc^{102?}$			<1m	β^-	$Mo^{102?}$ (12m)-β^-
	Tc^{105}			Short	β^-	Mo^{105}-β^-
	$Tc^{107?}$			<1.5m	β^-	U (n, f)
	Tc?			36.5h	β^-	Mo (p, ?)
	Tc?			18s	β^-	Mo (p, ?)
	Tc?			~2d	K	Mo (d, ?)
44	Ru^{95}			1.65h	β^+1.1 γ0.95 x	Mo^{92} (α, n) Ru^{96} (n, $2n$)
	Ru^{96}	5.68	95.945			
	Ru^{97}			2.8d	K, x 0.018 e^-0.2, γ0.23	Mo^{94} (α, n) Ru^{96} (n, γ) Ru^{96} (d, p)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Ru^{98}	2.22	97.943			
	Ru^{99}	12.81	98.944			
	Ru^{100}	12.70	99.942			
	Ru^{101}	16.98	100.946			
	Ru^{102}	31.34	101.941			
	Ru^{103}			42d	β^-0.665, 0.350 γ0.55 e^-0.217, 0.290	U (n, f) Ru^{102} (n, γ) Ru^{102} (d, p) Rh^{103} (n, p)
	Ru^{104}	18.27				
	Ru^{105}			4.5h	β^-1.35 γ0.76	Tc^{105}-β^- Ru^{104} (n, γ) Ru^{104} (d, p)
	Ru^{106}			1.0y	$\beta^-\sim$0.03 No γ	U (n, f)
	$Ru^{107?}$			4m	$\beta^-\sim$4	$Tc^{107?}$ (<1.5m)-β^-
45	Rh^{100}			19.4h	K, x 0.0193 e^-0.6, γ1.2 β^+3.0	Pd^{100}-K Ru^{99} (d, n) Ru^{100} $(d, 2n)$ Ru^{100} (p, n)

Rh^{101}			4.3d	K, x $\gamma 0.35$, e^-	Pd^{101}-K, β^+ Ru^{100} (d, n) Ru^{101} $(d, 2n)$ Ru^{101} (p, n)
Rh^{102}			210d	$\beta^- 1.04$ $\beta^+ 1.13$ γ	Ru^{102} $(d, 2n)$ Ru^{102} (p, n) Rh^{103} $(n, 2n)$ Pd^{104} (d, α)
Rh^{103}	100	102.941			
$Rh^{\underline{103}}$			54m	IT $e^- 0.034$ Rh-x	Rh^{103} (n, n) Ru^{103}-β^-
Rh^{104}			42s	$\beta^- 2.6$ $\gamma 0.4$	$Rh^{\underline{104}}$-IT Rh^{103} (n, γ) Rh^{103} (d, p) Ru^{104} (p, n)
$Rh^{\underline{104}}$			4.3m	IT $e^- 0.069$ $\gamma 0.08$	Rh^{103} (n, γ) Rh^{103} (d, p) Ru^{104} (p, n)
Rh^{105}			36.5h	$\beta^- 0.66$ $(e^- 0.3, \gamma 0.33)$	Ru^{105}-β^- Ru^{104} (d, n) Pd^{105} (n, p)
Rh^{106}			30s	$\beta^- 3.55, 2.30$ $\gamma 0.51, 0.73, 1.25$	Ru^{106}-β^- Pd^{108} (d, α)

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	$Rh^{107?}$			24m	β^-1.2	$Ru^{107?}$ (4m)-β^-
	Rh^{109}			<1h	β^-	U (n, f)
	Rh?			9h	β^-1.3 γ0.8	U (n, f)
46	Pd^{100}			4.0d	K, x γ1.8, 0.09	Rh^{103} (d, $5n$)
	Pd^{101}			9h	K, x β^+2.3	Rh^{103} (d, $4n$) Ru^{98} (α, n)
	Pd^{102}	0.8	101.941			
	Pd^{103}			17d	K, x	Rh^{103} (d, $2n$) Rh^{103} (p, n)
	Pd^{104}	9.3	103.941			
	Pd^{105}	22.6	104.942			
	Pd^{106}	27.2	105.941			
	Pd^{107}			Very short or $>10^8$y	β^-	Pd^{106} (n, γ)
	Pd^{108}	26.8	107.941			

	Pd^{109}			13.4h	$\beta^-1.1$ No γ	Pd^{108} (n, γ) Pd^{108} (d, p) U (n, f) Ag^{109} (n, p)
	Pd^{110}	13.5	109.941			
	Pd^{111}			26m	$\beta^-3.5$	Pd^{110} (n, γ) Pd^{110} (d, p) U (n, f)
	Pd^{112}			21h	$\beta^-0.2$ No γ	U (n, f)
47	$Ag^{102,104?}$			16.3m		Pd $(p, ?)$
	$Ag^{102,104?}$			73m		Pd $(p, ?)$
	Ag^{105}			45d	K γ0.282, 0.345, 0.430, 0.650, >1.0	Pd^{105} (p, n) Rh^{103} $(\alpha, 2n)$ Pd^{105} $(d, 2n)$
	Ag^{106}			8.2d	K, γ1.63, 1.06, 0.72? $e^-1.2$	Pd^{106} $(d, 2n)$ Pd^{106} (p, n) Rh^{103} (α, n) Ag^{107} $(n, 2n)$
	$Ag^{\underline{106}}$			24.5m	$\beta^+2.04$ No γ	Pd^{106} $(d, 2n)$ Pd^{106} (p, n) Rh^{103} (α, n) Ag^{107} $(n, 2n)$
	Ag^{107}	51.35	106.945			

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Ag^{107}			44.3s	IT 0.0935 e^- γ0.0935	Cd^{107}-β^+, K
	Ag^{108}			2.3m	β^-2.8	Ag^{107} (n, γ) Ag^{107} (d, p) Cd^{108} (n, p)
	Ag^{109}	48.65	108.944			
	Ag^{109}			40.0s	IT 0.0884 e^- γ0.0884	Cd^{109}-K Pd^{109}-β^-
	Ag^{110}			225d	β^-<0.1, 0.59 γ0.66, 0.90, 1.40 e^-, K?, x	Ag^{109} (n, γ) Ag^{109} (d, p)
	Ag^{110}			24s	β^-2.8 γ	Ag^{109} (n, γ) Ag^{109} (d, p)
	Ag^{111}			7.6d	β^-1.06	Pd^{110} (d, n) Pd^{111}-β^- Cd^{111} (n, p)
	Ag^{112}			3.2h	β^-3.6 γ0.86	Pd^{112}-β^- Cd^{112} (n, p) In^{115} (n, α)

	Ag^{113}		5.3h	$\beta^-2.2$	U (n, f)
	Ag^{115}		22m	$\beta^-3.0$ γ	U (n, f) Cd^{116} (γ, p)
48	Cd^{106}	**1.215**			
	Cd^{107}		6.7h	K $(\beta^+0.32)$ $(\gamma 0.846)$	Ag^{107} $(d, 2n)$ Ag^{107} (p, n) Cd^{106} (n, γ)
	Cd^{108}	0.875			
	Cd^{109}		330d (158d)	K	Ag^{109} $(d, 2n)$ Ag^{109} (p, n) Cd^{108} (n, γ)
	Cd^{110}	12.39			
	Cd^{111}	12.75			
	$Cd^{\underline{111}}$		48m	IT $\gamma 0.247$, (0.149) $e^-0.118$, 0.220	Cd^{111} (n, n) Cd^{110} (n, γ) Pd^{108} (α, n)
	Cd^{112}	24.07			
	Cd^{113}	**12.26**			
	$Cd^{\underline{113}}$		2.3m	IT?	Cd^{112} (n, γ)
	Cd^{114}	28.86			

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Cd^{115}			2.33d	β^-1.13, 0.6 γ0.52	Cd^{114} (n, γ) Cd^{114} (d, p) U (n, f)
	Cd^{115}			44d	β^-1.7 γ0.5?	Cd^{114} (n, γ) Cd^{114} (d, p) U (n, f)
	Cd^{116}	7.58				
	Cd^{117}			2.8h (3.75h)	β^-1.3–1.7	U (n, f) Cd^{116} (d, p) Cd^{116} (n, γ)
49	In^{108}?			5h	No β^+ γ0.65	
	In^{109}			5.2h 6.5h	K, β^+2.0 γ0.5	Ag^{107} $(\alpha, 2n)$ Cd^{108} (d, n)
	In^{110}			66m	β^+1.6	Ag^{107} (α, n) Cd^{110} (p, n) Cd^{110} $(d, 2n)$
	In^{111}			2.84d	K γ0.173, 0.247 e^-0.146, 0.220, 0.243, 0.169, 0.246, 0.172	Cd^{110} (d, n) Ag^{109} $(\alpha, 2n)$

In^{112}		9m	β^-1, $\beta^+1.7$ K, x	Cd^{112} $(d, 2n)$ Cd^{112} (p, n) In^{112} (23m)-IT
In^{112}		23m	IT 0.16 e^-, $\gamma 0.16$ x	Ag^{109} (α, n) Cd^{112} $(d, 2n)$ Cd^{112} (p, n)
In^{113}	4.23			
In^{113}		105m	IT 0.393 $e^-0.365$, 0.389 $\gamma 0.393$	Sn^{113}-K In^{113} (n, n) Cd^{112} (d, n)
In^{114}		72s	$\beta^-1.98$	In^{114} (48d)-IT In^{113} (n, γ)
In^{114}		48d	IT 0.192 $e^-0.162$, 0.187	In^{113} (n, γ) Cd^{114} $(d, 2n)$ Cd^{114} (p, n)
In^{115}	95.77			
In^{115}		4.53h	IT 0.338 $e^-0.308$, 0.332 $\gamma 0.338$	In^{115} (n, n) Cd^{114} (d, n) Cd^{115}-β^-
In^{116}		54.0m	$\beta^-0.85$ $\gamma 2.32$, 1.31, 1.12, 0.428	In^{115} (n, γ) In^{115} (d, p) Cd^{116} $(d, 2n)$ Cd^{116} (p, n)
In^{116}		13s	$\beta^-2.8$ No γ	In^{115} (n, γ) In^{115} (d, p)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	In^{117}			117m	β^-1.73 No γ	Cd^{117}-β^- Cd^{116} (d, n)
50	Sn			4.5h	K	Sb $(d, ?)$
	$Sn^{\leq 119}$?			25m	β^-	Cd (α, n)?
	$Sn^{\leq 119}$?			3h	β^-	Cd (α, n)?
	$Sn^{\leq 119}$?			13d	β^-	Cd (α, n)?
	Sn^{112}	0.90				
	Sn^{113}			105d	K, In-x e^-0.055, 0.080 γ0.085	In^{113} (p, n) In^{113} $(d, 2n)$
	Sn^{114}	0.61				
	Sn^{115}	0.35	114.940			
	Sn^{116}	14.07	115.939			
	Sn^{117}	7.54	116.937			
	Sn^{118}	23.98	117.937			
	Sn^{119}	8.62	118.938			
	$Sn^{\underline{119}}$			13–14d	IT 0.17 e^-0.13, γ	Cd^{116} (α, n) Sb^{121} (d, α)

Sn^{120}	33.03	119.937			
Sn^{121}			28h	$\beta^-0.4$ No γ	Sn^{120} (n, γ) Sn^{120} (d, p) Sb^{121} (n, p)
$Sn^{\leq 125}$			47h	$\beta^-0.8$	Sn (n, γ)
$Sn^{\leq 125}$			40m		Sn (d, p) Sn (n, γ)
$Sn^{\leq 125}$			~400d		Sn (d, p) Sn (n, γ)
Sn^{122}	4.78	121.945			
$Sn^{123,121}$			130d	$\beta^-1.5$ No γ	U (n, f)
$Sn^{123?}$			10d	$\beta^-2.6$ γ, x	U (n, f) Sn (n, γ) Sn (d, p)
Sn^{124}	6.11	123.944			
Sn^{125}			9m	β^-	Sn (d, p) Sn (n, γ)
$Sn^{126?}$			70m	$\beta^-0.7$ or 2.7 γ?	U (n, f)
$Sn^{?}$			~20m	β^-	U (n, f)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
51	Sb^{116}			60m	β^+1.45 γ0.70	In^{115} (α, $3n$)
	Sb^{117}			2.8h	K, x e^-0.46, 0.126	Sn^{116} (d, n) Sn^{117} (p, n)
	Sb^{118}			5.1h	K, x e^-, γ1.5, 0.26	In^{113} (α, n) Sn^{116} (d, $2n$) Sn^{116} (p, n)
	Sb^{118}			3.5m	β^+3.1	Te^{118}-K or β^+ In^{115} (α, n)
	Sb^{119}			39h	K, x	Te^{119}-K Sn^{118} (d, n)
	Sb^{120}			17m	β^+1.53	Sb^{121} (n, $2n$) Sn^{120} (d, $2n$) Sn^{120} (p, n)
	Sb^{120}			6.0d	K, γ1.1 Sn-x	Sb^{121} (d, H^3) Sn^{120} (d, $2n$) Sn^{120} (p, n)
	Sb^{121}	57.25				
	Sb^{122}			2.8d	β^-1.36, 1.94 γ0.57, e^-	Sb^{121} (n, γ) Sb^{121} (d, p)

Sb^{122}		3.5m	IT 0.140 e^-0.110 Sb-x	Sb^{121} (n, γ) Sb^{121} (d, p)
Sb^{123}	42.75			
Sb^{124}		60d	β^-0.68, 2.37, 0.48, 1.0, 1.6 γ0.603, 1.70, 0.71, 0.65, 2.06 e^-0.57, 0.09	Sb^{123} (n, γ) Sb^{123} (d, p) I^{127} (n, α)
Sb^{124}		1.3m	β^-3.2 γ	Sb^{123} (n, γ) Sb^{123} (d, p)
Sb^{124}		21m	IT ~0.020 e^-0.014 β^-	Sb^{123} (n, γ) Sb^{123} (d, p)
Sb^{125}		2.7y	β^-0.621, 288 γ0.174, 0.431, 0.125, 0.461, 0.609, 0.646, e^-, x	Sn^{125}-β^- U (n, f)
$Sb^{126?}$		~60m	β^-2.7 or 0.7 γ?	$Sn^{126(?)}$ (70m)-β^-
Sb^{127}		93h	β^-1.15 γ0.72	U (n, f)
Sb^{129}		4.2h	β^-	U (n, f)
Sb^{132}		~5m	β^-	U (n, f)
Sb^{133}		<10m	β^-	U (n, f)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Sb^{134}			<10m	β^-	U (n, f)
52	$Te^{<125?}$			2.5h	e^-	Sb $(d, ?)$
	Te^{118}			6.0d	K	Sb^{121} $(d, 5n)$
	Te^{119}			4.5d	K, e^-0.2, 0.5 γ1.5	Sb^{121} $(d, 4n)$
	Te^{120}	0.091				
	Te^{121}			17d	K, Sb-x γ0.61	Sb^{121} $(d, 2n)$ Sb^{121} (p, n)
	$Te^{\underline{\underline{121}}}$			143d	IT, e^-0.022 – 0.212 Te-x	Sb^{121} $(d, 2n)$ Sb^{121} (p, n)
	$Te^{\underline{121}}$			0.05μs	IT 0.225 γ0.225	$Sb^{\underline{\underline{121}}}$ (143d)-IT
	Te^{122}	2.49				
	Te^{123}	0.89				
	Te^{124}	4.63				
	Te^{125}	7.01				
	$Te^{\underline{125}}$			58d	IT 0.109, Te-x e^-0.077, 0.105, 0.109, 0.030	Sb^{125}-β^-
	Te^{126}	18.72				

Te^{127}		9.3h	β^-0.70 No γ	Te^{127} (90d)-IT Te^{126} (d, p) Te^{126} (n, γ)
$Te^{\underline{127}}$		90d	IT 0.086 e^-0.055, 0.082, 0.085 x 0.028	Te^{126} (d, p) Te^{126} (n, γ) Sb^{127}-β^-
Te^{128}	31.72			
Te^{129}		70m	β^-1.8 γ0.3, 0.8 x 0.030	Te^{128} (d, p) Te^{128} (n, γ) $Te^{\underline{129}}$ (32d)-IT
$Te^{\underline{129}}$		32d	IT 0.102 e^-0.070, 0.10 x	Te^{128} (n, γ) Te^{128} (d, p) Sb^{129}-β^-
Te^{130}	34.46			
Te^{131}		25m	β^-	$Te^{\underline{131}}$ (30h)-IT Te^{130} (d, p) Te^{130} (n, γ)
$Te^{\underline{131}}$		30h	IT 0.177 e^-0.147, 0.175	Te^{130} (d, p) Te^{130} (n, γ) U (n, f)
Te^{132}		77h	β^-0.28 γ0.22, e^- x	Sb^{132}-β^-
Te^{133}		60m	β^-	Sb^{133}-β^-

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Te^{134}			43m	β^-	Sb^{134}-β^-
	Te^{135}			<2m	β^-	U (n, f)
	$Te^{?}$			~1m	β^-	U (n, f)
	Te?			200d	β1.6	Te $(d, ?)$
53	I^{124}			4.0d	β^+	Sb^{121} (α, n) Te^{124} (p, n) Te^{124} $(d, 2n)$
	I^{125}			56d	K, Te-x No β, no γ	Te^{124} (d, n) Te^{125} $(d, 2n)$ Te^{125} (p, n)
	I^{126}			13.0d	β^-1.1, K γ0.5	Sb^{123} (α, n) I^{127} $(n, 2n)$ Te^{126} $(d, 2n)$ Te^{126} (p, n)
	I^{127}	100				
	I^{128}			25.0m	β^-2.02, 1.59 γ0.428	I^{127} (n, γ) I^{127} (d, p) Te^{128} $(d, 2n)$ Te^{128} (p, n)
	I^{129}			Long	β^-	Te^{129} (70m)-β^-

	I^{130}		12.6h	β^-1.03, 0.61 γ0.537, 0.667, 0.744, 0.417 (e^-)	Te^{130} (d, $2n$) Te^{130} (p, n) Cs^{133} (n, α)
	I^{131}		8.0d	β^-0.595, 0.315 γ0.363, 0.638, 0.283, 0.080, e^-	Te^{130} (d, n) Te^{131} (25m)-β^-
	I^{132}		2.4h	β^-1.0, 2.1 γ0.6, 1.4	Te^{132}-β^-
	I^{133}		22h	β^-1.3 γ0.55	Te^{133}-β^-
	I^{134}		54m	β^- $\gamma > 1$	Te^{134}-β^-
	I^{135}		6.7h	β^-1.00, 0.47, 1.40 γ1.27, 2.00	Te^{135}-β^-
	I^{136}		86s	β^-6.5 γ2.9	U (n, f)
	I^{137}		22.0s	β^- n 0.67	U (n, f)
	I^{138}		5.9s	β^-	U (n, f)
	I^{139}		2.6s	β^-	U (n, f)
54	Xe^{124}	0.095			
	Xe^{126}	0.088			

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Xe^{127}			34d	K, e^- $\gamma 0.9$	I^{127} (p, n)
	$Xe^{\underline{127}}$			75s	IT? Xe-x $e^- 0.0914, 0.140, 0.170$ $\gamma 0.125, 0.175$	I^{127} (p, n)
	Xe^{128}	1.917				
	Xe^{129}	26.24				
	Xe^{130}	4.053				
	Xe^{131}	21.24				
	Xe^{132}	26.93				
	Xe^{133}			5.3d	$\beta^- 0.35$ $\gamma 0.085$ $e^- 0.049$ x 0.031	I^{133}-β^- Cs^{133} (n, p) Xe^{132} (d, p) Xe^{132} (n, γ)
	Xe^{134}	10.52				
	Xe^{135}			9.2h	$\beta^- 0.94$ $\gamma 0.25$	I^{135}-β^- Xe^{134} (n, γ) Xe^{134} (d, p)
	$Xe^{\underline{135}}$			13m	IT 0.54 $e^- 0.50$ $\gamma 0.54$	I^{135}-β^- Xe^{134} (n, γ) Xe^{134} (d, p)

	Xe^{136}	8.93			
	Xe^{137}		3.9m	$\beta^- \sim 4$	I^{137}-β^- Xe^{136} (n, γ) Xe^{136} (d, p)
	Xe^{138}		17m	β^-	U (n, f)
	Xe^{139}		41s	β^-	U (n, f)
	Xe^{140}		16s	β^-	U (n, f)
	Xe^{141}		3s (1.7s)	β^-	U (n, f)
	Xe^{143}		1s	β^-	U (n, f)
	Xe^{144}		Short	β^-	U (n, f)
	$Xe^{145?}$		0.8s	β^-	U (n, f)
	$Xe^{?}$		68m	IT?	Xe $(d, ?)$ Xe $(n, ?)$
55	$Cs^{130?}$		30m		
	Cs^{131}		10.0d	K, Xe-x No β^+	Ba^{131}-K
	Cs^{132}		7.1d	K $\epsilon^-0.6$ $\gamma 0.68$	Cs^{133} $(n, 2n)$ Xe^{132} $(d, 2n)$ Xe^{132} (p, n)
	Cs^{133}	100			

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Cs^{134}			2.3y (1.7y)	β^-0.658, 0.09 γ0.602, 0.794, 0.568, 1.396 (e^-)	Cs^{133} (n, γ) Cs^{133} (d, p) Xe^{134} $(d, 2n)$ Xe^{134} (p, n)
	Cs^{134}			3.1h	IT 0.160, Cs-x e^- β^-2.4 γ0.7	Cs^{133} (n, γ) Cs^{133} (d, p) Xe^{134} $(d, 2n)$ Xe^{134} (p, n)
	Cs^{135}			2.1×10^6y	β^-0.21, No γ	Xe^{135}-β^-
	Cs^{136}			13.7d	β^-0.35 γ0.9	La^{139} (n, α)
	Cs^{137}			33y	β^-0.550	Xe^{137}-β^- Ba^{137} (n, p)
	Cs^{138}			32m	β^-2.6 γ1.2	Xe^{138}-β^- Ba^{138} (n, p)
	Cs^{139}			7m(10m)	β^-	Xe^{139}-β^-
	Cs^{140}			Short (40s)	β^-	Xe^{140}-β^-
	Cs^{141}			Short	β^-	Xe^{141}-β^-
	$Cs^{142?}$			1–2m	β^-	U (n, f)

	Cs^{143}		Short	β^-	Xe^{143}-β^-
	Cs^{144}		Short	β^-	Xe^{144}-β^-
	$Cs^{145?}$		Short	β^-	$Xe^{145?}$ (0.8s)-β^-
56	Ba^{130}	0.101			
	Ba^{131}		12.0d	*K*, Cs-*x* γ0.26, 0.5 (1.2) e^-0.42, 0.24	Ba^{130} (*n*, γ) Ba^{130} (*d*, *p*)
	Ba^{132}	0.097			
	Ba^{133}		>20y	*K*, Cs-*x* No β^+ γ0.32, 0.085 e^-0.047, 0.006, 0.29	Cs^{133} (*d*, 2*n*) Cs^{133} (*p*, *n*) Ba^{132} (*n*, γ)
	$Ba^{\underline{133}}$		38.8h	IT 0.276 e^-0.239, 0.270, 0.275 γ0.276	Cs^{133} (*d*, 2*n*) Cs^{133} (*p*, *n*)
	Ba^{134}	2.42			
	Ba^{135}	6.59			
	$Ba^{\underline{135}?}$		28.7h	IT, Ba-*x*, e^-0.280, γ	Ba (*d*, *p*)
	Ba^{136}	7.81			
	Ba^{137}	11.32			
	$Ba^{\underline{137}}$		2.6m	IT 0.663, Ba-*x*, γ0.663, e^-0.626, 0.657	Cs^{137}-β^-

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Ba^{138}	71.66				
	Ba^{139}			85m	β^-2.27 γ0.163, (1.05) e^-0.126, 0.159	Cs^{139}-β^- La^{139} (n, p) Ba^{138} (n, γ) Ba^{138} (d, p)
	Ba^{140}			12.8d	β^-1.05, 0.34? γ0.54 e^-0.50	Cs^{140}-β^-
	Ba^{141}			18m	β^-, γ	Cs^{141}-β^-
	$Ba^{142?}$			6m	β^-	$Cs^{142?}$ (1–2m)-β^-
	Ba^{143}			<0.5m	β^-	Cs^{143}-β^-
	Ba^{144}			Short	β^-	Cs^{144}-β^-
	$Ba^{145?}$			Short	β^-	$Cs^{145?}$ (short)-β^-
57	$La^{<139}$			10m	β^+2.1	Ba $(d, ?)$
	La^{135}			19.5h	K, γ0.76	Cs^{133} $(\alpha, 2n)$
	La^{136}			2.1h	β^+0.84 No γ	Cs^{133} (α, n)
	La^{137}			>400y		Ce^{137}-K

	La^{138}	0.089		Long?		Natural?
	La^{139}	99.91	138.953			
	La^{140}			40h	β^-1.4, 0.90, 2.12 γ1.64, 0.85, 0.49, 2.4, 0.335	Ba^{140}-β^- La^{139} (d, p) La^{139} (n, γ)
	La^{141}			3.7h	β^-2.8 γ?	Ba^{141}-β^-
	$La^{142?}$			74m	β^-, γ	$Ba^{142?}$ (6m)-β^-
	La^{143}			19m	β^-	Ba^{143}-β^-
	La^{144}			Short	β^-	Ba^{144}-β^-
	$La^{145?}$			Short	β^-	$Ba^{145?}$ (short)-β^-
58	Ce^{135}			16h	β^+	La^{139} (d, $6n$)
	Ce^{136}	0.193				
	Ce^{137}			36h	K, γ0.28, 0.75 e^-0.18	La^{139} (d, $4n$)
	Ce^{138}	0.250				
	Ce^{139}			140d	K, La-x γ0.184, 0.8 e^-0.135, 0.165	La^{139} (d, $2n$) La^{139} (p, n) Ce^{140} (n, $2n$)
	Ce^{140}	88.48				

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Ce^{141}			30d	$\beta^-0.55$ $\gamma 0.21$, (0.103) $e^-0.06$	La^{141}-β^- Ce^{140} (n, γ) Ce^{140} (d, p) Pr^{141} (n, p)
	Ce^{142}	11.07				
	Ce^{143}			33h	$\beta^-1.35$ $\gamma 0.5$	La^{143}-β^- Ce^{142} (n, γ) Ce^{142} (d, p)
	Ce^{144}			275d	$\beta^-0.348$ No γ $e^-0.075$, 0.12	La^{144}-β^-
	$Ce^{145?}$			1.8h	β^-	$La^{145?}$ (short)-β^-
	$Ce^{146?}$			14.6m	β^-	U (n, f)
59	Pr^{140}			3.5m	$\beta^+2.40$	Pr^{141} $(n, 2n)$ Ce^{140} $(d, 2n)$ Ce^{140} (p, n)
	Pr^{141}	100				
	Pr^{142}			19.3h	$\beta^-2.14$, 0.21 $\gamma 1.9$, 0.134, 0.329, 0.490, 0.624, e^-	Pr^{141} (n, γ) Pr^{141} (d, p) Ce^{142} $(d, 2n)$ Ce^{142} (p, n)

	Pr^{143}			13.8d	β^-0.83 No γ	Ce^{143}-β^- Ce^{142} (d, n)
	Pr^{144}			17.5m	β^-3.07 e^-0.091, 0.128, 0.103? γ0.135, 0.145?, 1.25	Ce^{144}-β^-
	$Pr^{145?}$			4.5h	β^-3.1 No γ	$Ce^{145?}$ (1.8h)-β^-
	$Pr^{146?}$			24.6m	$\beta^-\sim$3 γ1.4	$Ce^{146?}$ (14.6m)-β^-
60	$Nd^{141?}$			2.5h	β^+0.78	Pr^{141} (p, n) Pr^{141} (d, $2n$) Nd^{142} (n, $2n$)
	Nd^{142}	26.9				
	Nd^{143}	12.2				
	Nd^{144}	23.9				
	Nd^{145}	8.3	144.962			
	Nd^{146}	17.3	145.962			
	Nd^{147}			11.0d	β^-0.90, 0.4 γ0.58 e^-0.03 x 0.04	U (n, f) Nd^{146} (n, γ) Nd^{146} (d, p)
	Nd^{148}	5.75	147.962			

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Nd^{149}			1.7h	β^-1.5 γ or x	Nd^{148} (n, γ) Nd^{148} (d, p)
	Nd^{150}	5.65	149.964			
	Nd^{151}?			Short	β^-	Nd^{150} (n, γ)?
	Nd?			21m (11m)	β^-	Nd (n, γ)?
61	61^{144}?			~200d	K? γ0.67	Pr^{141} (α, n)? Nd $(d, ?)$
	61^{147}			3.7y	β^-0.20 No γ	Nd^{147}-β^-
	61^{148}			5.3d	β^-2.5 γ0.8	61^{147} (n, γ) Nd^{148} $(d, 2n)$ Nd^{148} (p, n)
	61^{149}			47h	β^-1.1 γ0.25	Nd^{149}-β^-
	61^{151}?			12m	β^-	Nd^{151}? (short)-β^-
	61^{153}			<5m	β^-	U (n, f)
	61^{156}			<5m	β^-	U (n, f)
	61?			2.7h	β^-2 γ	Nd $(d, ?)$ Nd $(p, ?)$

	61?		16d	β^-1.7 γ	Nd (d, ?)
	61?		12.5h	β^-	Nd (d, ?)
62	Sm^{144}	3.16			
	Sm^{145}		>70d	K?, γ0.95, 0.242 e^-0.194	Sm^{144} (n, γ)
	Sm^{147}	15.07			
	Sm^{148}	11.27			
	Sm^{149}	13.84			
	Sm^{150}	7.47			
	Sm^{151}		~20y	β^-0.06	Sm^{150} (n, γ) Eu^{151} (n, p)
	Sm^{152}	26.63	2.5×10^{11}y	α2.0	Natural
	Sm^{153}		47h	β^-0.78, (0.2) γ0.10, 0.61, 0.069 e^-0.021, 0.061, 0.062, 0.067, 0.069, 0.076, 0.081, 0.054, 0.095, 0.101, 0.103, 0.033, 0.034, 0.040	Sm^{152} (n, γ) Sm^{152} (d, p) 61^{153} (<5 m)-β^- Eu^{153} (n, p)
	Sm^{154}	22.53			

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Sm^{155}			25m	β^-1.9 γ~0.3	U (n, f) Sm^{154} (n, γ) Sm^{154} (d, p)
	Sm^{156}			~10h	β^-~0.8	61^{156}-β^-
63	$Eu^{147?}$			~53d		Sm $(d, ?)$
	$Eu^{149?}$			~14d		Sm $(d, ?)$
	$Eu^{150?}$			27h	β^+	Eu^{151} $(n, 2n)$?
	Eu^{151}	47.77				
	Eu^{152}			~5y	β^-0.75 e^-0.073, 0.114, 0.120	Eu^{151} (n, γ)
	Eu^{152}			9.2h	β^-1.88, K e^-0.113, 0.073, 0.675 γ0.725, 0.163, 0.123	Eu^{151} (n, γ) Eu^{151} (d, p) Sm^{152} $(d, 2n)$ Sm^{152} (p, n)
	Eu^{153}	52.23				
	Eu^{154}			~7y (~22y)	β^-0.62, 1.0 γ0.343, 0.408, 1.230, 0.247, 0.286, 0.773, 0.040 e^-0.394, 0.197, 0.236, 0.335, 0.343, 0.362	Eu^{153} (n, γ) Eu^{153} (d, p) Sm^{154} $(d, 2n)$ Sm^{154} (p, n)

	Eu^{155}			2y	$\beta^-\sim0.23$ $\gamma0.0844$	U (n, f) Sm^{154} (d, n)
	Eu^{156}			15.4d	$\beta^-0.5, 2.4$ $\gamma2.0$	Sm^{156}-β^-
	Eu^{157}			15.4h	$\beta^-1.0, 1.7$ $\gamma0.2, 0.6$	U (n, f)
	$Eu^{158?}$			60m	$\beta^-\sim2.6$	U (n, f)
	Eu?			40d		Sm $(d, ?)$
64	Gd^{152}	0.20				
	Gd^{153}			>72d (62d, 75d)	$e^-0.053, 0.094, 0.100$ $\gamma0.102$	Eu^{153} $(d, 2n)$ Eu^{153} (p, n)
	Gd^{154}	2.15	153.971			
	Gd^{155}	14.78	154.971			
	Gd^{156}	20.59	155.972			
	Gd^{157}	15.71	156.973			
	Gd^{158}	24.78	157.973			
	Gd^{160}	21.79	159.974			
	$Gd^{?}$			Long	$e^-0.039, 0.056$ $\gamma0.091, 0.108$	Gd (n, γ)
	$Gd^{?}$			4.5m		Gd (n, γ)

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	Gd?			8.6d		Gd (n, ?)
65	Tb^{152}?			4.5h	K?, e^-, x	Eu^{151} (α, $3n$)?
	Tb^{153}?			5.1d	K, x e^-0.15, 0.4	Eu^{151} (α, $2n$)?
	Tb^{154}?			17.2h	β^+2.6 K, e^-0.22, ~1, x γ1.4	Eu^{153} (α, $3n$)?
	Tb^{155}?			~1y	K?, e^-0.1, x	Eu^{153} (α, $2n$)?
	Tb^{159}	100				
	Tb^{160}			77d	β^-0.882, 0.546 e^- (many lines) γ0.086, 0.195, 0.213, 0.298, 1.15	Tb^{159} (n, γ) Tb^{159} (d, p) Gd^{160} (d, $2n$) Gd^{160} (p, n)
	Tb^{160}			3.9h	β^-	Tb^{159} (n, γ) Tb^{159} (d, p)
	Tb^{161}?			5.5d	β^-0.5 γ1.28	Gd (d, ?) Dy (n, ?)
	Tb^{161}?			18h	β^-0.85 γ0.3	Gd (d, ?) Dy (n, ?)

	Tb^{161}?		420d	β^-0.23, $\gamma\sim$0.1, 0.5	U (n, f)
66	Dy^{156}	0.05			
	Dy^{158}	$\sim$0.1			
	Dy^{160}	2.3			
	Dy^{161}	18.9			
	Dy^{162}	25.5			
	Dy^{163}	25.0			
	Dy^{164}	28.2			
	Dy^{165}		2.4h	β^-0.42, 0.88, 1.25 e^-0.0354, 0.0822 γ0.78, 0.37 (0.091)	Dy^{164} (n, γ) Dy^{164} (d, p) Ho^{165} (n, p)
	$\underline{Dy^{165}}$		1.25m	IT 0.18 e^-0.13	Dy^{164} (n, γ) Dy^{164} (d, p)
	Dy?		2.2m	β^+	Dy $(n, ?)$
67	Ho^{160}?		$\sim$20m	K?, x	Tb^{159} $(\alpha, 3n)$?
	$Ho^{161,162}$		60d	K?, x e^-0.16, 0.6 γ	Tb^{159} $(\alpha, 2n)$?
	$Ho^{162,161}$		4.5h	K, e^-0.3, x β^+, γ1.1	Tb^{159} (α, n)?

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Ho^{163}			7d	K, $e^-0.4$ γ, x	Dy (p, n)
	$Ho^{164?}$			35m (47m)	$\beta^-0.74$	Ho^{165} $(n, 2n)$? Dy (p, n)
	Ho^{165}	100				
	Ho^{166}			27.5h	$\beta^-1.8$	Ho^{165} (n, γ) Ho^{165} (d, p) Er^{166} (n, p)
68	Er^{162}	0.1				
	Er^{164}	1.5				
	$Er^{165?}$			1.1m	β^+	Er^{166} $(n, 2n)$?
	Er^{166}	32.9				
	Er^{167}	24.4				
	Er^{168}	26.9				
	Er^{169}			9.4d	$\beta^-0.33$ No γ	Er^{168} (n, γ) Er^{168} (d, p) Tm^{169} (n, p)
	Er^{170}	14.2				

	Er^{171}		7.5h	β^-1.05, 0.67, 1.49 γ0.305, 0.113, 0.81	Er^{170} (n, γ) Er^{170} (d, p) Yb^{174} (n, α)
	$Er^{169,171}$?		7m	β^-	Er (n, γ)
	$Er^{169,171}$?		20h	β^-0.6 γ	Er (n, γ)?
69	Tm^{166}		7.7h	β^+2.1, K, x γ1.5 e^-0.24, 1	Ho^{165} $(\alpha, 3n)$
	Tm^{167}		9d	K, e^-0.21, x γ0.22, 0.95	Ho^{165} $(\alpha, 2n)$
	$Tm^{168,167}$		~100d	K?, e^-0.16, 0.5, x	Ho^{165} $(\alpha, ?)$
	Tm^{169}	100			
	$Tm^{\underline{169}}$		~1μs	IT e^-0.12	Yb^{169}-K
	Tm^{170}		127d (105d)	β^-0.98 No γ	Tm^{169} (n, γ) Tm^{169} (d, p) Er^{170} $(d, 2n)$
	Tm^{171}		~2y	β^-0.10	Er^{171} (7.5h)-β^-
	$Tm^{\underline{171}}$		2.5μs	IT 0.113 γ0.113 e^-~0.1	Er^{171} (7.5h)-β^-

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
70	Yb^{168}	0.140				
	Yb^{169}			33d	K, Tm-x γ0.2, 0.4	Tm^{169} (d, $2n$) Tm^{169} (p, n)
	Yb^{170}	3.03				
	Yb^{171}	14.34				
	Yb^{172}	21.88				
	Yb^{173}	16.18				
	Yb^{174}	31.77				
	Yb^{175}			99h	β^-0.13, 0.50 γ0.35 Lu-x	Yb^{174} (n, γ) Yb^{174} (d, p) Lu^{175} (n, p)
	Yb^{175}?			45d	$\beta^-\sim$0.3	
	Yb^{176}	12.65				
	Yb^{177}			2.1h (1.9h, 2.4h, 3.5h)	β^-1.2	Yb^{176} (n, γ) Yb^{176} (d, p)
71	Lu^{170}			2.15d	K, e^-0.1, x γ1.5 β^+1.7	Tm^{169} (α, $3n$) Yb^{170} (d, $2n$) Yb^{170} (p, n)

	Lu^{171}		9d	K, x e^-0.17, 0.7 γ	Tm^{169} (α, $2n$) Yb^{171} (d, $2n$) Yb^{171} (p, n)
	$Lu^{172,171}$		>100d	K?, x, e^-0.11, 0.22, γ	Tm^{169} (α, n)?
	Lu^{175}	97.5			
	Lu^{176}	2.5	2.4×10^{10}y	K, Yb-x γ0.26 β^-0.40	Natural
	Lu^{176}		3.4h 3.67h	β^-1.15 No γ	Lu^{175} (n, γ) Lu^{175} (d, p)
	Lu^{177}		6.6d (6.8d)	β^-0.47	Lu^{176} (n, γ) Lu^{176} (d, p) Yb^{177}-β^-
72	Hf^{174}	0.18			
	Hf^{176}	5.30			
	Hf^{177}	18.47			
	Hf^{178}	27.10			
	Hf^{179}	13.84			
	$Hf^{177,179}$?		19s	IT e^-~0.19	Hf (n, γ)
	Hf^{180}	35.11			

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Hf^{181}			46d (55d)	β^-0.460 γ0.342, 0.128, 0.472 e^-0.061, 0.114, 0.275 Ta-x	Hf^{180} (n, γ) Ta^{181} (n, p) Hf^{180} (d, p)
73	Ta^{176}			8.0h	K, γ1.7, x e^-0.12, 0.18, 1.2	Lu^{175} $(\alpha, 3n)$ Hf^{176} (p, n) Hf^{176} $(d, 2n)$
	Ta^{177}			2.66d	K, x e^-0.11	Lu^{175} $(\alpha, 2n)$ Hf^{177} (p, n) Hf^{177} $(d, 2n)$
	Ta^{178}			16d	K e^-1.1?	Lu^{175} (α, n) Hf^{178} (p, n) Hf^{178} $(d, 2n)$
	Ta^{180}			8.2h	K e^-<0.5, γ β^-?	Ta^{181} $(n, 2n)$ Hf^{180} $(d, 2n)$ Hf^{180} (p, n)
	Ta^{180}?			14–21m		Ta^{181} $(n, 2n)$?
	Ta^{181}	100				
	Ta^{181}			22μs	IT 0.128 e^-0.114, 0.061, 0.42 γ0.472, 0.128	Hf^{181}-β^-

	Ta^{182}		16.2m	β^-0.4 γ or x	Ta^{181} (n, γ) Ta^{181} (d, p)
	Ta^{182}		117d	β^-0.525 γ1.22, 1.13, 0.22, 0.15 e^- many lines	Ta^{181} (n, γ) Ta^{181} (d, p) W^{182} (n, p)
74	$W^{179, 178}$		135m	K, x, e^-0.15, 0.45 γ ~0.5, 1.2	Ta^{181} $(d, ?)$
	W^{180}	0.122			
	W^{181}		140d	K, Ta-x γ1.83 e^-0.070	Ta^{181} $(d, 2n)$ Ta^{181} (p, n)
	W^{182}	25.80			
	W^{183}	14.24			
	W^{184}	30.68			
	W^{185}		73.2d	β^-0.43 No γ	W^{184} (n, γ) W^{184} (d, p) Re^{185} (n, p) Re^{187} (d, α)
	W^{186}	29.17			
	W^{187}		24.1h	β^-0.63, 1.33 γ0.48, 0.21, 0.69, 0.62, 0.138, 0.078 e^-0.066, 0.127, 0.007, 0.136	W^{186} (n, γ) W^{186} (d, p) Re^{187} (n, p)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
75	Re^{182}			64.0h	K, x e^-0.11, 0.27, 0.6 γ0.22, 1.52	Ta^{181} (α, $3n$) W^{182} (p, n) W^{182} (d, $2n$)
	$Re^{183, 184}$			13h	K?, x γ1.6	W (p, n)
	Re?			30–55m	β^+	W (p, n)?
	Re?			13m		W (p, n)?
	$Re^{184, 183}$			52d (~80d)	K, x, e^-0.1 γ1.0	W^{184} (d, $2n$) W^{184} (p, n) W^{183} (d, n) Ta^{181} (α, n)
	Re^{185}	37.07				
	Re^{186}			92.8h	β^-1.07 No γ	Re^{185} (n, γ) Re^{185} (d, p) W^{186} (p, n) W^{186} (d, $2n$)
	Re^{187}	62.93		4×10^{12}y	β^-0.043	Natural
	$Re^{\underline{187}}$			0.65μs	IT 0.13 e^-~0.13 x<0.1	W^{187}-β^-

	Re^{188}			18.9h	β^-2.05, 0.225 γ0.16, 0.64, 0.94, 0.48, 1.43 e^-0.12	Re^{187} (n, γ) Re^{187} (d, p) Os^{190} (d, α)
76	Os^{184}	0.018				
	Os^{185}			94.7d	K, x γ0.75	Re^{185} $(d, 2n)$ Re^{185} (p, n)
	Os^{186}	1.59				
	Os^{187}	1.64				
	Os^{188}	13.3				
	Os^{189}	16.1	189.04			
	Os^{190}	26.4	190.03			
	$Os^{191,193}$			15.0d	β^-0.142 e^- γ0.129, 0.039 x	Os (n, γ) Os (d, p) Ir (n, p)
	Os^{192}	41.0	192.04			
	$Os^{193,191}$			32h	β^-1.2 γ1.17, 1.58 e^-0.14	Os (n, γ) Os (d, p) Ir (n, p)
77	Ir^{190}			10.7d	K e^-0.091 γ0.25	Os^{190} $(d, 2n)$ Os^{190} (p, n)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Ir^{191}	38.5	191.04			
	Ir^{192}			70d	β^-0.67 γ0.137, 0.208, 0.296, 0.308, 0.317, 0.469, 0.488, 0.592, 0.607, 0.615 e^-	Ir^{191} (n, γ) Ir^{191} (d, p) Os^{192} $(d, 2n)$ Os^{192} (p, n)
	Ir^{192}			1.5m	IT 0.060, Ir-x e^-0.047 γ0.060	Ir^{191} (n, γ) Ir^{191} (d, p)
	Ir^{193}	61.5	193.04			
	Ir^{194}			19.0h	β^-2.2, 0.48 γ1.4, 0.3	Ir^{193} (n, γ) Ir^{193} (d, p)
78	Pt^{191}			3.00d	K, x e^-0.5 γ0.57, 1.8	Ir^{191} $(d, 2n)$ Ir^{191} (p, n)
	Pt^{192}	0.78				
	Pt^{193}			4.33d	K, x e^-0.115 γ0.17, 1.7	Ir^{193} $(d, 2n)$ Ir^{193} (p, n) Au^{193}-K
	Pt^{194}	32.8	194.039			

	Pt^{195}	33.7	195.039			
	Pt^{196}	25.4	196.039			
	$Pt^{196?}$			80m	IT e^-0.260, 0.323 (γ0.337)	Hg (n, α) Pt (d, ?)
	Pt^{197}			3.3d	β^- γ	Pt^{196} (n, γ) Pt^{196} (d, p) Au^{197} (n, p)
	Pt^{197}			18h	β^-0.7	Pt^{196} (n, γ) Pt^{196} (d, p) Au^{197} (n, p)
	Pt^{198}	7.23	198.05			
	Pt^{199}			31m	β^-1.8	Pt^{198} (n, γ) Pt^{198} (d, p)
79	$Au^{191?}$			~1d	K or β^+	Ir^{191} (α, $4n$)
	Au^{192}			4.7h	K?, x e^-~0.3, (>2) γ~3	Ir^{191} (α, $3n$)
	Au^{193}			15.8h	K?, x e^-<0.3	Ir^{191} (α, $2n$) Pt^{194} (d, $3n$)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	$Au^{194?}$			39.5h	K, x e^-0.251, 0.315, 0.208, 0.382, 0.446, 1.40 γ0.286, 0.329, 0.460, 1.48, 2.00	Ir^{191} (α, n) Pt^{194} (d, $2n$) Pt^{194} (p, n)
	Au^{195}			195d	K, x; no β^+ e^-0.082, 0.051, 0.115 γ0.096, 0.129	Pt^{194} (d, n) Pt^{195} (d, $2n$) Pt^{195} (p, n)
	Au^{196}			14h	β^- x	Au^{197} (n, $2n$) Pt^{195} (d, n)
	Au^{196}			5.5d	K, x, e^-0.280, 0.344, 0.061, 0.124 γ0.358, 0.139 β^-0.27, 0.43 γ0.334, 0.173 e^-0.251, 0.319, 0.091, 0.158	Au^{197} (n, $2n$) Pt^{195} (d, n) Pt^{196} (d, $2n$) Pt^{196} (p, n)
	Au^{197}	100	197.04			
	Au^{197}			7.4s	IT 0.273 e^-0.192, 0.260 γ0.273	Hg^{197} (23h)-K Au^{197} (n, n)

	Au^{198}		2.66d	β^-0.960 γ0.4112 ϵ^-0.330	Au^{197} (n, γ) Au^{197} (d, p) Hg^{200} (d, α)
	Au^{199}		3.3d	β^-0.38 γ0.025, 0.052, 0.164, 0.214 ϵ^-	Pt^{199}-β^- Hg^{201} (d, α)
	$Au^{200,202?}$		48m	β^-2.5	Hg (n, p) Tl (n, α)
80	Hg^{196}	0.155			
	Hg^{197}		64h	K, Au-x γ0.077 ϵ^-0.063, 0.0735, 0.0765	Au^{197} $(d, 2n)$ Au^{197} (p, n)
	$Hg^{\underline{197}}$		23h	K, Au-x γ0.165, 0.135, 0.250, 0.077 ϵ^-0.083, 0.151, 0.1615, 0.1205, 0.131	Au^{197} $(d, 2n)$ Au^{197} (p, n)
	Hg^{198}	10.12			
	Hg^{199}	17.01			
	Hg^{200}	23.21			
	Hg^{201}	13.66			
	$Hg^{\underline{199}, \underline{201}, \underline{204}}$		43m	IT, Hg-x ϵ^-0.139, 0.275, 0.351 (γ0.223, 0.365)	Hg $(n, ?)$ Hg (d, p)

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Hg^{202}	29.66				
	Hg^{203}			51.5d (43.5d)	β^-0.205 γ0.286 e^-0.20, 0.27	Hg^{202} (n, γ) Hg^{202} (d, p) Tl^{203} (n, p)
	Hg^{204}	6.69				
	Hg^{205}			5.5m	β^-1.62	Hg^{204} (n, γ) Hg^{204} (d, p) Tl^{205} (n, p)
81	$Tl^?$			10.5h	K? γ1.0 e^-	Hg $(d, 2n)$
	$Tl^?$			44h	K? e^-	Hg $(d, 2n)$
	Tl^{198}			1.8h	K, e^-0.4 γ1.3	Au^{197} $(\alpha, 3n)$
	Tl^{199}			7h	K, e^-0.5 γ1.5	Au^{197} $(\alpha, 2n)$
	Tl^{200}			27h	K, e^-0.4 γ	Au^{197} (α, n)
	$Tl^{200?}$			4m		Au^{197} (α, n)?

	Tl^{202}			11.8d 13d	K $\gamma 0.40$ e^-	Hg^{202} $(d, 2n)$ Hg^{202} (p, n) Tl^{203} $(n, 2n)$
	Tl^{203}	29.46	203.05			
	Tl^{204}			3.5y	$\beta^- 0.87$ No γ	Tl^{203} (n, γ) Tl^{203} (d, p)
	Tl^{205}	70.54	205.05			
	Tl^{206} (RaE″)			4.23m	$\beta^- 1.7$ No γ	Tl^{205} (n, γ) Tl^{205} (d, p)
	Tl^{207} (AcC″)			4.76m	$\beta^- 1.47$ $\gamma 0.46, 0.48$	Pb^{207} (n, p) Bi^{211} (AcC)-α
	Tl^{208} (ThC″)			3.1m	$\beta^- 1.805$, (others) $\gamma 2.62, 0.57, 0.51, 0.277$	Pb^{208} (n, p) Bi^{212} (ThC)-α
	Tl^{209}			2.2m	$\beta^- 1.8$	Bi^{213}-α
	Tl^{210} (RaC″)			1.32m	$\beta^- 1.80$	Bi^{214} (RaC)-α
82	$Pb^{201?}$			$\sim$5h	K, e^-, γ	Tl^{203} $(d, 4n)$?
	Pb^{203}			52h	K $e^- 0.15$ $\gamma 0.45$	Tl^{203} $(d, 2n)$ Tl^{203} (p, n)
	Pb^{204}	1.5	204.05			

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Pb^{204}			68m	IT 1.1 γ, e^-	Tl^{203} (d, n) Bi^{204}-K Pb^{204} (n, n)
	Pb^{206}	23.59	206.05			
	Pb^{207}	22.64	207.05			
	Pb^{208}	52.29	208.05			
	Pb^{209}			3.3h	β^-0.68 No γ	Pb^{208} (n, γ) Pb^{208} (d, p) Bi^{209} (n, p)
	Pb^{210} (RaD)			22y	β^-0.0255, x γ0.047, 0.0078, 0.0258	Po^{214} (RaC′)-α
	Pb^{211} (AcB)			36.1m	β^-0.5, 1.40 γ0.8	Po^{215} (AcA)-α
	Pb^{212} (ThB)			10.6h	β^-0.36, (0.59) γ0.43, 0.71, 0.238 e^-	Po^{216} (ThA)-α
	Pb^{214} (RaB)			26.8m	β^-0.65 γ0.0528, 0.2410, 0.2581, 0.294, 0.351 e^-	Po^{218} (RaA)-α

83	$Bi^{<203}$?			2m	$\alpha\sim5.5$	Bi^{209} (d, ?)
	$Bi^{<203}$			9m	$\alpha\sim5.5$	Bi^{209} (d, ?)
	$Bi^{<203}$			27m	K ($\alpha\sim5.5$)	Bi^{209} (d, ?) Pb^{204} (d, ?)
	$Bi^{<203}$			100m 1–2h	K ($\alpha\sim5.5$)	Bi^{209} (d, ?) Pb^{204} (d, ?)
	Bi^{204}			12h	K, γ $e^-0.20$, 0.75?	Tl^{203} (α, $3n$) Pb^{204} (d, $2n$)
	Bi^{206}			6.4d	K, x $\gamma 1.1$, 0.4 $e^-<0.85$	Pb^{206} (d, $2n$) Pb^{206} (p, n)
	Bi^{208}?			Long	e^-, γ	Bi^{209} (n, $2n$)?
	Bi^{209}	100	209.05			
	Bi^{210} (RaE)			5.0d	$\beta^-1.17$ ($\alpha 4.87$)	Bi^{209} (n, γ) Bi^{209} (d, p) Pb^{210} (RaD)-β^-
	Bi^{211} (AcC)			2.16m	$\alpha 6.62$, 6.26 (β^-) $\gamma 0.354$	Pb^{211} (AcB)-β^-
	Bi^{212} (ThC)			60.5m	$\beta^-2.256$ $\gamma 0.73$, 1.80 $\alpha 6.05$, 6.08, 5.76, 5.60, (5.62) $\gamma 0.15$ to 2.20	Pb^{212} (ThB)-β^-

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Bi^{213}			47m	β^-1.2 α5.86	At^{217}-α
	Bi^{214} (RaC)			19.7m	β^-3.15 (α5.50, 5.45) γ1.8 (0.607–2.20)	Pb^{214} (RaB)-β^-
84	Po^{206}			9d	K, x e^-, γ0.8 α5.2	Pb^{204} (α, $2n$) Bi^{209} (d, $5n$)
	Po^{207}			5.7h	K, x γ1.3, e^-? (α5.1)	Pb^{206} (α, $3n$) Bi^{209} (d, $4n$)
	Po^{208}			3y	α5.14	Pb^{206} (α, $2n$) Bi^{209} (d, $3n$)
	Po^{210} (RaF)			140d	α5.298 (γ0.773, 0.084)	Bi^{210} (RaE)-β^-
	Po^{211} (AcC′)			5×10^{-3}s	α7.43	Bi^{211} (AcC)-β^-
	Po^{212} (ThC′)			0.30μs 0.22μs	α8.78 (10.56, 9.47)	Bi^{212} (ThC)-β^-
	Po^{213}			4.2μs	α8.34	Bi^{213}-β^-

	Po^{214} (RaC′)	150μs	α7.68 (8.28–10.51)	Bi^{214} (RaC)-β^-
	Po^{215} (AcA)	1.83×10^{-3}s	α7.365 (β^-?)	Rn^{219} (An)-α
	Po^{216} (ThA)	0.158s	α6.77 (β^-)	Rn^{220} (Tn)-α
	Po^{218} (RaA)	3.05m	α6.00 (β^-)	Rn^{222} (Rn)-α
85	At^{207}?	1.7h	α5.76, K?	Bi^{209} (α, 6n)
	At^{209}?	4.5h	α5.66, K?	Bi^{209} (α, 4n)
	At^{210}	8.3h	K, α? γ1.0	Bi^{209} (α, 3n)
	At^{211}	7.5h	K α5.94	Bi^{209} (α, 2n)
	At^{212}	0.25s	α	Bi^{209} (α, n)
	At^{214}	Short	α8.78	Fr^{218}-α
	At^{215}	$\sim 10^{-4}$s	α8.00 (8.43?)	Fr^{219}-α Po^{215} (AcA)-β^-?
	At^{216}	$\sim 3 \times 10^{-4}$s	α7.79 (7.66?)	Fr^{220}-α Po^{216} (ThA)-β^-?
	At^{217}	0.020s	α7.02	Fr^{221}-α

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	At^{218}			Few s	α6.72, (β^-)	Po^{218} (RaA)-β^-
86	Rn^{216}			Short	α8.07	Ra^{220}-α
	Rn^{217}			$\sim 10^{-3}$s	α7.74	Ra^{221}-α
	Rn^{218}			0.019s	α7.12	Ra^{222}-α
	Rn^{219} (An)			3.92s	α6.82 (6.56, 6.44)	Ra^{223} (AcX)-α
	Rn^{220} (Tn)			54.5s	α6.28	Ra^{224} (ThX)-α
	Rn^{222} (Rn)			3.825d	α5.486	Ra^{226}-α
87	Fr^{218}			Short	α7.85	Ac^{222}-α
	Fr^{219}			$\sim$0.02s	α7.30	Ac^{223}-α
	Fr^{220}			27.5s	α6.69	Ac^{224}-α
	Fr^{221}			4.8m	α6.30	Ac^{225}-α
	Fr^{223} (AcK)			21m	β^-1.20 γ0.095	Ac^{227}-α
88	Ra^{220}			Short	α7.5	Th^{224}-α
	Ra^{221}			31s	α6.71	Th^{225}-α

	Ra^{222}	38s	α6.51	Th^{226}-α
	Ra^{223} (AcX)	11.2d	α5.72, 5.61, 5.53 γ0.143, 0.153, 0.157, 0.200, 0.269	Th^{227} (RdAc)-α
	Ra^{224} (ThX)	3.64d	α5.68	Th^{228} (RdTh)-α
	Ra^{225}	14.8d 14d	β^-~0.2 or <0.05	Th^{229}-α
	Ra^{226} (Ra)	1590y 1622y 1631y	α4.79, 4.61 (4.22, 4.17, 4.12, 4.05, 4.35)? γ0.19	Th^{230} (Io)-α
	Ra^{227}		β^-	Ra^{226} (n, γ)
	Ra^{228} ($MsTh_1$)	6.7y	β^-<0.005	Th^{232}-α
89	Ac^{222}	Short	α6.96	Pa^{226}-α
	Ac^{223}	2.2m	α6.64	Pa^{227}-α
	Ac^{224}	2.9h	K α6.17	Pa^{228}-α
	Ac^{225}	10.0d	α5.80	Ra^{225}-β^-
	Ac^{227}	21y	β^-<0.05 γ0.037 α4.95	Pa^{231}-α

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Ac^{228} ($MsTh_2$)			6.13h	β^-1.55 γ0.058–0.970 (α4.5) e^-	Ra^{228} ($MsTh_1$)-β^-
90	Th^{224}			Short	α7.20	U^{228}-α
	Th^{225}			7.8m	α6.57, K	U^{229}-α
	Th^{226}			30.9m	α6.30	U^{230}-α
	Th^{227} (RdAc)			18.9d	α5.99, 6.05, 5.76, 5.72 γ0.032–0.300	Ac^{227}-β^-
	Th^{228} (RdTh)			1.90y	α5.42, 5.34 γ0.085, 0.088	Ac^{228} ($MsTh_2$)-β^-
	Th^{229}			7×10^3y	α4.85	U^{233}-α
	Th^{230} (Io)			8.3×10^4y 8.0×10^4y	α4.68, 4.61, 4.51 γ0.068, e^-, x	U^{234} (U_{II})-α
	Th^{231} (UY)			25.5h	β^-0.21, e^-0.016, x γ0.035	U^{235} (AcU)-α Th^{232} (n, $2n$)
	Th^{232}	100	232.11	1.39×10^{10}y	α4.20	Natural
	Th^{233}			23m	β^-1.6 No γ	Th^{232} (n, γ) Th^{232} (d, p)

	Th^{234} (UX_1)	24.5d (24.1d)	β^-0.190 or 0.130? ϵ^-0.070, 0.085, 0.090 γ0.092	U^{238} (U_I)-α
91	Pa^{226}	1.7m	α6.81	Th^{232} (d, $8n$)
	Pa^{227}	38m	α6.46 K	Th^{232} (d, $7n$)
	Pa^{228}	22h	K α6.09	Th^{232} (d, $6n$)
	Pa^{229}	1.4d	K, α5.66	Th^{230} (d, $3n$)
	Pa^{230}	17d	$\beta^-\sim$1.1 γ0.94	Th^{232} (d, $4n$) Th^{232} (α, $p5n$)
	Pa^{231}	3.2×10^4y 3.43×10^4y	α5.04, 4.72, 4.69, 5.07, 5.13 γ0.29, 0.32	Th^{231} (UY)-β^-
	Pa^{232}	1.33d	β^-0.14, 0.4, 1.0 γ1.1	Th^{232} (d, $2n$) Th^{232} (p, n)
	Pa^{233}	27.4d	β^-0.23 e^- γ0.310, 0.338, 0.298, 0.084	Th^{232} (d, n) Th^{233}-β^-
	Pa^{234} (UZ)	6.7h	β^-0.45, 1.2 γ0.85, 0.75	Th^{234} (UX_1)-β^-

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Pa^{234} (UX_2)			1.14m 1.22m	β^-2.32, 1.50, 1.54, (0.82) γ0.78, 0.82, (1.5) (IT 0.394, e^-)	Th^{234} (UX_1)-β^-
92	U^{228}			9.3m	α6.72, K	$Th^{232}(\alpha, 8n)$ Pu^{232}-α
	U^{229}			58m	K, α6.42	Th^{232} (α, $7n$)
	U^{230}			20.8d	α5.85	Pa^{230}-β^- Th^{232} (α, $6n$)
	U^{231}			4.2d	K	Pa^{231} (d, $2n$)
	U^{232}			70y	α5.3	Th^{232} (α, $4n$) Pa^{232}-β^-
	U^{233}			1.63×10^5y	α4.82 e^-, γ0.31, 0.080, 0.040	Pa^{233}-β^-
	U^{234} (U_{II})	0.0052		2.32×10^5y	α4.75	Pa^{234} (UX_2, UZ)-β^-
	U^{235} (AcU)	0.719		7.07×10^8y 8.91×10^8y	α4.40 γ0.18	Natural

	U^{237}			6.8d 6.63d	β^-0.23 γ0.057, 0.203, 0.259, 0.032 e^-0.034–0.237 x	U^{238} $(n, 2n)$
	U^{238} (U_I)	99.276	238.12	4.51×10^9y	α4.21	Natural
	U^{239}			23m	β^-1.12, 2.06 γ0.073, 0.870 e^-0.051, 0.068	U^{238} (n, γ) U^{238} (d, p)
93	Np^{231}			53m	K, α6.2	U^{238} $(d, 9n)$
	Np^{234}			4.4d	K γ1.9	U^{235} $(d, 3n)$ Pa^{231} (α, n) Pu^{234}-K
	Np^{235}			435d	K, (α5.06)	U^{235} $(d, 2n)$
	Np^{236}			22h	β^-, γ	U^{235} (d, n) U^{238} $(d, 4n)$
	Np^{237}			2.25×10^6y	α4.73	U^{238} $(d, 3n)$ U^{237}-β^-
	Np^{238}			2.0d	β^-0.22, 1.39 γ1.2, 0.075, e^-	U^{238} $(d, 2n)$

Atomic Number	Symbol and Mass Number	Per Cent Abundance	Isotopic Mass	Half-life	Modes of Decay and Energies of Radiations (in Mev)	Some Modes of Formation
	Np^{239}			2.3d	β^-0.288, 0.406, 0.679, 1.181 γ0.275, 0.227, 0.206, 0.067, 0.061, 0.057 e^-0.034–0.255	U^{239}-β^-
94	Pu^{232}			22m	α6.6	U^{235} (α, 7n)
	Pu^{234}			8h	K (α6.1)	U^{235} (α, 5n)
	Pu^{236}			2.7y	α5.7	Np^{236}-β^-
	Pu^{237}			~40d	K, No γ	U^{238} (α, 5n)
	Pu^{238}			90y	α5.49	Np^{238}-β^-
	Pu^{239}			2.41×10^4y	α5.15 (γ0.42, 0.2)	Np^{239}-β^-
	Pu^{240}			~6×10^3y	α5.1	U^{238} (α, 2n)
	Pu^{241}			~10y	β^-0.01–0.02	U^{238} (α, n)
95	Am^{238}?			1.5h		Pu^{239} (d, 3n)? Np^{237} (α, 3n)?

	Am^{239}	12h	K, $\gamma 0.285$ e^-, x, ($\alpha 5.77$)	Pu^{239} (d, $2n$) Np^{237} (α, $2n$)
	Am^{240}	50h	K, e^-, $\gamma 1.3$, x	Pu^{239} (d, n) Np^{237} (α, n)
	Am^{241}	510y	$\alpha 5.45$ $\gamma 0.060$ Np-x	Pu^{241}-β^-
	Am^{242}	17h	$\beta^- \sim 0.8$	Pu^{239} (α, p) Am^{241} (n, γ)
	Am^{242}	$\sim$400y	$\beta^- \sim 0.5$ (α)	Pu^{239} (α, p) Am^{241} (n, γ)
96	Cm^{240}	26.8d	$\alpha 6.3$	Pu^{239} (α, $3n$)
	Cm^{241}	$\sim$55d	K	Pu^{239} (α, $2n$)
	Cm^{242}	150d	$\alpha 6.1$	Pu^{239} (α, n) Am^{242}-β^-

TABLE B

Isotopic Thermal Neutron Activation Cross Sections †

Target Isotope	Half-life of Product Isotope	Isotopic Cross Section for (n, γ) Reaction (Barns)
O^{18}	27s	0.00022
F^{19}	12s	0.0094
Na^{23}	14.8h	0.63
Mg^{26}	10.2m	0.048
Al^{27}	2.4m	0.21
Si^{30}	170m	0.159 *
P^{31}	14.3d	0.23
S^{34}	87.1d	0.26
Cl^{37}	38m	0.56
K^{41}	12.4h	1.0
Ca^{44}	180d	0.63
Ca^{48}	30m	0.55
Ca^{48}	2.5h	0.205
Sc^{45}	85d	22
Ti^{50}	6m	0.0075
Ti^{50}	72d	0.039
V^{51}	3.74m	4.50
Cr^{50}	26.5d	(11)
Cr^{54}	1.3h	($\sim$0.0061)
Mn^{55}	2.59h	10.7
Fe^{58}	47d	0.36
Co^{59}	5.3y	21.7
Ni^{64}	2.6h	2.31 *
Cu^{63}	12.8h	2.88 *
Cu^{65}	5m	1.83
Zn^{64}	250d	0.53
Zn^{68}	57m	1.02
Zn^{68}	13.8h	0.29
Ga^{69}	20m	1.40
Ga^{71}	14.1h	3.36
Ge^{70}	11d	($\sim$0.45)
Ge^{74}	89m	0.38
Ge^{76}	12h	0.085
As^{75}	26.8h	4.2
Se^{74}	127d	(22)
Se^{80}	18m	0.48 *
Se^{80}	58m	0.035 *
Se^{82}	25m	0.060

† From L. Seren, H. N. Friedlander, and S. H. Turkel, *Phys. Rev.* **72**, 888 (1947).

TABLE B (*Continued*)

ISOTOPIC THERMAL NEUTRON ACTIVATION CROSS SECTIONS

Target Isotope	Half-life of Product Isotope	Isotopic Cross Section for (n, γ) Reaction (Barns)
Br^{79}	18m	8.1
Br^{79}	4.4h	2.76
Br^{81}	34h	2.25
Rb^{85}	19.5d	0.72
Rb^{87}	17.8m	0.122
Sr^{86}	2.7h	1.29
Sr^{88}	54d	0.0050
Y^{89}	65h	1.24
Zr^{94}	65d	(0.43) *
Zr^{96}	17.0h	0.60 *
Cb^{93}	6.6m	($\sim$1.0)
Mo^{92}	6.7h	($<$0.007)
Mo^{98}	67h	0.42 *
Mo^{100}	14.6m	0.46 *
Ru^{102}	42d	1.2 *
Ru^{104}	4.5h	0.67
Rh^{103}	42s	[137]
Rh^{103}	4.3m	[11.6]
Pd^{108}	13.4h	11.2
Pd^{110}	26m	0.39
Ag^{107}	2.3m	44
Ag^{109}	22s	97
Ag^{109}	225d	2.3
Cd^{112}	2.3m	0.21 *
Cd^{114}	2.33d	1.0 *
Cd^{114}	44d	0.14 *
Cd^{116}	3.75h	1.3 *
In^{113}	48d	[56.0]
In^{115}	13s	[51.8]
In^{115}	54m	[145]
Sn^{112}	105d	($\sim$1.1)
Sn^{120}	28h	0.25 *
$Sn^{122?}$	10d	0.16 *
Sn^{124}	9m	0.57
Sb^{121}	2.8d	6.8
Sb^{123}	60d	2.5
Te^{126}	9.3h	0.80 *
Te^{126}	90d	0.075 *
Te^{128}	70m	0.137 *
Te^{128}	32d	0.016 *
Te^{130}	25m	0.21 *

TABLE B (*Continued*)

ISOTOPIC THERMAL NEUTRON ACTIVATION CROSS SECTIONS

Target Isotope	Half-life of Product Isotope	Isotopic Cross Section for (n, γ) Reaction (Barns)
Te^{130}	30h	(<0.008)
I^{127}	25m	6.25
Cs^{133}	3.1h	0.016
Cs^{133}	2.0y	25.6
Ba^{138}	85m	0.51
La^{139}	40h	8.4
Pr^{141}	19.3h	10.1
Sm^{152}	47h	135 *
Sm^{154}	25m	4.9 *
Eu^{151}	9.2h	1420 *
Gd^{160}	18h	(~4.3) *
Tb^{159}	3.9h	10.7
Dy^{164}	2.4h	[2660] *
Ho^{165}	27.5h	59.6
Tm^{169}	105d	106
Lu^{175}	3.4h	16.3 *
Lu^{176}	6.6d	3640
Hf^{180}	46d	10.0
Ta^{181}	16.2m	0.034
Ta^{181}	120d	20.6
W^{184}	73.2d	2.08 *
W^{186}	24.1h	35 *
Re^{185}	92.8h	101
Re^{187}	18.9h	75
$Os^{190?}$	32h	2.50
$Os^{192?}$	15d	5.34
Ir^{191}	1.5m	(260)
Ir^{191}	70d	1000
Ir^{193}	19h	128
Pt^{196}	18h	1.1
Pt^{196}	3.3d	(4.5)
Pt^{198}	31m	3.9
Au^{197}	2.7d	[96.4]
Hg^{202}	51.5d	2.44 *
Hg^{204}	5.5m	0.34
Tl^{203}	3.5y	7.5 *
Tl^{205}	4.23m	0.11 *
Bi^{209}	5.0d	0.015

The probable errors of the cross-section values are estimated as ±20 per cent, except where the cross section is given in parentheses (±40 per cent) or in brackets [±10 per cent].

Whenever the abundance value for the target isotope or the assignment of the active isotope used by Seren *et al.* differs from those given in table A, the isotopic cross section was recalculated from the atomic cross section given by Seren *et al.* with the data from table A. This is denoted by an asterisk.

TABLE C

THICK-TARGET YIELDS FOR SOME NUCLEAR REACTIONS OBTAINED WITH 14-MEV DEUTERONS *

Reaction	Yield in Rutherfords per microampere-hour
Mg^{24} (d, α) Na^{22}	0.065
Na^{23} (d, p) Na^{24}	410
Mg^{26} (d, α) Na^{24}	8.70
Al^{27} $(d, p\alpha)$ Na^{24}	1.66
Si^{30} (d, p) Si^{31}	37
P^{31} (d, p) P^{32}	8.5
Cl^{37} (d, p) Cl^{38}	1500
K^{41} (d, p) K^{42}	8.3
Cr^{52} $(d, 2n)$ Mn^{52}	3
Co^{59} (d, p) Co^{60} (5.3y)	0.040
Cu^{63} (d, p) Cu^{64}	92
Cu^{63} $(d, 2n)$ Zn^{63}	1160
Cu^{65} $(d, 2n)$ Zn^{65}	0.126
Br^{81} (d, p) Br^{82}	28.5
Sr^{88} (d, p) Sr^{89}	1.3
Sr^{88} $(d, 2n)$ Y^{88}	1.4
Te^{130} $(d, 2n)$ I^{130}	32
Te^{130} (d, n) I^{131}	3.2

* Data from E. T. Clarke and J. W. Irvine Jr., *Phys. Rev.* **70,** 893 (1946).

The yields listed are those that would be obtained with thick targets of the pure target elements of natural isotopic composition, and in bombardments short compared to the product half-lives.

TABLE D

TABLE OF PHYSICAL CONSTANTS AND CONVERSION FACTORS *

Velocity of light	c	$= (2.99776 \pm 0.00004) \times 10^{10}$ cm sec^{-1}
Planck constant	h	$= (6.624 \pm 0.002) \times 10^{-27}$ erg sec
Boltzmann constant	k	$= (1.38047 \pm 0.00026) \times 10^{-16}$ erg deg^{-1}

TABLE D (*Continued*)

TABLE OF PHYSICAL CONSTANTS AND CONVERSION FACTORS

Electronic charge	$e = (4.8025 \pm 0.0010) \times 10^{-10}$ absolute esu $= (1.60203 \pm 0.00034) \times 10^{-19}$ absolute coulomb
Electron mass	$m = (9.1066 \pm 0.0032) \times 10^{-28}$ g $= (5.4862 \pm 0.0017) \times 10^{-4}$ physical atomic weight unit
Electron rest energy	$mc^2 = 0.5107$ Mev
Neutron mass	1.00893 ± 0.00004 physical atomic weight units
Hydrogen atom mass	1.008125 ± 0.000009 physical atomic weight units
Ratio of physical atomic weight to chemical atomic weight	1.000272 ± 0.000005
Avogadro number (chemical scale)	$N = (6.0228 \pm 0.0011) \times 10^{23}$ mole^{-1}
Faraday constant (chemical scale)	$F = 96{,}487 \pm 10$ absolute coulombs g-equiv.$^{-1}$
Energy equivalent of one mass unit	931.05 ± 0.15 Mev
Energy in ergs of one absolute-volt-electron	$(1.60203 \pm 0.00034) \times 10^{-12}$ erg
Energy in calories per mole for one absolute-volt-electron per molecule	23,052 ± 3 cal_{15} mole^{-1}

* Mostly based on values given by R. T. Birge, *Rev. Modern Phys.* **13**, 233 (1941).

SELECTED EXAMINATION QUESTIONS FROM AN INTRODUCTORY COURSE IN RADIOCHEMISTRY

1. Diagram the decay of U^{238} to radium. Show the names, atomic numbers, mass numbers, half-lives, and modes of decay of the active substances.

2. $_{26}Fe^{56}$ is stable. $_{25}Mn^{56}$ is radioactive, half-life 2.59 hr, and emits negative β particles of 2.9 Mev maximum kinetic energy. Mn^{56} can be made by the action of neutrons on Fe^{56}. Is the reaction exoergic? Estimate the minimum practical neutron energy (threshold).

3. Suggest two different reasons for believing that attractive forces between nuclear particles have a very short range.

4. Define (*a*) isotopes, (*b*) isobars, (*c*) isomers, (*d*) Oppenheimer–Phillips process, (*e*) packing fraction, (*f*) secular equilibrium, (*g*) Compton effect, (*h*) Bragg curve, (*i*) saturation current, (*j*) betatron.

5. Why is He^5 unstable?

6. Derive the relationship between half-life and disintegration constant.

7. What is meant by the "three radioactive series"? Why do we say that one series is missing?

8. Define briefly: (*a*) (α, $2n$) reaction; (*b*) binding energy of a nucleus; (*c*) chemical scale of atomic weights; (*d*) physical scale of atomic weights; (*e*) "one over v" law.

9. In connection with nuclear reactions, why is there so much emphasis on *slow* neutrons? Why so little concern with *slow* protons, α particles, etc.?

10. Define the following: (*a*) internal-conversion coefficient of a γ ray; (*b*) half-life of a radioactive substance; (*c*) threshold of a Geiger–Müller counter; (*d*) probability; (*e*) a compound nucleus; (*f*) excitation function of a nuclear reaction; (*g*) exchange reaction.

11. The stable isotopes of a hypothetical element Z and its neighbors in the periodic table are given in the following chart:

$Z-2$	44	45	46		48		50							
$Z-1$				47		49								
Z					48		50	51	52					
$Z+1$										53				
$Z+2$											54	55	56	

Five radioactive isotopes of element Z have been prepared in the course of the bombardments listed in the following table. Their half-lives are 10 min, 45 min, 4 hr, 20 hr, and 14 days.

Target	Bombarding Particle	Bombarding Energy	Resulting Half-lives Chemically Identified as Belonging to Element Z
$Z-2$	α	30 Mev	10 min, 45 min, 4 hr, 20 hr, 14 days
$Z-1$	p	8 Mev	10 min, 20 hr, 14 days
$Z-1$	d	15 Mev	10 min, 45 min, 20 hr, 14 days
Z	n	Thermal	10 min, 4 hr, 14 days
Z	n	Up to 22 Mev	10 min, 45 min, 4 hr, 20 hr, 14 days
$Z+1$	n	Up to 22 Mev	4 hr
$Z+2$	n	Up to 22 Mev	4 hr

(*a*) To what mass number is each of the five radioactive isotopes to be assigned? State your reasoning.

(*b*) What mode of decay would you expect for each of these active isotopes? Where several possibilities appear about equally probable from the data given, state all alternatives.

12. Answer any four parts of this question. How would you distinguish experimentally between (*a*) α particles and β particles; (*b*) β particles and internal-conversion electrons; (*c*) X rays and γ rays; (*d*) positive and negative electrons; (*e*) K-electron capture and an isomeric transition in a given element.

13. (*a*) Discuss the conditions necessary for the operation of the Szilard–Chalmers process.

(*b*) Why is the chemical separation of nuclear isomers possible in some cases even though the recoil energy of the γ rays or conversion electrons is not sufficient to break the chemical bonds in question?

14. What is the weight of 1 "curie" of H^3?

15. 1.00 g of rubidium in the form of a thin foil is exposed to a thermal neutron flux of 1.00×10^{10} neutrons $cm^{-2}sec^{-1}$ for 48 hr. At the end of this irradiation what would be the total rate of β disintegrations (from the two induced radioactivities, Rb^{86} and Rb^{88})? The thermal neutron radiative capture cross sections are: for Rb^{85} $\sigma = 0.72$ barn, for Rb^{87} $\sigma = 0.122$ barn.

16. In a set of data, what is meant by (*a*) standard deviation; (*b*) probable error; (*c*) dispersion?

17. After ethyl bromide is bombarded with neutrons, a substantial fraction of the active bromine formed is found in the form of ethyl bromide. In what other chemical species would you expect to find bromine activity? How would you expect the "retention" of bromine activity in ethyl bromide to be affected by dilution of the ethyl bromide with ethyl alcohol before bombardment? Explain.

18. What conclusions would you draw from each of the following experimental results: (*a*) Br_2 and $AsBr_3$ are found to exchange active bromine rapidly in CCl_4 solution. (*b*) I_2 exchanges more readily with C_2H_5I than with C_6H_5I.

19. List three neutron sources that employ natural radioactivities and two neutron sources made possible by accelerating devices. Write down the nuclear reaction responsible for the production of neutrons in each case.

20. Describe in detail how you would best determine experimentally the maximum energy of the beta particles from a given radioactive substance without recourse to an electron spectrograph.

21. The stable isotopes in the region of the hypothetical element Z are:

$Z-2$		87							
$Z-1$	86		88		90				
Z				89		91			
$Z+1$					90		92	93	94
$Z+2$									95

Assuming you have available a cyclotron capable of producing 12-Mev deuterons, 6-Mev protons, or 24-Mev α particles, list the reactions by which you would expect to be able to produce in reasonably good yield each of the following isotopes: Z^{88}, Z^{90}, and Z^{92}.

22. Discuss the Bohr theory of nuclear reactions.

23. (*a*) Explain the principle of the Szilard–Chalmers reaction. (*b*) Suggest possible methods of applying the Szilard–Chalmers process to each of the following elements: iodine, lead, tellurium, arsenic.

24. How would you explain the following experimental facts: If Ag^+ is added as a carrier to a ThB solution, and 10 per cent excess I^- is added, 77 per cent of the ThB is carried by the AgI precipitate. If only 90 per cent of the stoichiometric amount of I^- is added, only 4 per cent of the ThB is carried.

25. (*a*) Briefly describe the main features of a cyclotron.

(*b*) What is the principal distinction between an ionization chamber and a Geiger counter?

26. Discuss the concept of probability and derive the radioactive decay law from probability considerations.

27. Discuss briefly the difficulties in the theory of β decay which led to the neutrino hypothesis.

28. What is wrong with this hypothetical table of stable isotopes?

Atomic Number	Mass Numbers					
Z − 2		49				
Z − 1			50	52	54	55
Z		49	51			
Z + 1	48		50	52	53	54
Z + 2				51	53	

29. Name three quantized nuclear properties; give (or guess reasonably) an illustrative value for each. Name three nonquantized nuclear properties; give (or guess reasonably) illustrative values for two of them.

30. Suggest methods for preparing samples of high specific activity of (*a*) $C_6H_4Cl_2$ containing radiochlorine; (*b*) $CH_2BrCOOH$ containing radiobromine; (*c*) $C_6H_4INO_2$ containing radioiodine.

31. What is meant by each of the following: (*a*) the plateau of a Geiger–Müller counter; (*b*) a proportional counter; (*c*) the observed threshold of a nuclear reaction; (*d*) the standard deviation of a set of numbers; (*e*) the cross section of a nuclear reaction.

32. Sketch and briefly describe the essential features of one of the following: (*a*) Lauritsen electroscope; (*b*) Geiger–Müller counter.

33. If M is the average counting rate per minute of a given sample, what is the probability of obtaining m counts per minute in a particular observation: (*a*) according to the Poisson distribution law? (*b*) according to the Gaussian distribution law? (*c*) Under what assumptions is the Gaussian distribution a good approximation to the Poisson distribution?

34. If a cyclotron is tuned to accelerate deuterons to 10 Mev, what must be done to the cyclotron (other than building a new one) so that it can accelerate: (*a*) α particles, (*b*) protons? What will be the energy of the α particles and protons? Explain.

35. For what reasons do we believe that nuclei consist of protons and neutrons, rather than protons and electrons?

From this point on the questions are of the "open-book" type.

36. A counting rate is very nearly 800 counts per min. How long must it be counted to give an answer with a probable error of 0.3 per cent? If this is a total rate for sample plus background, and the background is very nearly 100 counts per min as determined in a 10-min measurement, what is the percentage probable error in the net sample rate?

37. The fission of a uranium nucleus yields about 200 Mev. This is what fraction of the total energy content of that nucleus?

38. The energy of the sun is believed to result from a reaction which produces helium from hydrogen (that is, from protons and electrons). How many ergs result per gram of helium produced?

39. On a classical picture, what is the lowest-energy α particle (in million electron volts) that U^{238} can emit?

40. Write several nuclear reactions that could be used to produce $_{24}Cr^{51}$. Emphasize those that would be most practical with an average cyclotron. Outline how it could be made using a 0.8-Mev voltage multiplier set.

41. Define the cross section for a nuclear reaction by means of a suitable differential equation (explain the meaning of each term of your notation). Use this equation to calculate the intensity of a neutron beam emerging after passing through a 0.1-mm-thick cadmium foil. The intensity of the original beam, normal to the surface of the foil, is 1000 neutrons per sec per cm^2. The average cross section for capture of these neutrons is 2500 barns.

42. How might you measure the half-life of thorium? Make a practical answer; be specific as to sample sizes, types of instruments, etc.

43. The activity of a sample was measured as a function of time on a Lauritsen electroscope:

Time (hr)	1	2	3	4	5	6	8	10	12
Div. $min.^{-1}$	7.1	5.7	4.5	3.7	3.0	2.4	1.7	1.2	0.90

Time (hr)	15	18	21	24	27	30	35	40
Div. $min.^{-1}$	0.63	0.48	0.39	0.32	0.27	0.22	0.175	0.137

Several absorption curves were taken at different times with aluminum absorbers; all were similar in appearance. A typical one:

Absorber (mg cm^{-2})	0	70	130	200	300	400	500	600	700	800
Div. $min.^{-1}$	5.8	3.5	2.2	1.3	0.6	0.28	0.12	0.11	0.11	0.10

Give the half-lives of the activities obviously present, and the upper energy limit of the β spectrum. What type of decay might be suggested?

44. (*a*) A disk of iodine, area = 2 cm^2, weight = 1 mg, was exposed to a beam of thermal neutrons incident perpendicularly on the face of the disk. The beam intensity was 5×10^7 neutrons per cm^2 per sec. The irradiation was from 1:00 P.M. to 1:50 P.M. The activity of the I^{128} produced was measured by placing the sample directly underneath and very close to the large aluminum leaf window of an "infinitely" large air-filled ionization chamber. The saturation ionization current in the chamber was 4×10^{-12} ampere at 2:15 P.M. Estimate the cross section of the (n, γ) reaction on I^{127} for thermal neutrons. (*b*) Discuss briefly the effect on the measured ionization current of varying the thickness t of the iodine sample, its area remaining 2 cm^2.

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