# ARTIFICIAL RADIOACTIVITY

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#### *Received June S, 1940*

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#### I. INTRODUCTION

Artificial radioactivity was discovered by Curie and Joliot early in 1934 (C30). They made the exciting and important observation that the positrons which were emitted when aluminum was bombarded with alpha-particles from polonium continued to be emitted after the source of alpha-particles was removed. The intensity of positrons decayed exponentially with a half-life of about 3 min. Immediately following the discovery of Curie and Joliot, many more artificial radioactivities were produced by several groups of workers, mainly as the result of bombardment with high-speed charged particles from artificial sources. Many substances were rendered radioactive by bombardment with protons and deuterons which were accelerated in various types of high-voltage apparatus; Cockcroft, Gilbert, and Walton (C31) employed a voltage-multiplier circuit, Lawrence and coworkers (H32) accelerated the charged projectiles in the magnetic resonance accelerator or "cyclotron" (L33), Crane and Lauritsen (C32) used a "cascade transformer" potential source, and Tuve and Hafstad (H33) an electrostatic generator. A further large addition to the list of radioactivities was made during 1934 and 1935 as a result of the work of Fermi and his associates (Al), who produced the activities by the neutron bombardment of the elements. Since the Coulomb field of the nucleus does not oppose the entry of a neutron, as it does that of a charged particle, a large number of elements throughout the entire range of the Periodic Table were rendered radioactive in this manner.

By the end of 1935 about one hundred artificial radioelements were known, and a review of the subject of artificial radioactivity, which included a discussion of the methods of production, was published by Ridenour and Yost in *Chemical Reviews* (R18). In the meantime, further work with deuterons, neutrons, and protons, and in addition, high-energy alpha-particles and gamma-rays from artificial sources, made a large increase in the number of induced radioactivities. A number of tables of radioactivities have appeared in various physical journals (L34, D15, GlO). Livingood and Seaborg have published, in the *Reviews of Modern Physics* (January, 1940), a table which describes the properties of three hundred and thirty artificial radioactivities (L35). These reviews in the physical journals merely listed the radioactivities and their properties and methods of production; they were not concerned with the applications of the radioelements to scientific problems.

The large currents and high energies now available, especially with the cyclotron, have resulted in the discovery of radioactive isotopes of every element in the range of atomic numbers from 1 to 85, inclusive (as well as thorium and uranium). Of especial importance, because of their wide applicability to tracer problems, are the radioactive hydrogen of Alvarez and Cornog (A7) and the long-lived radioactive carbon recently discovered by Ruben and Kamen (R17). It is the purpose of this review to discuss the present situation in the field of artificial radioactivity, particularly with respect to its applications to chemistry. Sections II to V will be devoted to a discussion of the properties and methods of production of the artificial radioelements and will include a brief description of the methods used for the detection of the radiations. Section VI gives a complete table (table 2) of all the known artificial radioelements, including their half-lives, the properties of their radiation, the reactions by which they are produced, and references to the original literature which describe their production. Section VII discusses the application of these radioelements to chemistry. The applications to chemistry that have been made so far are just the

beginning of what is certain to become a very large and fruitful field of research.

#### II. GENERAL CONSIDERATIONS

The decay of the artificial radioelements follows, of course, the wellknown exponential law, just as for the natural radioactive bodies, and the decay rate for each element is described in terms of a half-life.<sup>1</sup> Likewise the growth of a radioactivity when an element is under constant bombardment follows the familiar growth law, that is, the fraction of the saturation 0.69i number of atoms formed in a time t is equal to  $1 - e^{-\frac{1}{T_1/2}}$ , where  $T_{1/2}$  is the half-life. After a time of activation long compared to the half-life of the substance, the activity reaches a saturation value which cannot be increased by further activation (see, e.g., R19, Chapter I).

Although the nomenclature for the artificial radioelements has not reached a standardized form, most of the investigators in the field have adopted the practice of using the contracted form "radioelement" rather than the more cumbersome term "radioactive element." For example, the well-known radioactivity of 14.8 hr. half-life induced in sodium, as the result of bombardment with either neutrons or deuterons, is often referred to as being due to *radiosodium* rather than the *radioactive isotope of sodium.*  The former, simpler nomenclature will be used in this review.

The radiations from the artificial radioelements are very similar in properties to those from the natural radioelements of the well-known uranium, actinium, and thorium families (R19). Single decay predominates, although numerous examples of chain decays, i.e., cases where the product of the first decay is also radioactive, are known in artificial radioactivity. The beta-particles emitted during the decay of artificial radioactivities have the same continuous type of distribution in energy, with a definite upper energy limit, which has long been known to exist for the natural radioelements.<sup>2</sup> The upper energy limit decreases as the halflife increases; this Sargent  $(S35)$  relation (log half-life  $\infty$  -log energy) holds only roughly over a range of mass values (B37) and must be modified when the nuclear spin change accompanying the disintegration is greater than zero  $(B37, F9)$ . Decay by alpha-particle  $(He^{++})$  emission, which occurs so often in natural radioactivity, is very uncommon in artificial radioactivity. While only negative beta-particles (electrons) are emitted

<sup>&</sup>lt;sup>1</sup> The half-life is the time required for one-half of the initial number of atoms to decay.

<sup>2</sup> It is this continuous distribution of beta-particle energy which has led to the introduction of the neutrino, a particle of small or zero mass, one-half unit of spin  $(1/2 \cdot h/2\pi)$  and no charge, in order to preserve the laws of conservation of energy and momentum in beta-decay (F9).

by natural radioelements, some of the artificial radioelements emit negative beta-particles and some of them emit positive beta-particles (positrons). Examples of radioelements which decay by the emission of both negative and positive beta-particles are known. Some of the decays are accompanied by gamma-rays (high-energy electromagnetic radiation) and some are not, just as in natural radioactivity, and these gamma-rays may be slightly or largely internally converted.<sup>3</sup> Gamma-rays ionize much less, and hence are much more penetrating, than beta-particles of similar energy.

Positron emitters may decay by the alternative process of orbital electron capture, a method of decay which, up to the present time, has been observed only with the artificial radioelements. It was suggested by Yukawa (Y3) and others, from considerations based upon the Fermi theory of betaray emission (F9), that an unstable nucleus might reach stability by the capture of an extranuclear electron. The first experimental observation of decay by " $K$ -electron capture" (so-called because by far the largest proportion of the orbital electrons captured by the nucleus come from the K-electron shell) was made by Alvarez  $(A4)$ , who found that an unstable gallium isotope of 83 hr. half-life decayed by this mechanism. Many examples of this type of decay are now known. Decay by  $K$ -electron capture may be unaccompanied by any detectable ionizing radiation except for the x-rays which must be emitted (since the vacant place in the inner shell must be filled with the emission of an x-ray or an Auger electron). However, many examples of  $K$ -electron capture are known where the resultant nucleus is left in an excited state which drops to the ground state with the emission of a gamma-ray or a line<sup>4</sup> of internal-conversion electrons or both. Some radioelements decay by both positron emission and *K*electron capture, some by  $K$ -electron capture alone, and some entirely by positron emission.

3 When a gamma-ray is "internally converted" or undergoes "internal conversion," it means that instead of the emission of a gamma-ray there is the ejection of an electron from the extranuclear structure of the same atom that contains the nucleus which is radiating, the kinetic energy of the ejected electron being equal to the difference between the energy of the gamma-ray and the binding energy of the electron.

4 The name "line of electrons" is often used to describe the *mono-energetic* electrons which are emitted from, for example, the K-electron shell, when gamma-rays undergo internal conversion. Some of the gamma-rays are converted in the *L-, M-,*  etc. electron shells. The term "line" arises from the fact that with an electron magnetic spectrograph these groups of mono-energetic conversion electrons appear as lines on a photographic plate in contrast to the beta-particles from a beta-ray emitter, whose continuous distribution in energy darkens the plate over an entire, very broad, energy range.

Another class of radioactive substances, not peculiar to artificial radioactivity but most thoroughly studied here, are "nuclear isomers." Each member of a pair of nuclear isomers has the same atomic number and the same atomic weight (isotopic isobars); they represent two different energy states, the upper and the ground state, of the same nuclear species, differing in energy content and degree of stability. A theory which Weizsacker (W21) has proposed in order to account for the existence of nuclear isomers ascribes the long lifetime of the upper, metastable state to a difference of several units of angular momentum between the metastable and ground state. This large spin difference forbids the transition from the upper to the lower state in a manner analogous to the forbidden transitions in optical spectra. Each isomer of a pair may be radioactive and decay by beta-emission with its own characteristic half-life, or the isomer corresponding to the upper energy state may be beta-active while that corresponding to the ground state may be stable. Some isomers are genetically related to each other; the upper state, rather than decaying by beta-particle emission to a neighboring isobar, decays by an *isomeric transition* to the ground state with the emission of a gamma-ray.<sup>5</sup> The first evidence for nuclear isomerism in artificial radioactivity was presented in 1935 by Kourtchatow and coworkers (K5), and in 1937 Snell (S9) and Bothe and Gentner (B28) simultaneously showed that an 18 min. period and a 4.4-hr. period must both be ascribed to a bromine isotope of atomic weight 80.

## III. METHODS OF CHEMICAL IDENTIFICATION AND CONCENTRATION

The chemical identification of artificial radioelements is based on the fact that isotopes are not appreciably separated by ordinary chemical reactions and the radioactive isotopes of a given element behave in the same manner as the stable isotopes of the element. The property of radioactivity does not influence the chemical behavior (except in the case of extremely strong activities, and then in the same way as an external source would do). Thus, after a transmutation has taken place leading to the formation of a new and unstable nucleus, the new atom has properties determined solely by its new atomic number and will behave chemically in all respects like its stable isotope or isotopes. Often only a few millions of atoms of the unstable transmutation product are formed. The behavior of an element at such a low concentration may be uncertain in many chemical procedures. For this reason, in order to establish the chemical identity of the transmu-

<sup>&</sup>lt;sup>5</sup> The present theoretical explanation for the phenomenon of nuclear isomerism leads to the prediction that the gamma-rays corresponding to an isomeric transition will undergo high internal conversion when the energy is small,—of the order of tens of kilovolts (H45.D20).

tation product, it is usually expedient and often absolutely necessary to add a small quantity of that element which is isotopic with the expected or suspected transmutation product. This added material is usually designated by the term "carrier." The carrier element is separated out chemically and in this manner one can establish a radioactivity, with a characteristic half-life, which is isotopic with the carrier element.

In many cases, when the radioelement is not isotopic with the element from which it is formed, the radioactivity can be concentrated in a very small amount of material by adding and separating out only a very small amount of carrier. Such a high ratio of activity to carrier material (high *specific activity)* is very desirable in many of the investigations which employ radioelements, especially in biological chemistry and physiological and biological studies. The *specific activity* may be defined as the ratio of the number of radioactive atoms to the total number of isotopic atoms with which the radioactive atoms are mixed.

When the radioactivity is isotopic with the element which is bombarded, the active isotope is necessarily mixed with a larger quantity of its inactive isotopes. However, it is sometimes possible to effect a separation of the radioactive isotope from the inactive isotopes and hence a concentration of the radioactive isotope in a small amount of material. Szilard and Chalmers (S28) were the first to show that radioactive iodine could be separated from ordinary iodine after irradiating with neutrons a nonionizing organic compound such as ethyl iodide. After the irradiation, a very small amount of free iodine was added to act as a carrier for the free radioactive iodine, and after this iodine was reduced and precipitated as silver iodide it was found to contain practically all of the radioactivity. This method of concentration, which has subsequently been used to concentrate a number of radioelements, is now known as the "Szilard-Chalmers method." Its success depends upon the removal of the newly formed radioactive nucleus from its chemical bond in the irradiated compound. The breaking of this bond is a result of the large amount of energy furnished by the recoil from the gamma-rays emitted during the neutroncapture process. The method is not limited to radioelements formed by irradiation with neutrons, but in principle can be applied to other methods of activation. It is, of course, essential that the radioactive atoms set free during the bombardment do not interchange with their isotopic atoms in the irradiated chemical compound. As examples of the application of this method, radioactive iodine, bromine, and chlorine can be concentrated, using either organic compounds or the inorganic halogenates. Similarly, manganese dioxide precipitated from an irradiated permanganate is found to carry most of the radiomanganese under certain conditions. Whenever it is desired to use this method in order to produce a radioactive

isotope with a high specific activity, a search is made for a compound which contains the element in a form which will not interchange with the freed radioactive atoms and from which the radioactive atoms will be liberated during and separable after the irradiation process.

Erbacher and Philipp (E8), Lu and Sugden (L42), and Roginsky and Gopstein (R21) have developed a number of excellent methods for the extraction with high yields of very concentrated radioactive halogens from neutron-irradiated organic halides. One of the techniques employed by Erbacher and Philipp (E8) depended upon the absorption of the radioactive atoms on active charcoal, and Roginsky and Gopstein (R21) have used aluminum oxide and active charcoal as absorbents. Majer (M25, M31) has used a trace of colloidal gold to supply condensation nuclei for the deposition of active gold atoms formed in a neutron-irradiated, alkaline gold chloride solution.

A method for the chemical separation of genetically related nuclear isomers, which is a modification of the Szilard-Chalmers method, has been invented by Segre, Halford, and Seaborg (SlO). The element which contains the radioactivity corresponding to an upper isomeric energy state is made into a compound suitable for the application of the Szilard-Chalmers method of concentration; the daughter radioactivity, which corresponds to the ground state and which is liberated from this compound as a result of the isomeric transition,<sup>6</sup> is then chemically separable from the parent radioactivity. Segre, Halford, and Seaborg (SlO) used this method to extract the bromine radioactivity of 18 min. half-life, in the form of hydrobromic acid, from its parent isomer of 4.4 hr. half-life, which was present as tertiary butyl bromide. DeVault and Libby (D12) made the same separation by precipitating silver bromide from an ammoniacal solution which contained the 4.4-hr. radioactivity in the form of the bromate, while Le Roux, Lu, and Sugden (L36) separated the 18-min. radioactivity as silver bromide from both ethylene dibromide and  $n$ -butyl bromide. Seaborg, Livingood, and Kennedy (S15) have applied this isomer separation technique to tellurium to extract three daughter radioactivities from their three parent radioactivities, and Langsdorf and Segre (L30) have separated a pair of isomers in selenium. A more complete discussion of the work which has been done on the Szilard-Chalmers and isomer separation methods appears in section VII.

In some cases when the transmutation product is not isotopic with the target element, it is possible to separate it from the target element without the use of carrier. This gives the pure radioactive element, or compound

<sup>6</sup> The mechanism of the bond-rupture process, which seems to depend upon internal conversion of the transition gamma-ray, has been investigated in some detail and will be discussed in section VII.

of the element, in much too small an amount to be seen, detectable only by its radioactivity. A few examples will serve to illustrate the type of physical and chemical properties which may serve to make such a separation feasible.

Haissinsky (H34) separated pure radiocopper, produced by neutron bombardment of zinc, by means of electrochemical deposition on lead, while Steigman (S29) effected the same separation by electrolysis. Segre (S30) found that the radiosodium present in a sample of magnesium hydroxide, after deuteron bombardment of the magnesium, could be dissolved out of the hydroxide quantitatively by treating it with water.

Partition between solvents affords another method for separating the radioactive isotope in its pure form in the absence of carrier material. Grahame and Seaborg (GlI) have used the partition between ether and 6 *N* hydrochloric acid to separate pure radiogallium from zinc as well as radiomanganese and radiocobalt from iron.

When there exists a large difference in the boiling points, this may be used to effect a separation. For example, a gaseous, radioactive transmutation product can be easily separated from a non-gaseous target element or parent element. Alvarez, Helmholz, and Nelson (A12) have separated practically pure radiocadmium by collecting the vapor after heating deuteron-activated silver to its melting point.

When the radioactive transmutation product forms an extremely insoluble compound, it should be possible to collect the invisible precipitate on the walls of the containing vessel after centrifugation, as has been done for many of the naturally radioactive elements (H52, C36, H39).

Another method of separation depends on the fact that the newly formed radioactive atom may have lost one or more of its extranuclear electrons at the moment of formation as a result of the recoil given to it by the gamma-ray emitted during the capture process. The charged atoms formed in this manner may be collected, during the bombardment of either a gaseous or a liquid substance, with the aid of an electric field. This method of concentration, which is sometimes applicable even when the radioactive product is isotopic with the target element, has been employed by Fermi and his associates  $(A1)$  and by Paneth and Fay  $(P11)$ and Govaerts  $(G15)$ . The collection of recoil radioactive atoms produced during bombardment with charged particles can also be used as a method for obtaining concentrated radioactive samples (S3).

Erbacher (ElO) and Maier-Leibnitz (M30) have devised means to extract radioactive P<sup>32</sup> of very high specific activity from carbon disulfide which has been bombarded with fast neutrons.

When the transmutation product is isotopic with an element of which no stable isotopes have been found in nature or which is very rare and not available for use as carrier for the radioactive isotopes, the matter of

chemical separation becomes more difficult. For example, Perrier and Segre (P12) found that the deuteron bombardment of molybdenum produced radioactive isotopes of element 43. Since no stable element 43 is available, the chemical identification was carried out with the aid of rhenium, an element in the same group of the Periodic Table, which was expected to have very similar chemical properties to element 43. Similarly, Corson, MacKenzie, and Segre (C46, C23) were able, with the aid of iodine as a carrier, to identify as element 85 a radioactive transmutation product produced from bismuth plus 32-Mev. alpha-particles. McMillan and Abelson (M28) were able to show by means of a series of chemical experiments that one of the radioactivities formed when uranium is bombarded with neutrons must be ascribed to element 93.

#### IV. METHODS FOR DETECTION OF RADIATIONS

There are a number of different types of instruments that are used for the measurement of the intensity of the radiations from radioactive substances (L25). No attempt will be made to describe all of them, but some of those which are being used at present by workers in this field will be mentioned briefly. The best type of instrument for a given problem depends upon the type and energy of the radiation and upon the sensitivity and stability which is required. A knowledge of the absorption curve for the radiation to be measured is often very important, especially if the radiation is of very low energy (i.e., very soft), since this will make it necessary to pay particular attention to the effect of self-absorption in the sample and to use a detection device of the proper design.

One of the simplest instruments, widely used, is the Lauritsen modification of the electroscope, known as the "quartz-fiber electroscope" (L44). The electroscope is usually used inside an ionization chamber, which may be filled with any gas, although air is usually used for convenience, and the rate of drift of the fiber across a scale is measured. This instrument, which is suitable for the detection of all types of radiations from radioactive substances, is one compact unit and requires for its operation only a D.c. potential source of the order of 200 volts. Its sensitivity is not as large as that of some of the other detection devices. The quartz-fiber electroscope can be used to best advantage with a radioactivity whose intensity is of the order of microcuries  $(10^{-6}$  curies<sup>7</sup>), although intensities which are 1 per cent, or even 0.1 per cent, as strong as this can be measured.

A more sensitive arrangement of the integrating type, also suitable for

<sup>&</sup>lt;sup>7</sup> The "curie," originally the name for the amount of radon in equilibrium with 1 g. of radium, is now usually taken as the name for the general unit of intensity in radioactivity; a curie of any radioactive material undergoes the same number of disintegrations per unit time as 1 g. of radium, namely,  $3.7 \times 10^{10}$  disintegrations per second.

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the detection of all types of radiation from radioactive substances, is obtained with an ionization chamber connected to some kind of an electrometer. Segre (S44), Amaldi and Fermi (A13), Barnes (B29), and Montgomery and Montgomery (M33) have given descriptions of the construction of various types of ionization chambers. These chambers, which usually consist of an outer cylinder with a central, coaxial electrode, require 300 to 400 volts of D.C. potential for their operation and may be designed so as to contain any gas. The gas may be at atmospheric pressure, which is necessary when a thin window is used, or higher pressures may be used to increase the detection sensitivity for penetrating radiation such as gamma-rays. The measurement of extremely soft radiation can be accomplished by introducing the sample into the inside of the ionization chamber. Any sensitive electrometer can be used; both the Edelmann electrometer and the Perucca electrometer have been used in practice and found to be particularly satisfactory. Various vacuum-tube electrometer systems have been described by Hafstad (H35), DuBridge and Brown  $(D16)$ , and Barth  $(B30)$ . The ionization current is amplified by a singletube D.C. amplifier and is read as a deflection on a sensitive galvanometer; a modification in which the "rate of drift" of the galvanometer current is read increases the sensitivity for the measurement of the radiations from read increases the sensitivity for the measurement of the radiations from<br>weak samples (intensities of the order of 10<sup>-4</sup> microcuries can be measured).

An ionization chamber of a different design can be used to detect single, heavily ionizing particles, such as protons, alpha-particles, etc., when connected to a high-gain, multiple-stage, A.C. (pulse) amplifier as described by Dunning (D17). Electrons and gamma-rays (which show themselves only by the electrons liberated from the material they pass through) are not recorded with this arrangement.

The most sensitive detection device is the Geiger counter (G12), also known as the "point" counter, an instrument which detects individual ionizing particles of all kinds. A modification of the original Geiger counter which is now widely used is the Geiger-Miiller counter (G13), which is also known as the "tube" counter and the "Zählrohr." With this instrument it is possible to detect the ionizing particles from a source as weak as 10-5 microcuries. The Geiger-Miiller counter consists of a cylinder and a coaxial wire which is insulated from the cylinder. It is filled with some gas such as air or argon at reduced pressure and is operated with a negative voltage of 500 to 5000 volts on the outer cylinder. The passage of an ionizing particle through the counter causes a temporary electrical breakdown between the outer cylinder and the central wire, and this pulse is usually amplified by an A.C. (pulse) amplifier to a stage where it is capable of operating a mechanical recorder in a recording circuit. In spite of the vast amount of research which has been done, it is felt by many

that the construction and operation of the Geiger-Miiller counter is still somewhat more of an art than a science. Neher (S31) gives an excellent description of the construction of several types of counters and of various vacuum-tube circuits for their operation. Hamblin and Johnson (H53) have also given a good discussion of counters and counting apparatus. Olson, Libby, Long, and Halford (03) and Bale, Haven, and LeFevre (B47) have described Geiger-Miiller counter arrangements which can be used to measure the radiations from radioactive substances in solution.

When the particle radiation from the radioactive element is of very low energy, the radioactive material, in the form of a gas, can be introduced into the interior of an ordinary ionization chamber (L30) or Geiger-Miiller counter (R17, S33). An arrangement which allows a radioactive sample of solid material to be introduced into the interior of a counter, and which allows background counts to be made while such a sample is inside, is the "screen-wall" counter described by Libby (L50) and Libby and Lee (L51). Of course, when the low-energy particle radiation is accompanied by gamma-radiation it may not be necessary to introduce the sample into the interior of the counter or ionization chamber. However, the lower efficiency of gamma-ray detection makes it necessary to have strong samples for measurements which depend entirely upon gamma-radiation.

The "proportional" counter, first described by Geiger and Klemperer  $(G14)$ , is a modification of the Geiger and the Geiger-Müller counter which is operated at a lower voltage (and with a higher gain pulse amplifier), so that only heavily ionizing particles such as protons, alpha-particles, etc., are detected. A description of this type of counter arrangement, which does not respond to electrons and gamma-rays, has been given by Brubaker and Pollard (B31).

For most work involving the use of the radioelements as indicators the choice of detection device will be made from the following group:  $(1)$ electroscope, *{2)* Geiger-Miiller counter, or (S) integrating ionization chamber.<sup>8</sup> These instruments can be used to obtain absorption curves

<sup>8</sup> Only a very rough comparison of the costs of the above instruments can be made. An ordinary Lauritsen electroscope can be purchased as a single unit (excluding batteries) for about forty-five dollars from the F. C. Hensen Company, 3311 E. Colorado Street, Pasadena, California. Geiger-Miiller counter equipment is also available commercially. Geiger-Miiller counter tubes, of various designs, can be purchased for twenty-five to thirty dollars each from the W. M. Welch Scientific Company, 1515 Sedgwick Street, Chicago, Illinois, and at a price of about ten to twenty dollars each from Herbach and Rademan, Inc., 522 Market Street, Philadelphia, Pennsylvania. The latter company also sells all the assembled auxiliary equipment necessary for the operation of a Geiger-Miiller counter, at a price ranging from one hundred to one hundred fifty dollars. Although the construction cost of an

in order to determine the energy of radiations from radioactive substances. The upper energy limits of beta-particles are usually determined with the help of the relationship of Feather (F13),  $R = 0.543E - 0.160$ , where R is the range in grams per  $cm<sup>2</sup>$  of aluminum and  $E$  is the energy in Mev. (good only for  $E > 0.6$  Mev.), while for gamma-rays the correlation of energy with absorption coefficient as listed in the table compiled by Gentner (G17) can be conveniently used.

Brief mention should also be made of other experimental arrangements which are used primarily for detailed studies of the properties and energy of radiations. The spectrograph or spectrometer  $(E11, L54)$  uses a magnetic or electric field to bend, to an extent dependent upon their energy, the beta-particles, the internal-conversion electrons, or the secondary electrons from gamma-rays. The particles are detected either by a photographic method or by a counter or ionization chamber. Another arrangement involves two or more counters connected to an amplifier of a type which records the counts only when the counters discharge simultaneously (B38). This "coincidence counter" arrangement is often used to determine the energy of gamma-rays by determining the absorption curve of the secondary electrons which are producing the coincidence counts. A complete discussion of the theory and practice of coincidence counting has been given by Dunworth (D23). Finally, there is the expansion chamber or cloud chamber of Wilson (W17) (usually used with a magnetic field), in which can be seen and photographed the water drops which condense along the path of an ionizing particle.

Another instrument which should be mentioned is the secondary electron multiplier tube as adapted to counting purposes by Allen (A15). This device, which detects single positive ions, electrons, and photons by producing a large current of secondary electrons inside a single vacuum tube, is most useful for special problems in physics where work in a vacuum and an extremely low background (< one count per minute) are paramount factors.

# V. TYPES OP REACTIONS AND METHODS OF PRODUCTION

Table 1 presents a list of the known stable isotopes together with their relative abundances.<sup>9</sup> (The more abundant natural radioelements, marked with the sign  $\dagger$ , are included.) All of the artificial radioelements

integrating ionization chamber will depend largely on the facilities available, the total price of such a chamber and good auxiliary equipment will probably amount to two hundred dollars or more.

9 This table is taken from the article of Livingood and Seaborg (L35). Attention is called to the very complete review article on the stable isotopes recently published by Hahn, Flügge, and Mattauch (H36). This includes exact mass and packing fraction values obtained from both mass spectrographic and transmutation data.

z	<b>ELEMENT</b>	$\boldsymbol{A}$	PER CENT ABUNDANCE	z	<b>ELEMENT</b>	$\boldsymbol{A}$	PER CENT ABUNDANCE
1	Η	1	99.98	16	S	32	95.0
		$\overline{2}$	0.02			33	0.74
						34	4.2
$\boldsymbol{2}$	He	3	$\sim$ 10 <sup>-5</sup>			$36\,$	0.016
		$\overline{4}$	100				
				17	Cl	35	75.4
$\bf 3$	Li	6	7.9			37	24.6
		7	92.1				
				18	A	36	0.307
$\overline{\mathbf{4}}$	Be	9	100			38	0.061
						40	99.632
5	в	10	18.4				
		11	81.6	19	Κ	39	93.3
						40+	0.012
6	$\rm ^{c}$	12	98.9			41	6.7
		13					
			1.1	20	Ca	40	96.96
						42	0.64
$\overline{7}$	${\bf N}$	14	99.62			43	0.15
		15	0.38			44	2.06
						46	0.0033
8	O	16	99.76			48	0.19
		17	0.04				
		18	0.20	21	$_{\rm Sc}$	45	100
9	F	19	100	22	Ti	46	7.95
						47	7.75
10	Ne	20	90.00			48	73.45
		21	0.27			49	5.51
		22	9.73			50	5.34
11	$N_{2}$	23	100	23	v	51	100
12	$_{\rm Mg}$	24	77.4	24	$_{\rm Cr}$	50	4.49
		25	11.5			52	83.77
		26	11.1			53	9.43
						54	2.30
13	Al	27	100				
				25	Mn	55	100
14	Si	28	89.6				
		29	$6.2\,$	26	Fe	54	6.04
		30	4.2			56	91.57
						$57\,$	2.11
15	$\mathbf P$	31	100			58	0.28

TABLE 1 *Stable isotopes of the elements* 

f Natural radioactivity.

 $\overline{\phantom{0}}$ 

z	<b>ELEMENT</b>	$\boldsymbol{A}$	PER CENT <b>ABUNDANCE</b>	Z	<b>ELEMENT</b>	$\boldsymbol{A}$	PER CENT ABUNDANCE
27	$\rm{Co}$	57	0.17	37	Rb	85	72.3
		59	99.83			87 <sup>†</sup>	27.7
28	Ni	58	68.0	38	$_{\rm Sr}$	84	0.56
		60	$27\,.2$			86	9.86
		61	0.1			87	7.02
		62	3.8			88	82.56
		64	0.9				
				39	Y	89	100
29	Cu	63	68	40			
		65	32		Zr	90	48
						91	11.5
30	Zn	64	50.9			92	22
		66	27.3			94 96	17
		67	3.9				1.5
		68	17.4	41	Сb	93	
		70	0.5				100
				42	Mo	92	15.5
31	Ga	69	61.2 38.8			94	8.7
		71				95	16.3
						96	16.8
32	Ge	70	21.2			97	8.7
		72	27.3			98	25.4
		73	7.9			100	8.6
		74	37.1				
		76	6.5	44	Ru	96	5 p.
						98	
33	As	75	100			99	12
						100	14 22
34	$\rm Se$	74	0.9			101 102	30
		76	9.5			104	17
		77 78	8.3				
		80	24.0 48.0	45	Rh	101	0.08
		82	9.3			103	99.92
35	Br	79	50.6	46	P <sub>d</sub>	102	0.8
		81	49.4			104	9.3
						105	22.6
36	Kr	78	0.35			106	27.2
		80	2.01			108	26.8
		82	11.53			110	13.5
		83	11.53				
		84	57.10	47	Ag	107	52.5
		86	17.47			109	47.5

TABLE *!—Continued* 

t Natural radioactivity.

z	<b>ELEMENT</b>	$\boldsymbol{A}$	PER CENT <b>ABUNDANCE</b>	z	ELEMENT	$\boldsymbol{A}$	PER CENT ABUNDANCE
48	$_{\mathrm{Cd}}$	106	1.4	55	$\rm Cs$	133	100
		108	1.0				
		110	12.8	56	Ba	130	0.101
		111	13.0			132	0.097
		112	24.2			134	2.42
		113	12.3			135	6.59
		114	28.0			136	7.81
		116	7.3			137	11.32
						138	71.66
49	In	113	4.5				
		115	95.5	57	La	139	100
50	$_{\rm Sn}$	112	1.1	58	Ce	136	$<$ 1
		114	0.8			138	$<$ 1
		115	0.4			140	90
		116	15.5			142	10
		117	9.1				
		118	22.5	59	Pr	141	100
		119	9.8				
		120	28.5	60	$\rm Nd$	142	25.95
		122	5.5			143	13.0
		124	6.8			144	22.6
						145	9.2
51	$_{\rm Sb}$	121	56			146	16.5
		123	44			148	6.8
						150	5.95
52	Te	120 122	< 0.1				
		123	2.9	62	Sm	144	3
		124	1.6			147	17
		125	4.5			148†	14
		126	6.0			149	15
		128	19.0			150	5
		130	32.8			152	26
			33.1			154	20
53	I	127	100				
				63	Eu	151	49.1
54	X <sub>e</sub>	124	0.094			153	50.9
		126	0.088				
		128	1.90	64	Gd	152	$0.2\,$
		129	26.23			154	1.5
		130	4.07			155	20.7
		131	21.17			156	22.6
		132	26.96			157	16.7
		134	10.54			158	22.6
		136	8.95			160	15.7

TABLE *!—Continued* 

t Natural radioactivity.

 $\bullet$ 

Z	<b>ELEMENT</b>	$\boldsymbol{A}$	PER CENT ABUNDANCE	z	<b>ELEMENT</b>	$\boldsymbol{A}$	PER CENT ABUNDANCE
65	Tb	159	100	75	$_{\rm Re}$	185	38.2
						187	61.8
66	$\mathbf{D}\mathbf{y}$	158	0.1				
		160	1.5	76	Os	184	0.018
		161	21.6			186	1.59
		162	24.6			187	1.64
		163	24.6			188	13.3
		164	27.6			189	16.1
						190	26.4
67	Ho	165	100			192	41.0
68	Er	162	0.25	77	Ir	191	38.5
		164	2.0			193	61.5
		166	35.2				
		167	23.5	78	Pt	192	0.8
		168	29.3			194	30.2
		170	9.8			195	35.3
						196	26.6
69	Tm	169	100			198	7.2
70	Yb	168	0.06	79	Αu	197	100
		170	2				
		171	8.8	80	Hg	196	0.15
		172	23.5			198	10.1
		173	16.7			199	17.0
		174	37.2			200	23.3
		176	11.8			201	13.2
						202	29.6
71	Lu	175	97.5			204	6.7
		176†	2.5				
				81	Tl	203	29.1
72	Hf	172?	${<}0.1$			205	70.9
		174	0.3				
		176	5	82	P <sub>b</sub>	204	1.48
		177	19			206	23.59
		178	28			207	22.64
		179	18			208	52.29
		180	30	83	Bi	209	100
73	Ta	181	100				
				90	Th	$232+$	100
74	w	180	$\sim 0.2$	91	Pa	231†	
		182	22.6				
		183	17.3	92	U	234†	0.006
		184	30.1			235†	0.71
		186	29.8			238†	99.28

TABLE *!—Concluded* 

t Natural radioactivity.

are prepared from these isotopes with the help of various kinds of bombarding particles, such as neutrons, deuterons, alpha-particles (helium ions), protons, high-energy gamma-rays, and x-rays. The artificial radioelements with mass numbers smaller than those of the stable isotopes of the same element reach stability by the emission of positrons (or by  $K$ -electron capture); those with larger mass numbers attain stability by the emission of negative beta-particles. A radioelement whose mass number lies between the mass numbers of two stable isotopes is usually a negative beta-particle emitter.

In order to identify completely a radioactivity it is necessary to establish the mass number of the active isotope as well as the atomic number, which is identified by the chemical separations. The mass number of the radioactive isotope can be deduced sometimes from a study of the known mass numbers of the stable isotopes of the target element after the chemical separation has established the type of reaction; often the identification must be made by the method of "cross bombardment," i.e., the preparation of the radioactive isotope by several independent nuclear reactions. The bombardment of separated isotopes should offer a powerful method for the isotopic identification of induced radioactivities, in view of the excellent isotope separation methods which have been recently developed by Urey and associates (H46), by Clusius and Dickel (C34), and by Beams and associates (B42). For example, Kennedy and Seaborg (K18) bombarded separated chlorine isotopes in order to make an isotopic assignment of a radiochlorine isotope. Isotope separation experiments performed after bombardment should also prove useful for isotopic identification. However, for the use of a radioactive substance in a chemical or biological problem a knowledge of the atomic number is sufficient, and radioactive isotopes whose mass numbers have not been established can be used.

When a radioactive isotope is formed as the result of a reaction which involves a rare isotope, the bombardment of the separated isotope will, of course, result in a larger yield. That this may be a matter of practical importance is strikingly shown in the case of the long-lived radiocarbon of Ruben and Kamen, which is produced by the deuteron-activation of carbon. Its long half-life and the fact that it is made from  $C^{13}$ , an isotope of 1 per cent abundance, leads, under the present methods of production, to rather low intensities. However, the bombardment of nearly pure  $C<sup>13</sup>$ , now available in good quantity as a result of the experiments of Urey and coworkers, makes it possible to increase the yield by a factor which may become nearly as large as 100.

The type reactions leading to the formation of the artificial radioelements will now be described (these reactions also often lead to the production of stable isotopes). For brevity it will be convenient to use a simplified notation, rather than to write out the entire reaction each time. For

example, the bombardment of iron with neutrons to produce radiomanganese will be described by  $\text{Fe}^{56}(n,p)$  Mn<sup>56</sup>, rather than by the more cumbersome equation

$$
_{26}\mathrm{Fe}^{56} + _{0}n^{1} \rightarrow _{25}\mathrm{Mn}^{56} + _{1}\mathrm{H}^{1}
$$

(where the superscripts denote mass numbers and the subscripts atomic numbers). The notation  $n =$  neutron,  $p =$  proton,  $\alpha =$  alpha-particle,  $d =$  deuteron, and  $\gamma =$  gamma-ray will be used.

The discussion which follows will also include a few statements regarding the yield of radioelements formed in the various reactions. It must be emphasized that only rough qualitative statements can be made, since the situation is too complex to allow a quantitative treatment in a few words. No summary of experimental data on the various reaction yields and the variation of the yields with energy and atomic number has been published; a summary of this type awaits more systematic data. Weisskopf and Ewing (W25) have recently published an excellent theoretical treatment of the yields from neutron, proton, and alpha-particle reactions.

It may be helpful to point out that the considerations of Bohr (B32, B33) have led to the view that the transmutation which occurs as the result of the impact of any particle with an atomic nucleus (with a few exceptions) proceeds by a mechanism which must be treated as two independent processes: namely, *(1)* the amalgamation of the particle with the nucleus to form an intermediate metastable compound nucleus, and *(2)* the eventual breaking up of the intermediate nucleus into the end products.

## *1. Neutron reactions*

Neutrons are obtained from two types of sources,—*(1)* artificial and *(£)*  those which utilize the radiations from the natural radioelements. The neutrons from the latter type are usually produced by the reaction  $Be^9$  $(\alpha, n)$  C<sup>12</sup> and the sources may be prepared by mixing powdered beryllium with alpha-particle emitters such as radium, radon, or polonium. Neutrons produced in this manner have a more or less continuous distribution in energy extending up to about 13 Mev. (million electron volts<sup>10</sup>) for radon alpha-particles (D21, B39). The action of mono-energetic gamma-rays (of sufficient energy) on beryllium and on deuterium gives rise to monoenergetic neutrons, usually known as "photo-neutrons," with energies of the order of hundreds of kilovolts when gamma-rays from the natural radioelements are used.

Artificial sources of neutrons are obtained by bombarding various ele-

10 The electron volt, a unit of energy widely used in atomic and nuclear physics, is equal to  $1.59 \times 10^{-12}$  ergs (the amount of energy acquired by a particle of electronic charge when it falls through a potential difference of 1 volt).

ments with fast-moving charged particles, the energy of the neutrons depending upon the energy of the bombarding particles. However, the neutron energies have a very strong dependence upon the reactions used for their production, so that, taking into account the experimental conditions being used at present for their production, certain very rough statements can be made with regard to the energies of neutrons from various artificial sources. Two common sources are *(1)* deuterons (say 5 to 10 Mev.) on beryllium (the  $Be + D$  source), giving neutrons with energies extending up to about 13 Mev., and  $(2)$  deuterons on deuterium (the  $D-D$ source). The  $D-D$  source is used to most advantage when deuterons with energy of the order of a few hundred kilovolts are available, and under these conditions gives rise to neutrons within the narrow energy range of 2.5 to 3 Mev. The bombardment of lithium or boron with deuterons produces neutrons with energies extending up to about 20 Mev. Neutrons in this high energy range are produced also by the bombardment of beryllium with very high energy (e.g., 16 Mev.) deuterons.

Common usage has evolved a *rough* classification of neutrons into groups on the basis of their energy. "Very fast" neutrons, often called "fast" neutrons, are those, e.g., from a lithium plus deuterons  $(Li + D)$  or boron plus deuterons  $(B + D)$  source, with energies extending from about 20 Mev. to about 10 Mev., while neutrons with energies within the range starting at about 10 Mev. and extending down to an indefinite energy region (of the order of thousands of electron volts) are known as "fast" neutrons or "medium fast" neutrons (especially those at the lower end of this energy range). "Slow" or "thermal" neutrons is the name given to those neutrons which have suffered a sufficient number of collisions, usually with hydrogen nuclei as the result of passage through paraffin or water, to slow them to thermal velocities, that is, about 0.025 electron volt of energy. Slow neutrons were discovered by Fermi and coworkers (Al), and these authors give a discussion of their properties and of the slowing process. Neutrons in the energy range immediately above the thermal range are sometimes designated as "resonance" neutrons, a result of the fact that many nuclei absorb such neutrons only within extremely narrow energy ranges, that is, absorb them in a resonance fashion.

Reactions between neutrons and nuclei produce artificial radioelements by four main types of transmutations: *(1)* the simple, radiative capture, known as the  $n, \gamma$  reaction; (2) neutron capture followed by proton emission, or the *n,p* reaction; (3) neutron capture with alpha-particle emission, the  $n, \alpha$  reaction; and (4) neutron capture followed by the emission of two neutrons (net expulsion of one neutron), or the *n,2n* reaction. The *n,y*  and *n,2n* reactions give radioelements which are isotopic with the target element, and hence the Szilard-Chalmers method of concentration is used

when it is desired to obtain a high ratio of activity to inactive material, that is, a high *specific activity*. When the  $n, p$  or  $n, \alpha$  reactions are used or the Szilard-Chalmers method employed, large amounts of material can be used effectively in order to obtain large specific activities. Radioelements formed in the *n,2n* reaction are largely positron emitters, while the other reactions usually lead to negative beta-particle emitters.

The  $n, \gamma$  type of activation occurs largely with slow neutrons and only to a smaller extent with fast neutrons. Elements throughout the entire range of the Periodic Table can be activated in this manner; the cross sections for slow neutron absorption, which are extremely large for some elements, vary in an irregular manner from element to element and from isotope to isotope. The  $n, p$  and  $n, \alpha$  transmutations require fast neutrons (except for two or three cases in the lightest elements). The energy required increases regularly as one proceeds up the Periodic Table, since the outgoing charged particles must escape the nuclear potential barrier, and for atomic numbers as high as 50 only the "very fast" neutrons are effective. The *n,2n* reaction requires "very fast" neutrons because the net result is the expulsion of a neutron, whose binding energy amounts to about 8 Mev. for most of the elements.

There is another type of activation by fast neutrons which involves those isomers in which the ground state is stable. The kinetic energy of the captured neutron excites the nucleus to its upper, radioactive, isomeric state, a neutron being reemitted, with reduced energy, after the excitation process. In keeping with our method of writing nuclear reactions, this method of excitation is known as an *n,n* process and a typical example is written In<sup>115</sup>  $(n,n)$  In<sup>115\*</sup>. (The asterisk, as used here, denotes a radioactive isomer of a stable nucleus.)

## *2. Deuteron reactions*

The most intense radioactivities are, in general, induced as the result of bombardments with high-energy deuterons which are produced in the "cyclotron" of Lawrence and Livingston (L37, L33). Most of the cyclotrons which are now in operation are producing deuterons of 5- to 10-Mev. energy and currents of 10 to 200 microamperes; the 60-in. cyclotron at Berkeley is furnishing 16-Mev. deuterons at 100 to 200 microamperes (L38). Other types of artificial sources induce radioactivities of lower intensities, since they produce deuteron beams of much lower energy.

Artificial radioelements, both negative and positive beta-particle emitters, are produced by deuterons in the following ways: *(1)* deuteron capture and proton emission, known as the *d,p* reaction, which is, since the net result is the capture of a neutron, equivalent to the  $n, \gamma$  reaction; (2) deuteron capture followed by neutron emission, or *d,n* reaction; and *[S)* 

deuteron capture with alpha-particle emission, the  $d_{\alpha}$  reaction. The yields from all of these reactions increase with increasing energy of the deuterons and, for a given deuteron energy, decrease with increasing atomic number of the target element. This decrease is most marked for the  $d, \alpha$  reaction, since the outgoing, doubly charged alpha-particles must penetrate the Coulomb barrier of the nucleus (for example, with 8-Mev. deuterons this reaction is not observed for nuclei of atomic number as high as 50, while the *d,p* and *d,n* reactions are observed throughout the entire range of the Periodic Table). It should be pointed out that the *d,p* reaction occurs largely by a mechanism known as the Oppenheimer-Phillips process (07, V9), wherein the deuteron, upon approaching the nucleus, is polarized in such a manner as to give rise to the capture of the neutron without the usual amalgamation of the bombarding particle (deuteron) to form a temporary intermediate nucleus.

Radioelements are also produced by the *d,2n* transmutation, especially when the deuterons have an energy as high as 16 Mev. The  $d, \gamma$  and *d,d* reactions have not yet been established.

A few examples will serve to illustrate the intense radioactivities which are induced with deuterons. The bombardment of copper for 20 min. with 20 microamperes of 8-Mev. deuterons produces Cu<sup>64</sup> (half-life 12.8 hr.), by the reaction Cu<sup>63</sup>  $(d,p)$  Cu<sup>64</sup>, with an intensity of about 5 millicuries,  $(i.e., 5 \times 10^{-3}$  curies). Such a sample would give a discharge rate corresponding to about 107 times the natural background of an ordinary, Lauritsen, quartz-fiber electroscope. In a typical experiment a 4-hr. bombardment of phosphorus with 100 microamperes of 16-Mev. deuterons produced about 50 millicuries of  $P^{32}$  (half-life 14.3 days or 1.24  $\times$  10<sup>6</sup> sec.) by the reaction  $P^{31}$   $(d,p)$   $P^{32}$ . With the aid of the relation  $-dN/dt = \lambda N$ , where  $-dN/dt$  is the number of disintegrations per second and  $\lambda$  the disintegration constant<sup>11</sup>, we find for  $N$ , the number of active atoms,  $3.7 \times 10^{10} \times 0.05 \times 1.24 \times 10^{6}/0.69 = 1/3 \times 10^{16}$ . This corresponds to approximately one-sixth of a microgram of radioactive P32. It seems certain that it will soon be possible to produce weighable amounts of the very long-lived transmutation products.

### *3. Alpha-particle (helium ion) reactions*

Helium ions which are accelerated by electrical means are entirely equivalent to alpha-particles from the natural radioelements and therefore are often called alpha-particles. However, artificially accelerated helium ions, because of the larger intensity of particles available, have largely displaced the natural alpha-particles for the production of radioelements.

<sup>11</sup> The disintegration constant,  $\lambda$ , defined by the equation  $-dN/dT = \lambda N$ , is equal to 0.69 divided by the half-life.

For example, 100 mg, of radium (with its decay products) emits about  $10^{10}$ alpha-particles per second, spread out in all directions, while 1 microampere of alpha-particles corresponds to  $10^{13}$  particles per second, directed upon the target. When the cyclotron is in adjustment for deuterons it is also almost in adjustment for alpha-particles, since deuterons and helium ions have nearly the same value of *e/m,* and because the alpha-particles have the same velocity and twice the mass they attain twice the energy that deuterons do when they are accelerated with the same voltage. Most of the cyclotrons now in operation furnish alpha-particles of 10- to 16-Mev. energy. The 60-in. Berkeley cyclotron is producing 32-Mev. alpha-particles at currents of 10 to 20 microamperes.

Artificial radioelements, both negative and positive beta-particle emitters, are produced as the result of alpha-particle capture followed by neutron emission, the  $\alpha$ ,*n* reaction, and by alpha-particle capture and proton emission, the  $\alpha, p$  reaction. The yields increase with increasing energy of the projectile and, for a given energy, decrease with increasing atomic number. The  $\alpha$ ,n reaction occurs with elements throughout almost the entire Periodic Table when 16-Mev. alpha-particles are used, while the  $\alpha, p$  reaction is a very rare occurrence for elements of atomic number as high as 50.

The  $\alpha, \gamma$  reaction has not yet been observed, although it might be expected to occur at an energy below that of the  $\alpha, n$  threshold. The  $\alpha, 2n$ reaction has been shown to occur with 32-Mev. alpha-particles, and the calculations of Weisskopf and Ewing (W25) predict that this reaction should be important for particles of such high energy. The  $\alpha, pn$  (or  $\alpha$ ,*d*) reaction has also been observed at high energies.

The excitation of nuclei by a process which might be designated as an  $\alpha$ , $\alpha$  reaction is another method of activation which occurs with alphaparticles. Theoretical considerations of Weisskopf (W26) suggest, however, that this activation occurs as a result of an interaction between the electric fields of the alpha-particle and the nucleus and not as a capture and reemission of the bombarding particle as in the case of the  $n, n$  reaction.

#### *4- Proton reactions*

Just as in the case of deuterons and alpha-particles, the cyclotron offers the best source of high-energy protons. Many investigators, especially DuBridge and coworkers, have prepared a large number of radioelements by means of proton bombardments.

The most common reaction is the capture of the proton followed by neutron emission, or the *p,n* reaction, producing mainly elements which decay by positron emission (or K-electron capture). The yield increases with the energy of the proton and decreases with the atomic number of the target element. This transmutation is observed throughout the entire range of the Periodic Table when 6.5-Mev. protons are used. If the radioactive substance formed in a *p,n* reaction emits positrons and thus returns to the target element, the energy threshold for the reaction is equal to the difference between the neutron and hydrogen mass (0.8 Mev.) plus the mass of two electrons (1.0 Mev.) plus the upper energy limit of the positron spectrum from the radioactive substance. (Similarly the energy threshold for the formation of a positron emitter from a *d,2n* reaction is equal to 4.0 Mev. plus the positron upper energy limit.) When the radioactive product decays by  $K$ -electron capture, the energy threshold may be as much as 1 Mev. lower than that which would be calculated for the formation of an emitter of zero-energy positrons.

The radiative capture of the proton, known as the  $p, \gamma$  reaction, is observed with the very lightest elements, and it has also been observed with a few elements of medium weight  $(Z = 30)$ . This reaction is important only at energies below or near the threshold for the *p,n* reaction. The yield from this reaction, especially for the very lightest elements, exhibits sharp maxima at certain sharply defined energies of the protons corresponding to definite "resonance levels."

The  $p, \alpha$  reaction has been reported only rarely but will certainly be a common occurrence when protons of higher energy become the object of experimentation. Similarly, the *p,2n* reaction is to be anticipated at high energies. The utilization of the kinetic energy of protons to excite nuclei by the  $p, p$  reaction occurs, as in the case of the  $n, n$  and the  $\alpha, \alpha$  reactions.

#### *5. Gamma-ray reactions*

Gamma-rays of very high energy are capable of ejecting neutrons from atomic nuclei to produce radioelements by the  $\gamma$ , *n* reaction, a type of transmutation also known as "photo-disintegration." Bothe and Gentner (B20), who have studied this reaction using the 17-Mev. gamma-rays produced in the reaction of protons with lithium and the 12-Mev. gamma-rays from boron plus protons, found that the yield varied irregularly from element to element. Mainly positron emitters are formed and the yield is comparatively low. The reactions giving rise to gamma-rays of very high energy are of the resonance type, occurring at voltages below 1 Mev. and hence the direct acceleration type of apparatus, operating at high beam currents, is the best source.

High-energy x-rays have been used to excite certain stable nuclei to their isomeric, radioactive states. This type of excitation has also been effected by bombardment with high-energy electrons (C39).

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#### *6. Uranium and thorium fission*

In January, 1939, Hahn and Strassmann (H14) reported their very important discovery that the bombardment of uranium with slow or fast neutrons resulted in its cleavage into pairs of radioactive products of medium atomic weight. The existence of this entirely new type of nuclear reaction was immediately confirmed in many laboratories throughout the world, and Meitner and Frisch (M23), after confirming the reaction, suggested the name "fission" for the process. Subsequent work by a large number of investigators has resulted in the chemical identification of many of the fission products. Hahn and Strassmann (H15) and others found that thorium also undergoes nuclear fission when bombarded with fast neutrons, and v. Grosse, Booth, and Dunning (G7) found that the same is true for protoactinium (slow neutrons are ineffective in these cases). The products of these cleavage processes, because of the high neutron to proton ratio in uranium and thorium, have an abnormally high neutron to proton ratio; hence all are negative beta-particle emitters and many chains of successive decay are found. The radioactive isotopes formed in this manner, some of which can be formed in no other way and others of which can also be formed by some of the methods outlined in the sections above, have already found application to chemical and biological problems. Turner (T8) has published in the issue of *Reviews of Modern Physics* for January, 1940, an excellent, complete review of nuclear fission.

The events leading to the discovery of the fission process present an interesting history. In their original work Fermi and coworkers (Al) bombarded uranium with neutrons and obtained a series of radioactivities which, on the basis of chemical experiments, they were led to assign to "transuranic elements," that is, elements with atomic number greater than 92. The experiments of Hahn, Meitner, and Strassmann (H37) and others appeared to confirm this point of view, and for several years the transuranic elements were the subject of much experimental work and discussion, including a review of their chemical properties which was published in *Chemical Reviews* (Ql) in 1938. Curie and Savitch (C33), in 1938, found a product of 3.5 hr. half-life which had the chemical properties of a rare earth, but they failed to give a complete interpretation of this astonishing discovery. Early in 1939 Hahn and Strassmann (H14) described experiments which made it certain that they had observed the production of radioactive barium isotopes as the result of the bombardment of uranium with neutrons. Subsequent work has shown that practically all of the radioactivities formerly ascribed to transuranic elements are actually due to fission products. More than fifty radioactive fission products are now known and are included in table 2. The fission of uranium and thorium by deuterons (K26) and by high-energy gamma-rays (H59) has also been observed.

It has been shown by von Halban, Joliot, and Kowarski (H38), and confirmed by many others, that secondary neutrons are emitted during the fission of uranium. The secondary neutrons might themselves produce still more fissions and the possibility of the occurrence of a catastrophic chain reaction, under the proper conditions, has been the subject of much discussion and speculation. The large energy per fission  $(\sim 200$  Mev.) shows that the propagation of such a chain might involve the release of terrific amounts of energy in a very short time.

#### VI. TABLE OF ARTIFICIAL RADIOELEMENTS

Table 2 presents a complete list of all the artificial radioelements known to date (covering publications received prior to August 1, 1940), together with a number of important features associated with them. The natural radioactivities are not included. The plan of presentation is the same as that used by Livingood and Seaborg (L35).

The first and second columns give the atomic numbers and the mass numbers associated with the radioactivities. The degree of certainty of each assignment is indicated, in the column headed "class," with a letter according to the following code:

- $A =$  isotope certain (mass number and element certain),
- $B =$  isotope probable, element certain,
- $C =$  one of few isotopes, element certain,
- $D = element certain$ ,
- $E = element$  probable,
- $F =$  insufficient evidence,
- $G =$  probably in error (e.g., impurity or inadequate half-life determination).

The fourth column lists the type of radiation, with the following meaning for the symbols:

- $\beta^-$  = negative beta-particles,
- $\beta^+$  = positive beta-particles (positrons),
- $\gamma =$  gamma-rays,
- *e~ =* internal-conversion electrons,
- $K = K$ -electron capture,
- *I.T.* = isomeric transition (transition from upper to lower isomeric state).

In the few cases where it is certain that no gamma-rays are emitted, this

fact is expressed explicitly by the symbol "No  $\gamma$ ." Annihilation gammarays<sup>12</sup> are not listed.

The half-life, followed by the relevant reference, is given in the fifth column. For the case where more than one value for the half-life has been reported, an attempt has been made to list the best value (an experimental value near the mean or one determined with a strong sample).

In the column headed "energy of radiation," the energy value is followed by the corresponding reference and by a description of the method used for the energy determination. The beta-particle energies correspond to the observed upper limits of the spectra; in those cases where only the Konopinski-Uhlenbeck (K14) extrapolated value has been reported, this is listed, followed by the designation "K.U." The methods used for the determination of the energy of the particles are described in each case with the aid of the following symbols: abs.  $=$  absorption, cl. ch.  $=$  cloud chamber with magnetic field, spect.  $=$  electron magnetic spectrograph or spectrometer.

The symbols used to describe the methods employed for the determination of the gamma-ray energies have the following meaning: abs.  $=$  absorption, cl. ch. recoil  $=$  secondary electrons in cloud chamber with magnetic field, cl. ch. pair = positron-electron pairs in cloud chamber with magnetic field, coincid. abs. = secondary electrons with coincidence counters and absorber, spect. conv. = internal-conversion electrons with magnetic spectrograph, spect.  $=$  secondary electrons with magnetic spectrograph, and abs. of  $e^-$  = absorption of internal-conversion electrons. When internal-conversion electrons are emitted the energy listed is always that of the corresponding gamma-ray transition.

The observed nuclear reactions (giving the target element, projectile, and residue, in order) by which the radioactive isotopes are formed, and the corresponding references, are listed in the last column ( $p =$  proton,  $n =$  neutron,  $\alpha =$  alpha-particle,  $d =$  deuteron,  $\gamma =$  gamma-ray). The neutron-induced fission reactions of the heavy elements are included and are designated by such symbols as  $U-n$ , Th-n, and Pa-n. In those cases where the radioactive fission product is known to be the second (or later) element in a chain decay its production is not designated by these symbols  $(U-n, etc.)$  but is listed as produced by the beta-decay of its immediate parent isotope.

No attempt has been made to list all of the publications connected with a given radioactivity, since it has been the aim to keep the table as compact

<sup>12</sup> Positron emission is always accompanied by "annihilation" gamma-radiation. Each positron, together with an electron (of equal mass), eventually undergoes annihilation with the emission of two gamma-rays, each with an energy (0.51 Mev.) corresponding to mc<sup>2</sup>, where m is the electronic mass.

# TABLE 2



# *Complete list of induced radioactivities*  (The literature has been covered up to July 15, 1940)

RADIOELEMENT			TYPE OF <b>RADIATION</b>	<b>HALF-LIFE</b>		ENERGY OF RADIATION IN MEV.	PRODUCED BY	
z	$\boldsymbol{A}$	CLA <sub>88</sub>			Particles	$\gamma$ -Rays		
8	O <sub>15</sub>	A	$\beta^+$	126 sec. (M3, B20)	$1.7$ (F1) cl. ch.		$N-d-n$ (M3, F1) $0 - \gamma - n$ (B20) $O-n-2n$ (P2) $N-p-\gamma$ (D2) $C$ - $\alpha$ -n (K3)	
	O19	A	β-	31 sec. (N1)			$F - n - p$ (N1, A1)	
9	F17	A	$\beta^+$	70 sec. $(N2)$	$2.1$ (K4) cl. ch.		$0-d-n$ (N2, F1) $N-\alpha$ -n (R3) $0-p-\gamma$ (D2)	
	F13	A	$B^+$	112 min. (S1)	0.7(T2) ci. ch.		Ne-d- $\alpha$ (S1) $O-p-n$ (D2) $F-n-2n$ (P2) $0-d-n$ (D22, Y2)	
	F20	A	$\beta^{\star}$ , $\gamma$ (B50, C47)	12 sec. $(C1)$	$5.0$ (F1, B50) cl. ch.	$2.2$ (B50) cl. ch. recoil	$F-d-p$ (F1, C1) $F-n-\gamma$ (N1) $N_{\alpha-n-\alpha}$ (N1)	
10	Ne <sup>19</sup>	A	$6^+$	20.3 sec. (W7)	$2.20 \, (W7)$ cl. ch.		$F-p-n$ (W7)	
	$Ne^{21}$	A	ß-	40 sec. (A1, B6)	4.1 (P21) abs.		$Na-n-p$ (A1, N1, P1) $Mg-n-\alpha$ (A1, B6) $Ne-d-p$ (P21, W24)	
11	Na <sup>21</sup>	в		23 sec. $(C27)$			$Ne-p-n$ $(C27)$ $Ne-d-n$ (P21)	
	Na <sup>22</sup>	A	$B^+$	$3.0 \text{ yr.} (L3)$	$0.58$ (L3) ci. ch.	$1.3 \ (O2)$ spect.	$Mg-d-\alpha$ (L3) $F-\alpha-n$ (L3, M4) $Ne-d-n$ (L3)	
	Na <sup>24</sup>	A	$\beta^-$ , $\gamma$	14.8 hr. $(V1)$	$1.4$ (L21) spect.	2.0, 1.46, $3.03 \text{ (C28)}$ spect.	$Na-d-p$ (L4, V1) Na-n- $\gamma$ (A1) $Mg-n-p (A1)$ $Al-n-\alpha$ $(A1)$ $Mg-d-\alpha$ (H4)	
12	$\rm Mg^{23}$	A	$\beta^+$	$11.6$ sec. $(W7)$	2.82 (W7) cl. ch.		$Na-p-n$ (W7, D9)	
	$Mg^{27}$	A	$\beta^-, \gamma$	10.2 min. $(H_4)$	$1.8$ (C13) cl. ch.	$0.9$ (R4) cl. ch. recoil	$Mg-d-p$ (H4) $Mg-n \gamma$ (A1) Al-n-p $(A1)$	
13 <sup>1</sup>	$Al^{26}$	A	$\pmb{\beta}^+$	7.0 sec. (W7, F2)	$2.99$ (W7) cl. ch.		$Na-\alpha (M4, F2)$ $Mg-p-n$ (W7, D9) $Mg-p-\gamma$ (C29)	

TABLE 2- *-Continued* 

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RADIOELEMENT			TYPE OF <b>BADIATION</b>	<b>HALF-LIFE</b>		ENERGY OF RADIATION IN MEV.	PRODUCED BY
z	$\boldsymbol{A}$	CLA88			Particles	$\gamma$ -Rays	
13 I	AI <sup>28</sup>	A	$6^-$ , $\gamma$	2.4 min. (A1, M5, E2)	$3.3$ (C <sub>6</sub> ) cl. ch.	$2.3$ (C6) cl. ch. recoil	$A1-d-p$ $(M5)$ $Al-n-\gamma$ (A1) $S_{i-n-p}$ (A1) P-n- $\alpha$ (A1) $Mg-\alpha-p$ (E2, R3)
	A129	A	$\beta^-$	6.7 min. (B25)	$2.5$ (B25) cl. ch. and abs.		$Mg - \alpha - n$ (B25, H21, F3)
14	Si <sup>27</sup>	A	$B^+$	$3.7-4.9$ sec. $(K8, 1)$ C27)	$3.74 \ (M21)$ cl. ch.		Al-p-n (K8, M21, C27)
	S <sub>1</sub> 31	A	$\beta^-$	170 min. (N3)	$1.8$ (K4) cl. ch.	No $\gamma$ (N3)	$Si-d-p$ (N3) $Si-n-\gamma$ (A1) $P - n - p$ (A1, P2) $S\nu\alpha$ (S2, C9)
15	$P^{29}$	A		$<$ 10 sec. (W11)			$Si-p-n$ (W11)
	$P^{30}$	A	$B^+$	$2.55$ min. $(R3)$	$3.0$ (B48) cl. ch. ä,		$Al-a-n$ (R3, C7) $S-d-\alpha$ (S2) $P-n-2n$ (P2) $P-\gamma-n$ (B20) $Si-p-n$ (B23) $Si-He^{k-p}$ (A7)
	P <sub>32</sub>	A	$B^-$	14.30 days (C8)	$1.69$ (L5) spect.	No $\gamma$ (K4)	$P-d-p$ (N3) $P-n-\gamma$ (A1) $S-n-p(A1)$ $Cl-n-\alpha$ (A1) $S-d\alpha$ (S2) Si- $\alpha$ - $p$ (F3)
16	S11			$<$ 10 sec. $(V4)$			$P-p-n$ (V4)
	$S^{36}$	A	ß-	88 days (L6, L58)	$0.107$ (L6) spect.		$CL-n-p$ (A3, L6, L58) $S-d-p$ (C25)
17	C133	A		$2.8$ sec. $(H31)$			$S-d-n$ (H31)
	Cl <sup>34</sup>	A	$6^+$	33 min. (S2, B21)	$2.5$ (B21) abs.		$P-\alpha-n$ (F2, R3, B21) Sdn(S2) $Cl-n-2n$ (P2) $Cl - \gamma \cdot n$ (B20) $S_{\alpha}$ - $p, n$ or $S_{\alpha}$ - $d$ (S45)
	C135	A	$\beta^+$ , K, $\beta^-$	$>1$ yr. (G8)	$0.7(f^{-})$ (G8) abs.		$Cl-n-\gamma$ (G8) $C1-d-p$ (G8)

TABLE 2—*Continued* 

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TABLE *2—Continued* 

# ARTIFICIAL RADIOACTIVITY 229

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## TABLE 2—*Continued*

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## TABLE *2—Continued*

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<b>RADIOELEMENT</b>			TYPE OF RADIATION	<b>HALF-LIFE</b>		ENERGY OF RADIATION IN MEV.	PRODUCED BY	
z	A	CLASS			Particles	$\sim$ Rays		
27	Co <sub>55</sub>	B	$B^+$ , $\gamma$	$18.2 \text{ hr.} (D5)$	1.50(L21) spect.	0.16, 0.21, 0.8, $1.2 \, (C20)$ cl. ch. recoil	$Fe-d-n$ (D5, L8) $Fe-p-\gamma$ (L9)	
	Co <sup>66</sup>	в	$K, \gamma, e^{-}$ ; $\beta^+(?)$ (L10)	270 days (L10)	$0.4~(B^{+})$ (L10)		Fe-d-2n (L9, B24, P4) $Ni-d-\alpha$ (L11) $F_{\mathbf{e}} p_n$ (L9)	
	Co <sup>8</sup>	A	$\beta^+$ , $\gamma$	72 days (L10)	$< 0.5$ (L10) abs.	$0.6$ (L10) abs. Pb	$Fe-d-n$ (L9, B24, P4) $Mn-\alpha-n$ (L9) $Ni-d-\alpha$ (L11) $F_{e-p-n}$ (L9) $Ni- n-p$ (V5, L10, L56)	
	Co <sup>60</sup>	A	$\beta^-, \gamma$	5.5 yr. (L10)	0.16, 1.5 (R9) abs.	$1.3$ (L9) abs. Pb	$C_0$ -d- $p$ (L9, B24) $Co-n-\gamma$ (R9, L9)	
	Co <sup>60</sup>	в	$I.T.(?)$ , $e^-$ (L10)	11 min. (H7, L10)			$C_{0}$ -n- $\gamma$ (H7, L8) $Ni-n-p$ (H8)	
28	Ni <sup>57</sup>	A	$\pmb{\beta}^+$	36 hr. (L11)	$0.67$ (L11) abs.		$F_{\theta-\alpha-n}$ (L11) $N_{1-n-2n}(?)$ (L11)	
	N <sub>103</sub>	A	$\beta^-$ , $\gamma$	$2.6 \text{ hr.} (L11)$	1.9(L11) abs.	1.1(L11) abs. Pb.	$Ni-d-p (L11)$ $Ni-n-\gamma$ (H8) $Cu-n-p$ (H8) $Zn-n-\alpha$ (H8) $Ni-n-2n$ (H8)	
29	Cu <sup>68, 60</sup>	с	ß+	81 sec. (D4)			$Ni-p-n$ (D4)	
	$Cu58$ , $60$	с	$B^+$	$7.9 \text{ min.} (D4)$			$N_i$ -p-n $(D4)$	
	Cu <sup>61</sup>	B		$\beta^+$ ; K (A4) 3.4 hr. (T1, R3)	$0.9$ (R3) abs.	No $\gamma$ (G2)	$Ni-d-n$ (T1) $Ni-p-n$ (D4) $Ni-p-\gamma$ (D4) $N$ i- $\alpha$ - $p$ (R3)	
	Cu <sup>52</sup>	A	$B^+$	10.5 min. (H8)	$2.6$ (C13) cl. ch.		$Cu-n-2n$ (H8) $Cu-\gamma-n (B20)$ $Co-\alpha$ (R3) $N_{1}$ - $p$ -n (S18) $Ni-p-\gamma$ (S18) Cu-d-p,2n(?) (K22)	
	Cu <sup>64</sup>	A	$\beta^-$ ; $\beta^+$ ; $K$ (A4)	12.8 hr. $(V2)$	$0.58~(B^-),$ $0.66~(\beta^{+})$ (T6, T11) spect.	$\mathrm{No} \gamma$ (T6)	$Cu-d-p$ $(V2)$ $Cu-n-\gamma (H8)$ $Cu-n-2n$ (H8) $Ni-p-n$ (S18, D4) $Z_{n-n-p}$ (H8)	

TABLE 2- *-Continued* 



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TABLE 2—*Continued* 

RADIOELEMENT			TYPE OF	HALF LIFE RADIATION		ENERGY OF RADIATION IN MEV.	PRODUCED BY
z	$\boldsymbol{A}$	CLA88			Particles	y-Rays	
31 <sup>1</sup>	$Ga^{70}$	A	$\beta^-$ , $\gamma$	20 min. (B20, A1)	$1.7$ (S25) cl. ch. (K.U.)		Ga- $n-\gamma$ (A1) $Ga-n-2n$ (P2) Ga- $\gamma$ -n (B20) $Z_{n-p-n}$ (D <sub>2</sub> , V <sub>7</sub> ) $Zn-a-p$ (M8)
	$Ga^{72}$	A	$\beta^-$ , $\gamma$	14 hr. (S6, L20)	$2.6$ (L28) abs.	1.0(S7) abs. Pb	Ga- $d$ -p (L20) Ga- $n-\gamma$ (S6)
32 I	Ge <sup>89</sup>	Е	$_{\beta^+}$	29 min. (S6)			Ge- $n-2n$ (S6)
	$Ge^{69}$ , 71	C		6-10 days (S6, L28)			Ge- $n-$ ? (S6) $Ga-d-2n$ (L28)
	Ge <sup>71</sup>	B	$\pmb{\beta}^+$	26-37 hr. (M8, S25)	$1.0$ (M8) abs.		$Zn-\alpha-n$ (M8) $Gen-\gamma$ (S6) $Ge-d-p$ (S6) $Ga-d-2n$ (L28) $Ge-n-2n$ (S25)
	$Ge^{87+89+71}$	Е		195 days (M8)			$Zn-\alpha-n$ (M8)
	Ge75, 77	Е	$\beta^-$	81 min. (S6)	$1.1$ (S25) el. ch. (K.U.)		Ge- $n-\gamma$ (S6) Ge- $d$ - $p$ (S6)
	$Ge^{75.77}$	Е	$\beta^-$	8 hr. (S6)	$1.9$ (S25) cl. ch. (K.U.)		$Gen-\gamma$ (S6)
33	As <sup>71</sup>	F	$\beta^+$ , $\gamma$	50 hr. (S26)			$Ge-d-n$ $(S26)$
	As <sup>71</sup>	F	$\beta^+$ , $\gamma$	88 min. (S26)			$Ge-d-n$ (S26)
	As <sup>†2</sup>	E	$\beta^+$	26 hr. (V4)			Ge-p-n $(V4)$
	As <sup>74</sup>	A	$\beta^-$ , $\beta^+$ , $\gamma$ (S26)	$16 \text{ days}$ (S26)	1.3 $(\beta^{-})$ , $0.9~(B^{+})$ (S26) cl. ch. (K,U.)		As- $n-2n$ (S26, C11) $Ge-d-n$ (S26) Se-d- $\alpha$ (F8) Ge-p- $n(D9)$
	As <sup>76</sup>	A	$\beta^-$ , $\gamma$ ; $B^+$ , K, $\gamma(?)$ (S23)	$26.8 \text{ hr.}$ (W9)	1.1, 1.7, 2.7 $(\beta^{\star})$ (S23, $W9$ . $0.7, 2.6$ ( $6^+$ ) (S23) cl. ch.	3.2, 2.2, 1.5 (S23) cl. ch. pair	As- $d \cdot p$ (C11, T3) As- $n-\gamma$ (C11) Br-n- $\alpha$ (C11) Ge- $p-n$ (V4) Se- $n-p$ (S26) Se-d- $\alpha$ (F8)
	$\mathrm{As}^{77}$	D		$f^-, \gamma$ (S26) 90 days (S26)	$0.12$ (S <sub>26</sub> ) cl. ch. (K.U.)		Ge- $d-n$ (S26)

TABLE 2—*Continued* 

	<b>RADIOELEMENT</b>		TYPE OF RADIATION	<b>HALF-LIFE</b>		ENERGY OF RADIATION IN MEV.	PRODUCED BY	
z	$\boldsymbol{A}$	<b>CLASS</b>			Particles	$\sim$ Rays		
33	As <sup>78</sup>	A	$\beta^-$ , $\gamma$	65 min. (S9)	$1.4$ (S26) cl. ch. (K.U.)	$0.27$ (S26) abs. Pb	$Br-n-\alpha$ (S9, C11, S26) Se- $n-p$ (S26)	
34	Se <sup>75</sup>	A	$K, \gamma, e^-$	48 days (D9)		$0.50$ (D9) spect. conv.	As- $p-n$ (D9)	
	Se79 81	C	$I.T., e^-$ (L30)	57 min. (S9, L30)		$0.098$ (L30) spect. conv.	Se-d- $p$ (S9, L30) Se- $n-\gamma$ (S9, H10) $Br-n-p$ (S9, L30) $S_{\mathbf{e}} - n$ (B20)	
	Se <sup>79</sup> , 31	с	в-	19 min. (L30)	$1.5$ (L30) abs.		Se- $d$ - $p$ (S9, L30) Se- $n-\gamma$ (S9, H10) $Se-\gamma-n (B20)$ $Br-n-p$ (L30) $Se^{79.51} (57 min.) I.T.$ (L30)	
	Se <sup>83</sup>	A	$6-$	30 min. (L30)			Se-d- $p$ (L30) Se- $n-\gamma$ (L30)	
	Se	D		Several hours (B15)			Th- $n$ (B15)	
	Se	D		Several days (B15)			Th- $n$ (B15)	
35	$Br^{78}$	A	$\beta^+$ , $e^-$ , $\gamma$	$6.4 \text{ min.} (S9)$	2.3 $(3^+)$ (S9) abs.	0.046, 0.108 (V7) spect. conv.	Se-d- $n$ (S9) <b>As-α-π</b> (S9) $Br-\gamma$ -n (B20, C5) $Br-n-2n$ (H10) Se-p-n (B13, V7)	
	Br <sup>80</sup>	A	(S10, V3, V7, G22)	$I.T., \epsilon^-, \gamma \mid 4.4 \text{ hr.}$ (B13)		0.049; 0.037 or 0.025 (V7) spect. conv. $0.037$ ( $G22$ ) abs. Al	$Br-n-\gamma$ (S9, S10, A2) $Br-d-p(S9)$ Se-p-n $(B13, V7)$ $Br-\gamma \cdot n$ (B20) $Br-n-2n$ $(P2)$	
	Br <sub>80</sub>	A	$\beta^-$ , $\gamma$	18 min. (S9, S10)	2.0(A2) spect.	$< 0.5$ (B13, S9) abs.	$Br-n-\gamma$ (S9) $Br-d-p(S9)$ Se- $p-n$ (B13) $Br-\gamma-n$ (B20) $Br-n-2n$ (P2) $Br^{80}$ (4.4 hr.) $I.T.$ (S10)	
	Br <sup>82</sup>	A	$\beta^-$ , $\gamma$	34 hr. (S9)	$0.7$ (B13)	$0.65$ (K5) cl. ch. recoil and abs.	$Br-n-\gamma$ (K5, S9) $Br-d-p(S9)$ Se- $p$ - $n$ (B13) Se-d-2n (S9) $Rb-n a (S9, P2)$	

 $\text{TABLE 2—}Continued$
RADIOELEMENT			TYPE OF	<b>HALF-LIFE</b>		ENERGY OF RADIATION IN MEV.	PRODUCED BY
z	A	CLASS	RADIATION		Particles	y-Rays	
35	Br <sup>88</sup>	A	в-	140 min. (L30)	$1.05$ (L30) abs.	No $\gamma$ (S9)	Se-d-n (S9) Se <sup>33</sup> $\beta$ <sup>-</sup> decay (S9, L30) Th-n (B15, L30) $U-n$ (L30)
	Br > 82	D		40 min. $(D6)$			$U-n$ (D6, H22, H57)
	Br > 32	F		22 hr. (B15)			Th- $n$ (B15)
	Br > 82	D		$3.8 \text{ hr.} (\text{H22})$			$U-n$ (H22)
36	Kr79, 81	с	$\beta^{+}$ (B41)	34 hr. (B41)	$0.5$ (B41) cl. ch.		$Kr-d-p$ (C45, S9) $Br-p-n$ (B41, C41) Se- $\alpha$ -n (C45)
	$Kr^{79.31}$	с	γ; $no\beta^+(C41)$	$I.T.(?)$ , $e^-$ , 13 sec. (C41)		$0.187$ (C41) spect. conv.	$Br-p-n$ (B41, C41)
	$Kr^{79.81}$	с	$\bm{\gamma}$ : $no\beta^+(C41)$	$I.T.(?)$ , $e^-$ , 55 sec. (C41)		$0.127$ (C41) spect. conv.	$Br-p-n$ (B41, C41) Se- $\alpha$ -n(?) (K3)
	$Kr83*$	Α	$I.T., e^-$ (L30)	113 min. (L30)		$0.049$ (L30) abs. of $e^-$	$Br^{38} \beta$ <sup>-</sup> decay (L30) $Se$ - $\alpha$ - $n$ (C45) $Kr-d-p(C45)$
	$Kr^{37}$	Е	ß-	74 min. (S9)			$Kr-d-p$ (S9) Se- $\alpha$ - $n(?)$ (K3)
	$\mathbf{K} \mathbf{r}$ 87	в	6	4.5 hr. $(S9)$			$Kr-d-p$ (S9, C45)
	Kr <sup>89</sup>	в	ß-	$\sim$ 2 min. (G9, G21)			$U-n$ (G9, G21)
	Kr <sup>88</sup>	A	ß-	3 hr. (L27, H28)			Th- $n$ (H29, A5, L27) $U-n$ (H <sub>28</sub> , H <sub>11</sub> , G <sub>9</sub> , G <sub>21</sub> )
	Kr > 90	D	$\theta^-$	$< 0.5 \,\mathrm{min.} \,(H28)$			U-n $(H28)$ Th- $n$ (H29)
37	$Rb^{32}$	в		20 min. (H51)			$Br-\alpha n$ (H51)
	Rb <sup>34</sup>	в		$6.5 \text{ hr.} (\text{H}51)$			$Br-\alpha n$ (H51) $Kr-d-n$ $(H51)$
	Rb	F		$42 \text{ min.} (\text{H}51)$			$Kr-d-n$ (H51)
	Rb	F		$200 \text{ hr.} (\text{H}51)$			$Kr-d-n$ $(H51)$
	$Rb^{ss}$	A	ß-	18 min. (S9)	$4.6$ (G21) abs.		$Rb-n-\gamma$ (S9, P2) Pa- $n(G7)$ $Kr^{ss} \beta$ <sup>-</sup> decay (H28, L27, H <sub>11</sub> , G <sub>21</sub> )
	Rb <sup>s9</sup>	в		$\beta^{-}$ , $\gamma$ (G21) 15 min. (G9, G21)	3.8(G21) abs.		$Kr^{89} \beta$ <sup>-</sup> decay (G9, G21)

TABLE *2—Continued* 

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	<b><i>RADIOELEMENT</i></b>		<b>TYPE OF</b> RADIATION	<b>HALF-LIFE</b>		ENERGY OF RADIATION IN MEV.	<b>PRODUCED BY</b>		
z	A		CLA88			Particles	$\gamma$ -Rays		
37	$Rb^{86}$ $88$	С	$\theta^-$	18 days (S9)			$Rb-n-\gamma$ (S9)		
	$Rb$ >00	D	ß-	80 sec. (H28)			$Kr$ >90 $\beta$ <sup>-</sup> decay (H28)		
38	Sr <sup>86</sup>	A		$K, \gamma$ (D13) 65 days (D13)		$0.8$ (D13, D25) abs. Pb	$Rb-p-n$ (D13, D25)		
	Sr <sup>85</sup>	A	$I.T., e^-, \gamma$ (D25)	70 min. (D25)		$0.170$ (D <sub>25</sub> ) spect. conv.	$Rb-p-n$ (D13, D25)		
	$Sr87+$	A	$I, T., e^-, \gamma$ (D11)	$2.7 \text{ hr.} \quad (D11)$		$0.37$ (D11) spect. conv.	$Sr-n-n$ (D13, R15, D25) $Rb-p-n$ (D11) $Sr-d-p$ (D11) $Sr-n-\gamma$ (D11, R15) $Y^{37}$ (80 hr.) K decay (D11, D25) $Sr-p-p(?)$ (D25) $Z_{r-n-\alpha}$ (S46)		
	Sr <sup>89</sup>	A	$\beta^-$	55 days (S24)	1.50(S24) cl. ch.	No $\gamma$ (S24)	$Sr-d-p$ (S11, S24) $Sr-n-\gamma$ (S11, S24) $Y_{-n-p}$ (S12) Rb <sup>39</sup> $\beta$ <sup>-</sup> decay (G9, H28, G21 $Z_{r-n-\alpha}(?)$ (S46)		
	$\mathrm{Sr}\geq 90$	D	ß-	7 min. (L26)			$U_n$ (L26, H28)		
	Sr > 90	D	β	6 hr. (H28, L26)			$Rb$ <sup>200</sup> $\beta$ <sup>-</sup> decay (H <sub>28</sub> )		
39	$Y^{86}$	в		$K,\gamma$ (D25) 105 days (D25)		$2(?)$ (D13, $(D25)$ abs.	$Sr-p-n$ (D13, D25)		
	$\mathbf{Y}^{37}$	в	$I.T., \epsilon^-, \gamma$ (D25)	14 hr. (S24, D13)		$0.5$ (D25) abs.	Sr-d-n (S24, D13, D25) $Sr-p-n$ (D13, D25)		
	$Y^{37}$	A	$K$ (D13)	80 hr. (D25)		No $\gamma$ (?) (D25)	Sr-p-n (D13, D25) Sr-d-n (D13, S24, D25)		
	$Y^{ss}$	A	$\beta^+$	$2.0 \text{ hr.} (S24)$	$1.2$ (S11) cl. ch. (K.U.)		$Sr-dn$ (S11, S24) $Y - n - 2n$ (S11) $Sr-p-n$ (D13, D25)		
	$Y^{90}$	Α	ß-	60 hr. (S11)	$2.6$ (S11) cl. ch. (K.U.)		$Y-d-p$ (S11) $Y_{\cdot}n_{\gamma}$ (S11, S12) $Ch-n \alpha$ (S42, S13) $Z_{r-n-p}$ (S46) $Zr-d-\alpha$ (S46)		
	Y > 90	D		$3.3 \text{ hr.} (\text{H28})$			$Sr>90$ (6 hr.) $\beta$ <sup>-</sup> decay (H28, L26) $Z_{r-n-p}(?)$ (S46)		

TABLE *2—Continued* 



TABLE 2—*Continued* 

<b>RADIOELEMENT</b>			TYPE OF RADIATION	<b>HALF-LIFE</b>	ENERGY OF RADIATION IN MEV.		PRODUCED BY
z	А	CLASS			Particles	$\gamma$ -Rays	
42	$Mo^{34}$	F		7 hr. (D9)			$\text{Cb-}p-n(?)$ (D9)
	$Mo^{91.93}$	с	$\beta^+$	17 min. (B20, S12)	$2.65$ (S46) cl. ch. (K,U.)		$Mo-n-2n$ (H10, S12, S46) $M_0$ - $\gamma$ -n (B20)
	Mo <sup>99</sup>	в	β-, γ	67 hr. (S14)	$1.5$ (S14) abs.	$0.4$ (S14) abs.	$Mo-d-p$ (S14) $M_0$ -n- $\gamma$ (S14, S12) $U-n$ (H23) Th- $n$ (H24) $M_0 - n - 2n$ (S46)
	Mo <sup>101</sup>	в	$\beta^-$	19 min. (S40, S22)	$1.8$ (S40) cl. ch. (K,U)		$M_0$ -n- $\gamma$ (S40, S22, S46)
43	4396	в	$\beta^+(?)$	$2.7 \text{ hr.} (D4)$			$C_{\mathbf{b}-\alpha-n}$ (K3) $Mo-p-n$ (D4) $Mo-d-n$ (S14)
	4399	в	$I.T., \epsilon^-, \gamma$ (S14)	$6.6 \text{ hr.}$ (S14)		$0.136$ (S14) spect. conv.; $\sim 0.18$ (S14) abs.	$Mo^{99} \beta^-$ decay (S14)
	43101	в	ß-	9 min. (840, S22)	$1.1$ (S40) ci. ch. (K,U.)		$M0$ <sup>101</sup> $\beta$ <sup>-</sup> decay (S40, S22 S46)
	43	D	$K, \sigma$	90 days (C12)		$0.096$ (H25) spect. conv.	$Mo-d-n$ (C12, C24)
	43	D	$K, \gamma$	$62 \text{ days } (C12)$			$Mo-d-n$ (C12, C24)
	43	D	$K(?)$ , $e^{-}$ , $\gamma$ (E5)	110 hr. (E3)	$0.6$ (E3)	0.05, 0.5 (E5)	$M_0$ -p-n (E3, E5)
	43	Е	$\beta^-$ , $\gamma$ (E3)	55 min. (E5)	$2.5$ (E5) abs.		$M_0$ -p-n (E3, D4, E5)
	43	Е	$6-$	$36.5 \text{ hr.} \; (\text{D4})$			Mo-p-n $(D4)$
	43	Е	ß-	18 sec. $(D9)$			$Mo-p-n$ (D3, D9)
	43	D	K	$\sim$ 2 days (S14)			Mo- $dn$ (S14)
44	Ru*	F		20 min. (D7)			$Ru-n-2n(?)$ (D7, P2)
	$Ru^{103}$	в	$\mathbf{G}^-$	$4 \text{ hr.}$ (D7, L13)			$Ru-n-\gamma$ (D7) $Ru-n-2n$ (D7, P2) $Ru-d-p$ (L13)
	$Ru^{105}$	в	$\beta^-$	20 hr. (D7)			$Ru-n \gamma$ (D7)

TABLE 2—*Continued* 

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	RADIOELEMENT	8	<b>TYPE OF</b> RADIATION	<b>HALF-LIFE</b>		ENERGY OF RADIATION IN MEV.	PRODUCED BY		
z	A			CLA			Particles	$\gamma$ -Rays	
47	Ag107*, 109*	с	$I.T., e^-$	40 sec. (A12)		$0.093$ (V7, A12) spect. conv.	$Cd107$ , 109 (6.7 hr.) K decay (A12) Cd <sup>107</sup> 109 ( $\sim$ 90 days) K decay (H25) $Ag-n n (A12)$		
	$Ag^{108}$	Α	$\beta^-$	2.3 min. (A1, B20) 2.8 (N4)	cl. ch.		$Ag-n-\gamma$ (A1) $Ag-\gamma-n$ (B20) $Pd-p-n$ (D2, E6) $Cd-n-p$ (P6) $Ag-d-p$ (K12)		
	Ag <sup>110</sup>	A		$\beta^-$ , $\gamma$ (P6) 22 sec. (A1, P6)	2.8(G4) cl. ch. (K.U.)		$Ag-n-\gamma$ (A1) $Cd-n-p$ (P6)		
	Ag108 110	C	$\beta^-$	225 days (L14, R10			$Ag-n-\gamma$ (R10, L14, A8, M12) $Ag-d-p$ (K12)		
	Ag <sup>111</sup>	A	$\sigma$	7.5 days (K6, P6)		$\overline{\text{No }\gamma}$ (K6, P6)	$Pd-d-n$ (K6, P6) $Pd-a-p$ (P6) $Cd-n-p$ (P6) Pd <sup>111</sup> $\beta$ <sup>-</sup> decay (K6) $U-n$ (N9)		
	$\rm Ag^{112}$	A	$\beta^-, \gamma$	$3.2 \text{ hr.}$ (P6)	$2.2$ (P <sub>6</sub> ) ci. ch.		$Cd-n-p$ (P6) In-n- $\alpha$ (P6) $U-n$ (N9)		
48	Cd107, 109	C	$K, \gamma$ (D4, V7, W11, A12)	$6.7 \text{ hr.}$ (D <sub>4</sub> , R <sub>5</sub> )		$0.53$ (V7) abs. Pb	$Ag-p-n$ (D4, R5, V7, W11) $Ag-d-2n$ (K12, A12)		
	$Cd$ 107, 109	с	Κ	$\sim$ 90 days (H25)			$Ag-d-2n$ (H25)		
	Cd <sub>103</sub>	Е	$\pmb{\beta}^+$	33 min. (P2)			$Cd-n-2n$ (P2)		
	$Cd^{\text{116}}$	A	$\beta^-$ , $\gamma$	2.5 days (G5)	$1.11 \text{ (C14)}$ spect.	$0.55$ (L57) cl. ch. recoil	$Cd-d-p$ (C14) Cd-n- $\gamma$ (G5, M10) $C_{d-n-2n}$ (G5) $U-n$ $(N9)$		
	$Cd^{117}$	A	ß-	$3.75 \text{ hr.}$ (C14)			$Cd-d-p$ (C14) $Cd - n - \gamma$ (M10, G5) $U-n$ $(N9)$		
	$Cd*$	D	$I.T., e^-$	50 min. (D8)			$Cd-n-n$ (D8) $U-n$ $(?)$ $(N9)$		
49	In <sup>110</sup>	D	$B^+$	65 min. (B17)	$1.6$ (B17) spect.		$Cd-p-n$ (B17) $Ag-\alpha-n$ (K9) $Cd-d-2n$ (L57)		
	In <sup>110</sup>	D	$\beta^+$ , $\gamma$ , $e^-$	20 min. (B17)	1.7 $(\beta^+)$ (L57) el. ch.	$0.16$ (B17) spect.conv.	$Cd-d-2n(L57)$ $Cd-p-n$ (B17) Ag- $\alpha$ -n (K3, K9)		

TABLE 2—*Continued* 

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# TABLE *2—Continued*

\* Radioactive isomer of stable nucleus.

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<b>RADIOELEMENT</b>			TYPE OF RADIATION	HALF-LIFE	ENERGY OF RADIATION IN MEV.		PRODUCED BY
z	A	CLA88			Particles	$\gamma$ -Rays	
50	Sn < 126	D	$\beta^-$	26 hr. (L17)			$Sn-d-p (L17)$ $Sn-n-\gamma$ (L17)
	Sn <sup>126</sup>	D	$\beta^-$	10 days (L17)			Sn- $d_{\tau}p$ (L17) $Sn-n-\gamma$ (L17)
	Sn < 123	D		$\sim$ 400 days (L17)			$Sn-d-p (L17)$
	$Sn^{125}$	B	$\beta^-$	$9 \text{ min.} (L17)$			$Sn-d-p (L17)$ $Sn-n-\gamma$ (L17)
51	Sb	E	$\beta^-$	$3.5 \text{ min.}$ (D9)			$Sn-p-n$ (D9)
	Sb116, 118	Е	$\pmb{\beta}^+$	$3.6 \text{ min.} (R16)$			$In-\alpha-n (L16, R16)$
	$Sb^{120}$	A	$B^+$	17 min. (H10, L18)	$1.53$ (A10) cl. ch.		$Sb-n+2n$ (P2, H10) $Sb-\gamma-n$ (B20) $Sn-d-n (L18)$ $Sn-p-n$ (D9)
	$Sb^{122}$	A	ß-	$2.8 \text{ days}$ (L <sub>28</sub> )	0.81, 1.64 (A10, M35) cl.ch.and abs.	$0.96$ (M35) coincid abs.	$Sb-d.p$ (L18) $Sb-n-\gamma$ (A1, L18) $Sn-d-2n$ (L18) $Sn-p-n$ (D9)
	Sb124	A	$B^-$	60 days (L18)	$1.53$ (M35) abs.	$1.82 \ (M35)$ coincid. abs.	$Sb-d-p$ (L18) $Sb-n-\gamma$ (L18) I-n- $\alpha$ (L18)
	Sb < 126	D	$\sigma$	3 hr. (L18)			$Sn-d-n (L18)$
	$Sb<$ 128	D		$\sim$ 45 days (L18)			$Sn-d-n (L18)$
	Sb < 120	D		$\sim$ 2 yr. (L18)			$Sn-d-n$ (L18)
	$Sb^{127}$	A	$\theta^{-1}$	80 hr. (A6)			$U_n(A6)$
	$Sb^{129}$	A	$6-$	4.2 hr. $(A6)$			$U_n$ (A6)
	Sb > 111	D	$\beta^-$	$<$ 10 min. (A6)			$U-n$ (A <sub>6</sub> , S <sub>21</sub> ) Th- $n$ (S21)
	$Sb$ > 181	D	$\beta^-$	$<$ 10 min. $(A6)$			U- $n(A6)$
	Sb > 11	D	$\beta^-$	$5 \text{ min.} \quad (A6)$			U-n $(A6)$
52	Te <sup>121</sup>	A	$K, \epsilon^-$	125 days (S15)			$Sb-d-2n$ (S15) $Sn - \alpha - n$ (S15) $8b-p-n$ (S15)
	Te <sup>127</sup>	A	$I.T., e^-$ (S15)	90 days (S15)		$\sim$ 0.10 (H <sub>25</sub> ) spect. conv.	Te-d- $p$ (S15) I- $n-p$ (S15)

TABLE *2—Continued* 

# ARTIFICIAL RADIOACTIVITY 243





# ARTIFICIAL RADIOACTIVITY 245





TABLE *2—Continued* 

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# ARTIFICIAL RADIOACTIVITY 247



RADIOELEMENT			TYPE OF RADIATION	<b>HALF-LIFE</b>	ENERGY OF RADIATION IN MEV.		<b>PRODUCED BY</b>
z	$\boldsymbol{A}$	CLA88			Particles	y-Rays	
77	$Ir^{192,194}$	C	$\mathbf{S}^-$	19 hr. (M15, A1)	2.2(A2) spect.		$Ir-n-\gamma$ (M15, A1, P2, J4) Au-d- $\alpha$ , $p(?)$ (C18)
	$Ir^{192,104}$	c	$\beta^-$	60 days (M15, F6)			$Ir-n-\gamma$ (M15, F6, J4)
78	Pt <sup>197</sup>	в	ß-	18 hr. (M15)			$Pt-n-\gamma$ (M15) $Pt-d-p$ (C19)
	$Pt^{197}$	в	o-	3.3 days (M15)			$Pt-n-\gamma$ (M15, P2)
	$Pt^{199}$	A	$\beta^-$	31 min. (M15)			Pt-n- $\gamma$ (M15, A1, M14) $Pt-d.p$ (C19)
79	$Au^{100}$	в	ß-	13 hr. (M15)			Au-n-2n (M15)
	$Au^{195}$	в	$\beta^-$ , $\gamma$	$4-5$ days $(M15)$	$0.36$ (C43)	$0.41 \ (C43)$	Au- $n-2n$ (M15)
	$Au^{198}$	A	$\beta^-$ , $\gamma$	$2.7 \text{ days } (M15, A1)$	$0.8$ (M15, R2) abs. and cl. ch.	0.28, 0.44, $2.5$ (R <sub>2</sub> , S <sub>17</sub> ) cl.ch.recoil	Au-n- $\gamma$ (M15, A1, P2) Au-d- $p$ (C18)
	Au <sup>199</sup>	A	ß-	3.3 days (M15)			$Pt^{199}\beta^-$ decay (M15)
80	$Hg^{197}$	в	$K, e^-$ , $\gamma$ (R11, A4)	43 min. (H10, M15)	< 0.4 (M15) abs.	$< 0.25$ (M15) abs.	$Hg-n-2n$ (M15, H10, P2)
	Hg203,205	с		25 hr. (M15)			$Hg \cdot n \cdot \gamma$ (M15, A9)
81	T1200	F		4 min. (K3)			Au- $\alpha$ -n(?) (K3)
	T1200	F		$3.8 \text{ hr.} (\text{K3})$			Au- $\alpha$ -n(?) (K3)
	T1204	B	ß-	4.23 min. (F17)	$1.6$ (F17) abs.	No $_{\gamma}$ (F17)	Tl-n- $\gamma$ (P10, P2, H10) $T1-d-p$ (F17) $Tl-n-2n$ (F17, P2, H10)
	Ti <sup>205</sup>	B	ß-	$1-2$ yr. $(F17)$			$Tl$ - $n-\gamma$ (F17) $T1-d\cdot p$ (F17)
82	Pb <sup>200</sup>	в		80 min. (D10)			$Pb-n-2n$ (D10)
	Pb204*, 206*	с	$I.T.$ (?) $e^-$ , $\gamma$	52 hr. (F17)		$~0.5$ (F17) abs. of e-	$T1-d-n$ (F17)
	Pb <sup>209</sup>	в	ß-	$3.0\,\text{hr.}$ (T5)			$Pb-d-p$ (T5)
	$Pb*$	D	$I.T., e^-$	$1.6 \text{ min.}$ (W27)		~10.3~(W27) abs. of $e^-$	$Pb-x-rays (W27)$

TABLE *2—Continued* 

RADIOELEMENT			TYPE OF RADIATION	<b>HALF-LIFE</b>	ENERGY OF RADIATION IN MEV.		PRODUCED BY
z	$\boldsymbol{A}$	CLABB			Particles	$\gamma$ -Rays	
83	<b>Bi210</b>	А	$6-$	$5 \text{ days}$ (L13)			$Bi-d-p$ (L13, C26, H27) $\text{Bi-}n-\gamma$ (M29)
84	$P0$ 210	A	$\pmb{\alpha}$	136 days			Bi <sup>210</sup> $\beta$ <sup>-</sup> decay (L13, C26, H27 Bi-d-n (V4, C26, H27)
	$P0$ 211	A	$\alpha$	$\sim$ 10 <sup>-3</sup> sec. (C46, C23)	7.5 (C46, C <sub>23</sub> abs.		85211 K decay (C46, C23)
85	85211	A	$\alpha$ , K	$7.5 \text{ hr.} (\text{C46, C23})$	$6(\alpha)$ (C46, C <sub>23</sub> abs.		$Bi - \alpha - 2n$ (C46, C23)
90	UV <sub>231</sub>	в	$\beta^-$	$24.5 \text{ hr.} (N5)$			$Th-n-2n(N5)$
	Th <sup>233</sup>	A	В-	26 min. (M17)			Th- $n-\gamma$ (M17)
91	Pa <sup>233</sup>	F	в-	25 days (M17)			Th <sup>233</sup> $\beta$ <sup>-</sup> decay(?) (M17)
92	TJ239	A	$\beta^-$	23 min. (I1, S4)			$U - n - \gamma$ (H18, H14, I1, M19
	TT237	в	$\beta$ , $\gamma$ (M37)	$\sim$ 7 days (M37, N8) 0.26 (M37)	abs.		$U - n - 2n$ (M37, N8)
93	93289	А	$\beta^-$	2.3 days (M28, M19	$0.47$ (M28) abs.	0.22, 0.27 (H25) spect. conv. and spect.	$U^{239} \beta^-$ decay (M28)

TABLE 2— *Concluded* 

as possible. References to the original discoveries are not given when better data are available in more recent publications. The references which are listed usually give a key to the complete literature.

The half-lives of H<sup>3</sup>, Be<sup>10</sup>, and C<sup>14</sup> have been estimated from the measured intensities of the radioactivities together with reasonable assumed values for the yields.

## VII. APPLICATIONS TO CHEMISTRY

Some of the applications of artificial radioactivity which have been made to chemistry are described in this section. The use of the natural radioelements as "indicators" or "tracers" in physical and chemical investigations has found a wide application. Descriptions of the methods and the

types of problems which have been solved may be found in the books written by Hahn (H39), Paneth (P12), and Hevesy and Paneth (H50). The treatises on radioactivity by Curie (C38) and Meyer and Schweidler (M32) also contain discussions of such problems. These investigations were limited to those few elements which have naturally occurring radioactive isotopes. Those elements which exist both in the form of a stable type and as radioactive isotopes, such as lead (ThB), bismuth (ThC), and thallium (ThC"), are particularly suited to investigations of this kind. The "tagging" of atoms (for example, hydrogen, carbon, nitrogen, and oxygen) has also been accomplished by the use of separated (inactive) isotopes. This method, which is applicable in principle to any element which has two or more separable isotopes, has led to the solution of a large number of problems; an excellent review of this work was written in 1939 by Reitz (R22).

The large number of artificial radioactivities now known (about three hundred and fifty) makes it possible to extend the powerful radioactive indicator method to nearly every element. Although there is at least one radioactive form of every element, the restrictions imposed by the half-life, which must be sufficiently long for convenient manipulation, prevent the use of some of the radioactivities.

The application of the indicator method, by the use of the artificial radioelements, has also been made to the fields of physiology, zoology, biology, biochemistry, physics, medicine, and botany. Only the applications to chemistry will be discussed in this review. Hevesy has written a review article on the application of radioactive indicators in biology and biochemistry, which will appear in the *Annual Reviews of Biochemistry*  for 1940.

The discussion of the applications which is to be given in this section, although not detailed, should reveal the type of information that can be obtained by application of the radioactive method and should give a key to the literature on the subject. Whenever it is necessary in this discussion to designate a radioactive form of an element in a chemical equation, this will be done with the aid of an asterisk; for example, bromide ions which have been "labelled" as the result of the presence of radioactive bromide ions will be written Br<sup>-\*</sup>. (The asterisk as used in table 2 has another meaning,—namely, to denote radioactive isomers of stable nuclei. It is not likely that this usage, which is in conformity with the practice of nuclear physics, will give rise to confusion.)

#### *1. Exchange reactions*

Perhaps the most extensive chemical application of the artificial radioelements as indicators has been to the study of "exchange reactions." The first exchange experiments were performed by Hevesy and coworkers (H56), who used the natural radioactive lead isotope ThB to demonstrate a rapid exchange between the lead atoms in aqueous lead nitrate and lead chloride and also between plumbous and plumbic acetates in acetic acid solution. Reitz  $(R22)$  and Rosenblum and Flagg  $(R20)$  have written reviews (1939) on the applications of artificial radioelements for the measurement of exchange reactions. Exchange reactions were discussed also by Breineva and Roginsky (B49). In exchange experiments the atoms of an element, in one of its valence forms or types of chemical combination, are "labelled" by admixture with some radioactive isotope of the element which is in the same form of chemical combination. To this system is added the element in another state of valence or form of combination (containing none of the radioactive isotope); the presence of radioactivity in this second chemical form, after it has been separated from the first, shows that an effective exchange of atoms between the two different states of the element has taken place. Complete exchange has been attained when the radioactivity has distributed itself between the two chemical forms in the same ratio as the amounts of the element in the two forms, that is, when the specific activity is the same. In all exchange reactions, regardless of the order, the rate varies with time according to the law for first-order reversible reactions, since the chemical composition of the reaction mixture remains unchanged (M34).

The following description of exchange work should serve to illustrate the type of information which can be obtained from such experiments. For example, these experiments give information on chemical bond types, the strength and reactivity of chemical bonds and the effect of solvents on these properties, the reactivities of ions and compounds, the structure of ions and compounds, the mechanism of reactions, and the mechanism of catalysis. In addition, exchange reactions often offer an excellent and convenient method for the introduction of radioactive atoms into compounds.

The experiments of several groups of investigators have shown that the thermal exchanges between halogens and the corresponding halide ions in aqueous solution at room temperature are practically instantaneous. Radioactive Cl<sup>38</sup>, Br<sup>80</sup>, Br<sup>82</sup>, and I<sup>128</sup> were used as indicators. Long and Olson (L39) were able to show, by means of a method of competing reactions, that radiochlorine and chloride ion, in acidified aqueous solution, undergo an extremely fast exchange, and Halford (H58) has pointed out that their results, taken together with other data, show that the half-life for the exchange is less than  $10^{-4}$  sec. for 1 *M* Cl<sup>-</sup>. Roginsky and Gopstein (R21) and Dodson and Fowler (D18) showed that the brominebromide exchange is very rapid in aqueous solution, while Hull, Shiflett,

and Lind (H40), Juliusberger, Topley, and Weiss (Jl), and Dodson and Fowler (D18) showed that the same is true for the iodine-iodide exchange. Topley and Weiss (T9) have shown that bromine and hydrobromic acid exchange bromine atoms rapidly also in dry carbon tetrachloride at room temperature, and Libby (L55) has shown that iodine and antimony triiodide undergo exchange within 20 min. in dry pentane at 37<sup>o</sup>C.

Brejneva, Roginsky, and Schilinsky (B34) found complete exchange at room temperature within 10 min. between *solid* aluminum bromide  $(A<sub>2</sub>Br<sub>6</sub>)$ and either gaseous bromine or hydrogen bromide. Exchange experiments between gaseous bromine and solid cupric bromide led Roginsky and Gopstein (R21) to the unexpected conclusion that the two bromine atoms in solid cupric bromide are not equally reactive. Kolthoff and O'Brien (K20) found an exchange, more than 50 per cent complete in 100 min. at room temperature, between bromine gas and solid silver bromide.

Hull, Shiflett, and Lind (H40) showed that there is no very rapid exchange between iodine and iodate ion in  $1 N$  sulfuric acid solution. The experiments of Libby (L43) show that there is no appreciable rate of exchange at room temperature between perchlorate ion and chlorate ion or between perchlorate ion and chlorine or chloride ion in either acid or alkaline solution. He finds that chlorate and radiochlorine exchange at a measurable rate in acid solution, the half-time of the reaction being about 95 min. in 1 *M* sodium chlorate and 2 *M* sulfuric acid. Libby (L43) and Rollefson (R24) found that bromate and bromine exchange at a faster rate in acid solution, while Kennedy (K17) and Libby (L43) used radioactive I<sup>31</sup> to show that iodate and iodine exchange in acid solution. According to Libby (L43), the exchange rates seem to increase in the same order as the ordinary reactivities of the halate ions. Polessitsky (P17) found that bromate and bromide, as well as iodate and iodide, do not exchange even after hours at  $100^{\circ}$ C. (presumably in slightly alkaline solution).

It has been found by many investigators (S28, H40, M24), using radioactive Cl<sup>38</sup>, Br<sup>80</sup>, Br<sup>82</sup>, and I<sup>128</sup> as indicators, that the non-ionizing alkyl halides (as well as the alkyl dihalides, haloforms, and tetrahalides) do not undergo thermal exchange at any appreciable rate at room temperature with free halogens or halide ions. Szilard and Chalmers (S28), who were probably the first to make this observation, irradiated with neutrons compounds such as ethyl iodide and bromoform, from which they separated the radioactive atoms in concentrated form as silver halides. For iodine and either methyl iodide (M24) or ethyl iodide (H40) exchange was not observed even after about 15 min. at  $90-100^{\circ}$ C. However, exchanges of this type do take place when certain solvents are used for the mixtures of alkyl halides and inorganic halides or halogens. McKay (M24) and Juliusberger, Topley, and Weiss (Jl) found that iodide undergoes rapid

exchange (1 to 2 min.) with methyl iodide at room temperature in ethyl alcohol (J1, M24), acetone (M24), or amyl alcohol (M24). When iodine was substituted for iodide, the exchange required an elevated temperature  $(M24)$ ; complete exchange occurred in alcohol after 15 min. at 100<sup>o</sup>C. However, McKay (M24) and Juliusberger, Topley, and Weiss (Jl) did not observe exchanges at room temperature between iodide and ethyl, n-propyl, isopropyl, and methylene iodides in ethyl alcohol or between iodide and iodoform in acetone; McKay (M24) found that all of these exchanges did occur within 15 min. in solvents at 100°C. Hull, Shiflett, and Lind (H41) also showed that the exchange between iodide and ethyl iodide in alcohol was slow at room temperature and very rapid  $(< 1$  min.) at 100°C. There is a pronounced difference between the behavior of methyl iodide and the other alkyl halides in these experiments.

McKay (M24) has demonstrated that phenyl iodide, p-nitroiodobenzene, and  $p$ -iodoaniline do not exchange with iodide at  $100^{\circ}$ C. in ethyl alcohol, and it was shown by Friedmann, Solomon, and Werthessen (FlO) that the bromine atoms of phenyl bromide and sodium bromide do not interchange. Juliusberger, Topley, and Weiss (J2) showed that only the negative, ionizing iodine of diphenyliodonium iodide exchanges with iodine in a water-alcohol mixture at room temperature.

Brejneva, Roginsky, and Schilinsky (B34, B35) found in a detailed study that the very reactive reagent aluminum bromide, at room temperature with no auxiliary solvent, did rapidly exchange bromine atoms with certain alkyl bromides, benzyl bromide, and all the bromines in certain alkyl polybromides; the exchange also occurred with the aromatic bromides, but was much slower. These experiments give important information on the strength of a number of bromine-carbon bonds. They also observed a fast exchange between aluminum iodide and ethyl iodide, and suggest that the use of radioactive aluminum halides should offer a general convenient method for the synthesis of radioactive organic halides. The radioactive aluminum halide can be prepared by taking advantage of the complete exchange between solid aluminum halide and gaseous radioactive halogen or hydrogen halide. In a later communication Brejneva, Roginsky, and Schilinsky (B46) show that, although two alkyl bromides do not undergo exchange alone, the presence of aluminum bromide effects exchange according to the following mechanism:

> $RBr^* + AlBr_3 \rightleftharpoons RBr + AlBr_2Br^*$  $R'Br + AlBr<sub>2</sub>Br^* \rightleftarrows R'Br^* + AlBr<sub>3</sub>$

 $RBr^* + R'Br \rightleftarrows R'Br^* + RBr$ 

In the presence of aluminum bromide and no auxiliary solvent, exchange between the pairs ethylene dibromide and ethyl bromide, bromoform and ethyl bromide, amyl bromide and ethyl bromide reached practical completion in experiments of 45 min. duration.

Some thermal and photochemical exchange reactions of bromine have been studied by Wilson and Dickinson (W22). They found a rapid thermal exchange (within 10 min.) between radioactive bromine and either arsenious bromide or stannic bromide at room temperature in carbon tetrachloride solution. In the same solvent no thermal exchange was observed to occur between radioactive bromine and either ethylene dibromide (65 min.) or trichlorobromomethane (40 min.) in the dark at  $100^{\circ}$ C., but a rapid exchange (practically complete in 20 min.) between bromine and trichlorobromomethane is induced by green light at  $76^{\circ}$ C.

Grinberg and Filinov (G20) showed that radioactive bromide ions in aqueous solution at room temperature undergo complete exchange with the four bromine atoms of  $K_2PtBr_4$  and the six bromine atoms of  $K_2PtBr_6$ . They state that this probably means that the six positions of the bromines in the octahedron of  $PtBr_6^-$  are equivalent. Polessitsky (P17) performed exchange experiments to demonstrate the equivalence of the four iodine atoms in  $K_2HgI_4$ .

Brejneva, Roginsky, and Schilinsky (B34) used radiobromine to show that gaseous bromine and hydrobromic acid undergo complete exchange within 15 min. at room temperature. In a more detailed investigation Liberatore and Wiig (L45, L46) showed this exchange to be homogeneous and non-photochemical in character and almost complete within 2 min. As a mechanism they suggest a chain involving bromine atoms reacting according to

$$
Br + HBr^* = HBr + Br^*
$$

**and** 

$$
Br + BrBr^* = BrBr + Br^*
$$

followed by similar reactions of Br\* with hydrogen bromide and bromine, and they suggest that the primary bromine atoms are produced in the ionization processes associated with the radioactive bromine radiations. Libby (L47) also observed a fast exchange, and he suggests as an alternative mechanism a direct bimolecular reaction between bromine and hydrogen bromide through an  $HBr<sub>3</sub>$  complex. The question of the mechanism has not yet been settled. Libby (L47) found that iodine and hydroiodic acid undergo exchange either in the gaseous state or in dry pentane.

Liberatore and Wiig (L46) showed that gaseous hydrogen bromide and ethyl bromide do not undergo exchange at room temperature even when

exposed to the full light of a 500-watt tungsten lamp for 9 hr. However, rapid thermal exchange takes place when hydrogen bromide and ethyl bromide are heated to  $200-300^{\circ}$ C., and they suggest as an explanation the equilibrium  $C_2H_5Br = C_2H_4 + HBr.$ 

Ruben, Norris, and Nahinsky (R31) used radioactive C<sup>11</sup> to show that there is no exchange between gaseous carbon dioxide and carbon monoxide within 1.5 hr. at  $200^{\circ}$ C.

Andersen  $(A3)$  used radioactive  $S<sup>35</sup>$  (88 days half-life) to demonstrate that there is no exchange between the two sulfur atoms in thiosulfate ion, and thus proved that the sulfur atoms in thiosulfate are not equivalent. Voge and Libby (V8) and Voge (V6) have also used the radioactive  $S^{35}$  to study the exchange of sulfur atoms between various valence forms of sulfur in aqueous solution. They found that an exchange took place (within the time taken for the experiment, 1 hr.) in polysulfide at  $100^{\circ}$ C., thus demonstrating a probable equivalence of the sulfur atoms in the polysulfide ion. They also observed a much slower exchange of sulfur atoms between sulfide ion and thiosulfate ion (after about 20 hr.) at  $100^{\circ}$ C. Their experiments showed that there is no appreciable exchange between sulfide ion and sulfate ion in alkaline solution or between sulfite and sulfate in either alkaline or acid solutions, even after 36 hr. at 100°C. They showed that there is no exchange of sulfur between aqueous thiosulfate and sulfite within 20 hr. at  $20^{\circ}$ C., but thiosulfate and sulfite completely exchange one sulfur within an hour at  $100^{\circ}$ C. This is in agreement with the results of Andersen concerning the non-equivalence of the two sulfur atoms in thiosulfate. Gaseous sulfur dioxide and sulfur trioxide did not exchange at temperatures appreciably below those at which dissociation of the trioxide might be expected. Cooley, Yost, and McMillan (C25) have shown that no exchange occurs when elementary sulfur is dissolved in carbon disulfide and the solution kept at a temperature of  $100^{\circ}$ C. for 68 hr.

The exchange reactions of manganese in its various valence states in aqueous solution have been investigated by Polissar (P13), who used the radiomanganese  $(Mn^{66})$  of 2.6 hr. half-life as indicator. His experiments showed that manganous ion and manganic ion (present as the manganic oxalate complex ion), undergo a rapid and complete exchange. He found that there was no rapid exchange between the following pairs of valence states: (a) permanganate ion-manganous ion, (b) permanganate ion $\pm$ manganic ion (oxalate complex), (c) permanganate ion-manganese dioxide (solid), and  $(d)$  manganous ion-manganese dioxide (solid). Libby  $(L43)$ found a very rapid exchange at room temperature between manganate and permanganate ions; it seems certain that the mechanism involves an electronic exchange between the two ions.

Perrier and Segrè (P14) used the 14.3-day radiophosphorus ( $P^{32}$ ) to investigate the possibility of an exchange between phosphate and hypophosphite in aqueous solution. Their experiments showed that there was no exchange either during 10 days at room temperature or 24 hr. at  $100^{\circ}$ C. in neutral or acid solution. Wilson (W18) showed that there is no exchange between phosphate and phosphite either in acid or alkaline solution within 1 day at 100°C., or in acid solution after 26 days at  $25^{\circ}$ C. His results also showed that, if the phosphorus atoms in hypophosphoric acid  $(H_4P_2O_6)$  are equivalent, the equilibrium constant for the formation of hypophosphoric acid from phosphorous and phosphoric acids is less than  $8 \times 10^{-5}$  (mole<sup>-1</sup> liters) at  $25^{\circ}$ C. in 5.6 *M* hydrochloric acid.

Wilson and Dickinson (W19) used radioarsenic (As<sup>76</sup>, 26 hr. half-life) to show that arsenate and arsenite do not exchange, in either acid or alkaline solution, even after  $3 \text{ hr}$ , at  $100^{\circ} \text{C}$ .

Some exchange reactions of iron were studied by Kennedy, Ruben, and Seaborg  $(K15)$ , who used  $Fe<sup>59</sup>$  (47 days) as indicator. They studied oxidation-reduction exchanges and found that ferrous and ferric ions undergo instantaneous exchange (< 10 sec.) in 6 *N* hydrochloric acid solution at room temperature, while ferrocyanide and ferricyanide ions do not exchange in slightly acid or slightly alkaline solution within 3 days at room temperature. Ferric ion and ferricyanide ion do not exchange. Likewise, no exchange of iron atoms occurs in precipitates of either Prussian blue or Turnbull's blue. Ruben and Nahinsky (R31) used the 43-min.  $He<sup>197</sup>$  to show that mercurous and mercuric ions undergo rapid exchange in aqueous solution at room temperature.

Ruben, Kamen, and Frenkel (R32) have employed the  $10.2$ -min. Mg<sup>27</sup> to study the nature of the magnesium bonds in chlorophyll. There was no exchange in 40 min. at room temperature between  $Mg^{++}$  and highly purified samples of either chlorophyll *a* or chlorophyll 6 in a buffered 80 per cent acetone solution. The exchange between Mg<sup>++</sup> and the magnesium compound of 8-hydroxyquinoline proceeds rapidly in aqueous ethanol solution at room temperature. These investigators also used radioactive Fe<sup>59</sup> to show that there is no exchange between  $\overline{Fe^{+++}}$  and ferrihemoglobin in aqueous solution or between  $Fe^{+++}$  and ferriheme in ethanol at room temperature even in experiments lasting several weeks. Since it is known from magnetic susceptibility measurements (P20) that the iron in both of these molecules is held by ionic bonds, it seems that the highly symmetrical electrostatic field of the porphin ring is sufficiently strong to prevent any reversible equilibrium involving the central metal ion.

A few solid-phase-liquid-phase exchange reactions have been studied. Rollin (R26) has investigated the exchange between metals and their ions in aqueous solution. In a qualitative experiment he found appreciable exchange when inactive zinc metal, in the form of a powder, was shaken for 1 hr. at room temperature with a solution of zinc chloride which contained radioactive  $Zn^{65}$  prepared by the deuteron bombardment of zinc. Also, Rollin used the 8.2-day Ag<sup>106</sup>, prepared by the deuteron bombardment of palladium, to measure the exchange between metallic silver, in the form of a mirror on a glass surface, and silver ions in a silver nitrate solution. In some experiments the metal was labelled with the radioactive  $Ag^{106}$ , while in others the ions were radioactive. He found that exchange amounting to ten to one hundred atomic layers took place within an hour or two at room temperature. This seems to be explicable on the basis of a mechanism which involves local electrolysis caused by the existence of irregularities on the surface of the metal. Hevesy and Biltz (H49) had previously used a naturally radioactive lead isotope to demonstrate an exchange between metallic lead and a solution of plumbous salt in contact with it.

Kolthoff and O'Brien (K20) used the 4.4-hr. Br<sup>80</sup> for the study of the exchange between bromine and solid silver bromide at room temperature with ethyl bromide as solvent for the bromine. They found that complete exchange occurs in a few hours with fresh silver bromide, and that the rate of exchange decreases when the age of the precipitate is increased, until finally no exchange occurs with drastically aged silver bromide. They also measured the exchange between radioactive iodine (using  $I^{128}$ ) and fresh silver bromide with ethyl iodide as solvent for the iodine. In a similar manner Kolthoff and Yutzy (K19) measured the exchange between silver chloride and chloride ions, and Kolthoff and O'Brien (K21) and Polessitsky (P17, P18) measured the exchange between silver bromide and bromide ions in aqueous solution. Such experiments lead to the determination of the rates of aging of precipitates. Kolthoff and coworkers have previously applied naturally radioactive elements to problems of this type.

Although the subject of exchange reactions is too complicated to make accurate, complete generalizations possible, it seems profitable to make a few rough statements about homogeneous exchange reactions. If we consider exchanges of a given element between two sorts of molecules or ions in which it is held by electron-pair bonds to different numbers or kinds of other atoms, we may say in general that such exchange reactions do not proceed with appreciable rates except in those cases where there are reversible reactions which enable the exchanging atoms to reach equivalent states of chemical combination. For example, there is no exchange of atoms between phosphate and phosphite ions, sulfate and sulfite ions, sulfur and carbon disulfide, iodide ion and iodoform, etc. On the other hand, exchanges have been found between chlorine and chlorate ion (due to the oxidation-reduction equilibrium), between lead nitrate and lead

chloride (an extreme example of the ionization exchange mechanism), and between iodide ion and iodine (through the formation of a symmetrical intermediate,  $I_3^-$ ). Most of the homogeneous exchange reactions reviewed in this section may be classified as one of these particular types. When the two exchanging molecules differ only in their net charge, another exchange mechanism—the transfer of an electron from one to the other may become possible. For example, exchanges have been observed between  $Fe^{++}$  and  $Fe^{+++}$  and between  $MnO_4^{--}$  and  $MnO_4^{--}$ . It is no doubt true that some exchanges occur through a simple transfer of atoms between molecules during a collision; such a mechanism is a special case of exchange through the formation of an intermediate. In many cases, the observation of exchanges of this sort suggests the existence of unstable intermediates which might not be known from other reaction studies.

## *2. Study of reaction mechanisms*

The artificial radioelements offer a powerful tool for the study of rearrangements and reaction mechanisms in general. A few examples will serve to illustrate the type of problems which can be solved by this method.

Hughes, Juliusberger, Mastermann, Topley, and Weiss (H42) have used radioiodine  $(I^{128})$  to make a careful measurement of the rate of exchange between iodide ion and sec.-octyl iodide in acetone solution. They showed that this rate was equal to the velocity of racemization of the *d-sec*octyl iodide by iodide ion under identical conditions and hence established in a direct way the fact that the inversion is preceded by a substitution, in agreement with the ideas of Olson and Long (04). Hughes, Juliusberger, Scott, Topley, and Weiss (H43) employed the same method to prove that the racemization of  $\alpha$ -phenylethyl bromide in the presence of bromide ion is preceded by a substitution.

Rollefson and Libby  $(R25)$  used radioactive Cl<sup>38</sup> to study the nature of primary photochemical processes in solution. They use the fact that no exchange of chlorine atoms occurs within 30 min. at room temperature in an illuminated solution of radiochlorine in carbon tetrachloride to help them deduce that the high efficiency of photodissociation processes in solution is due to a high primary efficiency rather than to reaction of the dissociation products with the solvent. The availability of long-lived radioactive isotopes of antimony, tellurium, arsenic, zinc, etc. should make it possible to extend the scope and sensitivity of Paneth's mirror technique (P19) for the detection of free radicals. Leighton and Mortenson (L53) have already used the natural radioactive lead isotope RaD in an application of the highly sensitive radioactive method to test for free radicals in photochemical reactions.

Olson, Halford, and Hornel (05) used radiochlorine to demonstrate

that the rearrangement of N-chloroacetanilide to  $o$ - and  $p$ -chloroacetanilide proceeds, not by means of an intramolecular rearrangement, but through a mechanism which involves a chlorine intermediate. Long (L40) was able to show that the racemization of chromioxalate ion,  $Cr(C_2O_4)_3$  – –, does not proceed through an ionization mechanism, since his experiments with radiocarbon  $(C<sup>11</sup>, 21$  min. half-life) showed that there is no exchange between oxalate ion and chromioxalate ion. Ettle and Johnson (E12) used radioactive Cl<sup>38</sup> to show that the interconversion in aqueous solution of green 1:6-dichlorobisethylenediaminocobaltic chloride to the violet 1:2-isomer does not proceed by an intramolecular mechanism.

Wilson and Dickinson (W19), using the radioarsenic  $(As^{76})$  of 26 hr. half-life, found a measurable rate of exchange between trivalent and pentavalent arsenic in acid solution in the presence of iodine and iodide ion. They used the radioactive indicator method to measure *k* and *k' at equilibrium* for the reaction

$$
H_3\text{AsO}_3 + I_{\overline{3}} + H_2O \xleftarrow{\hat{\kappa}} H_3\text{AsO}_4 + 3I^{-} + 2H^{+}
$$

and showed that the values which they obtained agreed with those obtained from measurements made far from equilibrium.

Experiments of the kind performed by Brejneva, Roginsky, and Schilinsky (B35) described above (study of aluminum bromide-organic bromide exchanges) should offer an insight into the mechanism of halogenation catalysis by substances of the aluminum bromide type. These same investigators (B44) made a detailed study of the kinetics of the aluminum bromide-ethyl bromide exchange reaction in carbon disulfide solution, where the rate is sufficiently slow for convenient measurement. They found an activation energy of 11 kg-cal. and favor a mechanism which involves the formation of an intermediate complex with the formula RAlBr4.

Brejneva, Roginsky, and Schilinsky (B45) have mixed solutions of bromide ions in water and in ethyl alcohol in order to measure, with the aid of the 4.4-hr. Br<sup>80</sup> and the 34-hr. Br<sup>82</sup>, the rate of exchange between bromide hydrate and bromide alcoholate:

# $\text{Br}^-(\text{H}_2\text{O})_n + \text{Br}^-(\text{C}_2\text{H}_5\text{OH})_p \rightleftarrows \text{Br}^-(\text{H}_2\text{O})_k(\text{C}_2\text{H}_5\text{OH})_i$

Surprising results were obtained. They found a rather slow rate; a time considerably longer than 10 min. was required for complete exchange at  $-31^{\circ}$ C. Further experiments on this "inter-solvate" exchange were performed by Roginsky and Tartakovskaja (R30), who showed that chloride and iodide as well as bromide ions showed slow solvate exchange at room temperature for the following pairs of mixed solvents: methyl alcohol-water, acetone-water, glycerol-water, glycerol-acetone, and ethyl alcohol-water. Roginsky and Afanasiev (R35) have made a detailed study of the kinetics of this type of exchange, using bromide ions and water-acetone mixtures at temperatures of  $0^{\circ}$ ,  $16^{\circ}$  and  $32^{\circ}$ C.

Le Roux and Sugden (L41) have used radioactive  $Br^{80}$  and  $Br^{82}$  to make a detailed study of the kinetics of the slow exchange of bromide ion and n-butyl bromide in aqueous acetone (90 per cent acetone by volume) over a range of temperatures from  $0^{\circ}$  to  $65^{\circ}$ C. Elliott and Sugden (E7) have extended these experiments to include three more alkyl bromides. The activation energies found are: n-propyl, 18.12; n-butyl, 18.87; isobutyl, 20.21; isopropyl, 22.94 kg-cal. The speed of these bimolecular reactions is of the same order of magnitude as that predicted for a gaseous system on the collision hypothesis; isopropyl bromide shows the highest value for the effective collision diameter.

Tuck  $(T10)$  has measured the rate of exchange of radioiodine  $(I^{128})$ with tertiary butyl iodide in liquid sulfur dioxide under conditions which enabled him to deduce the velocity of electrolytic dissociation of the butyl iodide from the measurements.

It should be pointed out that the long-lived radiohydrogen (H3) and radiocarbon  $(C<sup>14</sup>)$  offer opportunities for the study of many very important reaction mechanisms, and a few such experiments are in progress at the present time.

#### *S. Reactions of high-energy atoms*

The work which has been done on the mechanism of the Szilard-Chalmers method of concentration and on the mechanism of the nuclear isomer separation method will be described in this section. This will include a discussion of the reactions and behavior of the atoms and molecules of very high energy which are formed as the result of neutron capture and isomeric transitions.

Szilard and Chalmers (S28) first demonstrated that a neutron-capture radioactivity can be very highly concentrated by suitable chemical methods. They irradiated ethyl iodide containing traces of iodine with neutrons and found that they could extract most of the radioactivity with water; this activity  $(I^{128})$  was associated with an amount of iodine which was equivalent to only a small fraction of the total irradiated iodide. Later, Fermi and coworkers (Al) separated concentrated radioactive halides from neutron-irradiated halogenates and radioactive manganese dioxide (Mn<sup>66</sup>) from irradiated potassium permanganate. D'Agostino (D19) studied these processes in more detail and also separated concentrated active arsenic  $(As^{76})$  from irradiated cacodylic acid,  $(CH_3)_2AsOOH$ .

The formation of a radioactive isotope by neutron capture (the  $n, \gamma$ reaction) is accompanied by the emission of gamma-rays with an average

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energy of 3 to 6 Mev. (R23, F11, K16). The emission of a gamma-ray of energy *E* (Mev.) imparts a recoil energy of 535 *E<sup>2</sup> /M* electron volts to an atom of atomic weight *M* when the nucleus of that atom captures a slow neutron. This energy, which, for example, amounts to about 100 electron volts in the case of manganese, is practically always sufficient to disrupt the molecule which contains the irradiated nucleus.

When the newly formed radioactive atoms are in, and remain in, a chemical form which is separable from the bulk of irradiated compound, there is obtained a high percentage of *extraction* of the radioactivity from, and hence small *retention* by, the irradiated compound. D'Agostino (D19), for example, found that he could extract as silver chloride practically 100 per cent of the 37-min. Cl<sup>38</sup> when solid or aqueous sodium chlorate or perchlorate was irradiated with slow neutrons. He showed in similar experiments that there is also practically no retention of  $I^{128}$ by solid or neutral aqueous sodium iodate; the same is true for As<sup>76</sup> when cacodylic acid is irradiated.

However, there are several factors which may act to decrease the percentage of extractable activity. The newly formed radioactive atoms may undergo thermal exchange  $(cf.$  section VII, part 1) with the inactive atoms in the irradiated compound and hence reenter the original form. Or the high-energy fragments which contain the radioactive recoil atoms may react while activated with surrounding molecules so as to form new molecules or re-form the irradiated molecules. The mechanism of the expulsion process and the reactions of such high-energy atoms have been the subject of rather detailed study by several groups of investigators.

Erbacher and Philipp (E8) studied the separation of radioactive halogens by aqueous extraction of irradiated alkyl halides. However, these investigators were only interested in obtaining the radiohalogens as free as possible from inactive halogens and did not make a detailed study of the efficiency of the extraction processes.

Glückauf and Fay (G16) have shown that the high-energy halogen atoms expelled by recoil from capture gamma-rays can frequently give rise to substitution products. They found, for example, dibromobenzene from irradiated bromobenzene, methylene iodide from methyl iodide, bromoform from methylene bromide, carbon tetrabromide from bromoform, etc. Identical yields of methylene iodide from irradiated methyl iodide were obtained over an extremely wide temperature interval  $(+15^{\circ}$ to  $-195^{\circ}$ C.), which shows that it is not an ordinary thermal reaction but rather a substitution of a hydrogen atom in methyl iodide by a high-energy recoil radioactive iodine atom. Considerable activity remains in the original methyl iodide, showing that the recoil iodine atoms can replace halogen as well as hydrogen. They also showed that the recoil halogens

can replace hydroxyl, amino, carboxyl, and  $-CH<sub>2</sub>OH$  groups. That such reactions offer a method for the synthesis of concentrated radioactive halogen compounds is illustrated by the fact that practically pure radioactive phenyl iodide can be separated from methyl iodide irradiated in benzene solution.

The experiments of Lu and Sugden (L42) were designed with the view of finding methods for increasing the yield of extractable radioactive halogens from irradiated organic halides. They used, as extracting agents, acidic and alkaline aqueous solutions as well as finely divided metallic powders and metallic foils. With bromobenzene and iodobenzene the presence or absence of free halogen during irradiation has no effect; with the aliphatic halides the presence of halogen increases the percentage of extraction. Radioiodine is separable mainly as the free element; radiochlorine and radiobromine as anions. They found that a small amount of aniline present during the irradiation has a very marked effect in increasing the fraction of activity which can be extracted by acidified water. They suggest that this effect is explained by the reaction

$$
\rm R\,+\,X^*\,+\,NH_2C_6H_5\rightarrow C_6H_5NH_2R^+\,+\,X^{*-}
$$

where R represents the highly energetic molecular residue which remains after the disruption of the molecule RX following the recoil of the radiohalogen X\*. However, other explanations of this effect may also be advanced.

Libby (L43) has made a study of the reactions of the high-energy recoil atoms which result after the formation of radioactive Cl<sup>38</sup>, Br<sup>80</sup>, Br<sup>82</sup>,  $I^{128}$ ,  $P^{32}$ ,  $Mn^{56}$ , and  $As^{76}$  by neutron capture. The halogenates show practically zero retention of activity in neutral or alkaline aqueous solution, while some retention occurs in acid solution. The time independence of the retention in the case of acid chlorate indicates that it is the highenergy recoils which are interchanging instantaneously to form radioactive chlorate. The increase of retention with increase of time between irradiation and extraction indicates that thermal exchange is occurring between acid bromate and iodate and their products *(cf.* section VII, part 1). Libby suggests that the retention of activity by the organic halides is best explained largely by the recombination of a stopped  $X^*$  particle with its residual free radical or ion in the same reaction "cage." The lack of dependence upon environment in the case of the phosphate retention experiments (approximately 50 per cent of the activity extractable as phosphite in all cases) indicates that the initial recoil entirely determines the retention. The recoil products from irradiated permanganate undergo a wide range of follow reactions as determined largely by the environment. The remarkable complete retention of activity observed in the case of both

arsenate and arsenite is explained on the basis of no change in valence during the recoil, followed by hydration reactions more rapid than any oxidation or reduction reactions with water. Suess (S48) studied the reactions of the high-energy bromine atoms formed as the result of neutron capture by gaseous ethyl bromide.

Another source of high-energy atoms is furnished by isomeric transitions. Segre, Halford, and Seaborg (SlO) first showed that the energy released during the radioactive transition from an upper to a lower isomeric state of a nucleus can be used to effect a chemical separation of two genetically related nuclear isomers. Subsequent work by Willard (W20) and Seaborg, Friedlaender, and Kennedy (S32) has shown that, for isomeric transitions, activation and bond rupture occur, not as the result of recoil energy (which is usually very small in isomeric transitions), but as a consequence of the high state of electronic excitation which results from the vacancy in the *K-* or L-shell created by the emission of the internal-conversion electron. Willard deduced this mechanism from the fact that isomeric transitions can initiate reactions which could not occur with energies as small as the recoil energies, while the other investigators were able to show that bond rupture occurs only when the transition gamma-rays are internally converted and not in the case of isomeric transitions with *unconverted* gammarays of even higher energy. Fairbrother (F12) also suggested this mechanism on the basis of experiments similar to those of Willard.

The radioactive isomeric transition from  $Br^{80}$  (4.4-hr. half-life) to  $Br^{80}$ (18-min. half-life) has been used to initiate a variety of chemical changes. Segre, Halford, and Seaborg (SlO) used this transition to activate the hydrolysis (or decomposition) of *tert*.-butyl bromide; Le Roux, Lu, and Sugden observed the decomposition of ethylene dibromide (L36, L42) and the hydrolysis (or decomposition) of n-butyl bromide (L36); DeVault and Libby (D12) observed the rupture of bromate ion; Fairbrother (F12) studied the decomposition of ethyl bromide. These reactions were all established by the presence of the 18-min. activity in the reaction products. Lu and Sugden (L42) found that the activated bromine from the decomposed ethylene dibromide reacts to some extent with metals, such as zinc powder and aluminum, copper and silver foil, as shown by the extraction of the 18-min. activity by these metals. Willard (W20) found that this transition activates bromine in such a way as to enable it to replace one of the chlorines in carbon tetrachloride; experiments over a wide range of temperature show the reaction to be independent of temperature, as should be expected. In a continuation of this work Willard (W23) has observed a wide range of reaction efficiencies for a number of reactions activated by this isomeric transition. He studied the reaction of bromine with ether and with mineral oil, the gas-phase decomposition

of ethylene dibromide, the decomposition of ethyl bromide, the reaction of cinnamic acid dibromide with carbon tetrachloride, and the reaction of bromine with carbon tetrachloride in the presence of high concentrations of bromine. He observed no reaction of this nature between bromine and carbon tetrachloride in the gas phase.

Libby and DeVault (L52) observed reaction efficiencies ranging from 0 to 90 per cent (as measured by the amount of the 18-min. activity found in the reaction products) for a number of reactions activated by the Br<sup>80</sup> isomeric transition. Decomposition reactions and reactions with solvents were studied. The experiments included observations on gaseous ethyl bromide, bromoform, and ethylene dibromide as well as ethyl bromide, bromoform, phenyl bromide, dibromopentane, and tertiary butyl bromide in various solvents. They suggest that their observed maximum reaction efficiency of 90 per cent corresponds to the internal-conversion coefficient for the  $Br^{80}$  isomeric transition. Imre  $(12)$  has effected a separation of the Br<sup>80</sup> isomers by means of an exchange experiment between bromide ions and solid silver chloride. Suess (S47) used the transition to activate the reaction between gaseous hydrobromic acid and acetylene.

As has already been mentioned, isomeric transitions in tellurium and selenium have been used to effect the reduction of tellurate to tellurite (S15) and selenate to selenite (L30). Seaborg, Friedlaender, and Kennedy (S32) found that the reduction of tellurate to tellurite proceeds with almost 100 per cent efficiency even when the solution is kept frozen at the temperature of liquid air. The decomposition of gaseous tellurium diethyl has also been observed (S32).

## *4. Behavior of material at extremely small concentrations*

The use of radioactive isotopes in the absence of carrier material offers a unique method for the study of the behavior of substances at extremely small concentrations. Extensive applications of the natural radioelements to such problems have been described in the books of Hahn (H39), Paneth (P12), and Hevesy and Paneth (H50).

An indication of the type of experiments which can be performed has already been given in section II, where a description was given of the methods used for the isolation of radioactive transmutation products in the absence of carrier. Among the problems which can be investigated by this method there should be mentioned the measurement of the vapor pressure of non-volatile metals and other substances, the measurement of the absorptive power of solids for extremely small amounts of gases, the behavior of extremely small amounts of insoluble substances, electrodeposition and electrochemical deposition of extremely small amounts of material, the behavior of "radiocolloids," the behavior of fractionation processes at small concentrations, etc.

Grahame and Seaborg (G11) used radioactive Ga<sup>67</sup>, Ga<sup>68</sup>, Co<sup>56</sup>, Mn<sup>56</sup>, and Fe<sup>59</sup> in order to measure at extremely small concentrations the distribution of the chlorides of these elements between ether and 6 *N* hydrochloric acid. They found distribution ratios identical with those at higher concentrations for all except ferric chloride.

Langsdorf and Segrè (L30) have used radioactive Br<sup>33</sup> and Kr<sup>33</sup> to study "emanation methods." By a technique which involved the introduction of the radioactive halogen into a silver-nitrate-impregnated silica gel they effected a very efficient emanation of the radioactive rare-gas decay product from the halogen. Hahn (H48) has discussed the diffusion rates out of uranium and thorium precipitates of the rare-gas radioactive fission products, xenon and krypton. Hahn and coworkers had previously applied this "emanation method," with the aid of the naturally radioactive rare gases radon and thoron, to a long series of investigations of the structure and the change of structure of solid substances (H39, Zl). Hahn (H48) points out that a comparison of the diffusion rates of krypton, xenon, and radon out of uranium and thorium precipitates should give the atomic weight dependence of the diffusion constants. A theoretical discussion of the emanation method and the information to be obtained from it is given by Flügge and Zimens  $(F15)$ .

Seaborg, Kennedy, and Wahl (S33) used radioactive H<sup>3</sup> to study the thermal diffusion process in hydrogen at small concentrations. They showed that the isotope separation arrangement of Clusius and Dickel  $(C34)$  gives, at mole fractions as low as  $10^{-12}$ , separation factors which are essentially the same as those obtained at much higher concentrations. Beams (B36) is also using radioactive  $H<sup>3</sup>$  in an investigation of the separation of isotopes by means of the ultracentrifuge.

An obvious application of the artificial radioelements will be to the problems of "radiocolloids" and colloid chemistry, which have been so extensively investigated with the natural radioelements (C36, C37). This has been the subject of a detailed treatment by Hahn (H39).

Another use of the artificial radioelements should be as indicators in the preparation of new compounds in a manner similar to Paneth's (P16) use of ThC (an isotope of bismuth) to demonstrate the existence of bismuth hydride.

## *5. Analytical chemistry*

The artificial radioelements can be used as an aid in analytical chemistry in many ways:— to provide a method of analysis for the presence of certain elements, as a means of studying the completeness of chemical separations, as a means of studying coprecipitation problems, etc.

The presence of certain elements in a sample which is to be analyzed can often be established, with or without the help of a chemical separation,

by means of their characteristic half-lives, after the substance has been activated in some manner. The analysis can often be made without destroying or even changing the form of the sample. A quantitative estimate of the amount of the element can be made in those cases where the yield of the reaction involving the formation of the radioactivity has been previously determined. Hevesy and Levi have applied this method of analysis to the rare-earth elements, where it is especially useful because of the extreme difficulty of application of ordinary analytical methods. In one experiment (H20) they found the 2.5-hr. dysprosium period in a sample of yttrium, after activation with neutrons, which showed the presence of dysprosium impurity to the extent of 1 per cent. Neutron activations of gadolinium samples (H19) were used as tests for small amounts of europium impurities by means of the 9.2-hr. europium period. Seaborg and Livingood (S38) bombarded a sample of iron with deuterons  $\frac{1}{2}$  and  $\frac{1}{2}$  radioactivities, the presence of gallium impurity to the extent of 6 parts per million. They used this same method to demonstrate the presence of small amounts of copper in nickel, and of iron in cobalt, as well as of phosphorus and sulfur in various substances. King and Henderson (K9) were able to detect less than 1 part in 10,000 of copper in silver by bombarding the silver with alpha-particles. However, it must be pointed out that extreme care must be exercised in the application of these sensitive methods to analysis. This is especially true when a sample is bombarded with deuterons, protons, or alpha-particles, because of the danger that there might be introduced into the sample small amounts of impurities, from recoil atoms and volatilization, in the target chamber. Even when samples are bombarded through a window with the target outside of the target outside of the target outside of the samples are bombarded unough a whidow with the target outside or the acceleration apparatus, care must be taken to prevent the introduction of extremely small amounts of impurities during the preparation of the target.

Application of the radioactive method to the analytical problems of organic chemistry also shows some promise. Brejneva, Roginsky, and Schilinsky (B34) have pointed out that measurements on the rate of exchange with aluminum bromide should help one to trace the position of the bromine in the compound during the synthesis of complicated organic bromides. The aluminum bromide undergoes only a slow rate of exchange with bromine which is attached directly to the benzene or naphthalene rings, but exchanges quickly with aliphatic bromides and benzyl bromide.

Relative intensity measurements of induced radioactivity can be used for isotopic analysis of separated isotopes (K18).

The efficiency of separation processes in analytical chemistry can be tested with the help of radioactive indicators. This is a very convenient method, because the amount of a given element which remains in any fraction after a chemical separation can be quickly determined by means of its radioactivity. For example, Erbacher and Philipp (E9) used radioactive Au<sup>198</sup> to study the completeness with which gold can be separated from platinum and iridium by a gravimetric procedure in which the elements are weighed after reduction to the metallic state. Saunders (S43) has used radioactive Te<sup>127</sup> and Te<sup>129</sup> to study, over a wide range of conditions, the amount of tellurium which is carried down with antimony when the antimony is precipitated as an oxide from a solution of boiling, concentrated nitric acid. Radioactive indicators have been used time after time in the chemical work connected with the Radiation Laboratory at Berkeley for the purpose of making quick, convenient, and effective tests of the efficiency of separation procedures in analytical chemistry.

Similarly it is possible to determine the solubility of very slightly soluble substances; for example, Ferla (F14) used radiophosphorus to study the completeness of precipitation of ammonium phosphomolybdate. Radioactive cobalt was used by Cacciapuoti and Ferla (C40) to measure the solubility in water of cobaltic hydroxide  $(5.6 \times 10^{-3} \text{ mg. per liter})$ .

Kolthoff and Yutzy (K19) have applied radioactive Cl<sup>38</sup> to a measurement of the specific surface of a silver chloride precipitate by treating the solid with a solution of radioactive chloride ions and measuring the rate of exchange as a function of time. Kolthoff and O'Brien (K20, K21) and Polessitsky (P17, P18) have used the 4.4-hr. Br<sup>80</sup> to study the surface and also the aging of silver bromide precipitates by means of exchange experiments.

#### *6. Chemical properties of rare elements*

The fact that extremely small amounts of radioelements can be detected by the radioactive methods makes it possible to use radioactive transmutation products to study the properties of elements which do not exist in nature or are so very rare that they have not yet been isolated. It must be emphasized that experiments of this type are performed with very small, unweighable amounts (approximately  $10^{-10}$  g.) of the element, usually in the presence of foreign carrier material, and therefore the interpretation of the results, especially for precipitation processes, may sometimes be uncertain.

Perrier and Segre (P15) were able to show that the deuteron bombardment of molybdenum produces radioactive isotopes of element 43, and they used these to study the hitherto unknown chemical properties of this element. Their experiments showed that the chemical properties resembled those of the heavier homolog, rhenium, to a much greater extent than they resembled those of manganese, the lighter homolog. They used rhenium as a carrier for the radioactivity in order to show that element 43

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is precipitated by hydrogen sulfide from alkaline or acid (less than 10 *N)*  solution. They investigated other properties, including the volatility of the oxide and chloride and the conditions for the electrolytic deposition of the metal. They worked out analytical procedures for the isolation of element 43 and specifically for its isolation from rhenium. Segre (S36) has written a review of the known properties of the radioactive isotopes of element 43.

The recent discovery of radioactive element 85, from bismuth plus 32- Mev. alpha-particles, by Corson, MacKenzie, and Segre (C23) makes possible an investigation of its properties. Segre, MacKenzie, and Corson (S30, C46) have carried out an investigation of its chemical properties. The general behavior is that of a metal, with little resemblance to the other halogens. It is precipitated by hydrogen sulfide in 6 *N* hydrochloric acid solution with various carriers, and the sulfide is insoluble in ammonium sulfide. It is precipitated by stannous chloride in acid solution but not by sodium stannite in alkaline solution. Volatility at comparatively low temperatures is observed; a piece of bombarded bismuth loses most of the activity before melting  $(275^{\circ}C)$ . There is no precipitation upon the addition of silver nitrate to a dilute nitric acid solution using iodide as carrier. Element 85 can be extracted with carbon tetrachloride with iodine carrier but with yields small compared with iodine under similar conditions. Worth mentioning here is the interesting observation of Hamilton and Soley (H44) that element 85 concentrates in the thyroid gland to an extent which has heretofore been peculiar only to iodine.

The decay scheme of element 85, which has a half-life of 7.5 hr. and probably has the mass number 211, is very interesting. The radioactive 85<sup>211</sup> undergoes a branching decay, going to a bismuth isotope (Bi<sup>207</sup>) by alpha-emission and to a polonium isotope  $(Po<sup>211</sup>)$  by K-electron capture. The Po<sup>211</sup> is the well-known naturally radioactive AcC' and decays to stable Pb<sup>207</sup> by alpha-particle emission with a half-life of  $5 \times 10^{-3}$  sec. The Bi<sup>207</sup> probably also decays to Pb<sup>207</sup>, since Bi<sup>207</sup> does not exist in nature as a stable isotope, but as yet no radioactivity corresponding to this decay has been found.

The remarkable discovery by McMillan and Abelson (M28) that a 2.3-day activity, formed by the bombardment of uranium with neutrons, is an isotope of element 93 makes possible an actual investigation of the chemical properties of element 93. The properties of element 93 have been the source of much speculation for a long time. The 2.3-day activity was originally discovered by McMillan (M19), who found that this radioactive isotope is not found among the highly energetic recoil products which are formed when uranium undergoes neutron-induced fission. McMillan and Abelson have shown that this 2.3-day activity is the

daughter of the  $23$ -min. U<sup>289</sup>, which is formed from U<sup>238</sup> by the capture of slow neutrons, and hence must be assigned to the isotope  $93^{239}$ . . The radioactive 93<sup>239</sup> decays by beta-particle emission to the isotope 94<sup>239</sup>, and although it is probable that the isotope  $94^{239}$  is radioactive, perhaps decaying by the emission of alpha-particles to the naturally radioactive U 235 (actinouranium), McMillan and Abelson have shown that the halflife for alpha-decay of 94<sup>239</sup> must exceed one million years.

A striking confirmation of the assignment of the 2.3-day activity to element 93 has been obtained in the physical measurements of Helmholz (H25). He placed a sample of the 2.3-day activity in an electron magnetic spectrograph and found electron lines corresponding to the internal conversion of gamma-rays in the  $K$ - and  $L$ -electron shells. Presumably these gamma-rays are emitted immediately following the beta-emission and hence the internal conversion occurs in element 94. Helmholz found that his measurements gave an energy difference of about 98 kilovolts between the *K-* and L-shells, which is very close to the difference expected for element 94. The corresponding difference between the *K-* and L-electron binding energy amounts to about 96 kilovolts in element 93 and about 94 kilovolts in uranium.

McMillan and Abelson have used this 2.3-day activity to investigate the chemical properties of element 93. They found that the chemical behavior of element 93 is much more similar to uranium than to its homolog, rhenium. For example, element 93 precipitates with sodium uranyl acetate, the reaction which is so characteristic of uranium. Likewise it precipitates with uranium in alkaline solution and redissolves upon the addition of ammonium carbonate, another reaction which is characteristic of uranium. Element 93 is not precipitated by hydrogen sulfide in dilute acid solution. It precipitates with the rare-earth elements, probably in the  $+4$  oxidation state, upon the addition of fluoride or oxalate, and is chemically similar to the rare-earth elements in many other precipitation processes. The element also precipitates with insoluble iodates, a precipitation process which is characteristic of the  $+4$  oxidation state of cerium and thorium. Chemical separation from the rare earths is effected by taking advantage of a higher oxidation state; for example, in the presence of bromate ions rare-earth precipitates do not carry the activity.

These chemical investigations with element 85 and element 93 have demonstrated that the chemical behavior of these elements differs rather markedly from that of their nearest homologs, iodine and rhenium.

Radioactive H<sup>3</sup>, although not a new element as is the case for the other elements discussed in this section, will differ enough from hydrogen (H) and deuterium (D) in its chemical and physical properties to make an investigation of some of its properties worth while. Many investigations

which have been based on differences in properties between hydrogen and deuterium can be extended to radioactive  $H<sup>3</sup>$  so as to obtain additional useful information. Such investigations include measurements of equilibrium constants of reactions and especially exchange reactions, reaction rate measurements which emphasize differences in the kinetic behavior of the three isotopes, determinations of the separation factors and efficiencies of isotope fractionation processes, study of diffusion and adsorption processes, etc. Some of these experiments would yield, for example, information on the zero point energy of the molecules HT and DT. (T is used here to represent radioactive H<sup>3</sup> , triterium.) Of course the extent of such investigations would be limited, because  $H^3$  is available only in very small, unweighable amounts. A detailed discussion of the investigations involving hydrogen and deuterium is given in the book written by Farkas (F16).

## *7. Self-diffusion processes*

The radioactive indicator method has made it possible to measure the rate of diffusion of a substance into itself, that is, to measure the rate of "self-diffusion." For example, the radioactive lead isotopes (ThB and RaD) have been used to investigate the rate of self-diffusion in lead, both liquid and solid (H54, H55, G19). The discovery of artificial radioactivity extended the possibility for the application of this method to a large number of elements.

Sagrubskij (S39) and McKay (M27) have measured the rates of selfdiffusion in gold at elevated temperatures. They activated gold with neutrons to form the radioactive gold (Au<sup>198</sup>) of 2.7 days half-life and then measured the rate of diffusion of this radioactive gold through samples of ordinary inactive gold. As an example of the results obtained, McKay found the diffusion coefficient at  $941^{\circ}$ C. to be  $9.7 \times 10^{-6}$  mm.<sup>2</sup> per minute. McKay found an activation energy of 51 kg-cal. per mole for this selfdiffusion process.

Rollin (R27) and Steigman, Shockley, and Nix (S37) have used the radioactive copper ( $Cu<sup>64</sup>$ ) of 12.8 hr. half-life to measure the rate of selfdiffusion in copper. Rollin, who activated the copper by direct bombardment with 8-Mev. deuterons, found that the diffusion coefficient at  $940^{\circ}$ C. is a few times smaller than that found by McKay at the same temperature for self-diffusion in gold. The results of Rollin show that the coefficient of self-diffusion in copper is smaller than might be predicted from the known rates of diffusion of several other elements in copper. His results gave an activation energy of 60 kg-cal. per mole for the self-diffusion process. Steigman, Shockley, and Nix electroplated the radiocopper, which was prepared by neutron bombardment of zinc, upon disks of inactive copper. They found an activation energy of 57.2 kg-cal. per mole and diffusion rates similar to those found by Rollin.
Banks and Day (B40) have measured the rate of self-diffusion in single crystals of metallic zinc. A thin deposit of radioactive Zn<sup>65</sup>, obtained from copper by proton bombardment, was electrolytically deposited on a flat, polished, and etched surface of a single crystal of ordinary inactive zinc. They obtained, for example, a value at 400°C. of 5.17  $\times$  10<sup>-5</sup> mm.<sup>2</sup> per minute for the diffusion coefficient parallel to the hexagonal axis. The value for the activation energy obtained by them is 17.6 kg-cal. per mole, a value which is much smaller than those found for the self-diffusion in gold and copper. Miller and Day (M26) measured the rates of selfdiffusion in several samples of polycrystalline zinc. Their values for the diffusion coefficients, which were close to those found by Banks and Day, varied as much as 25 per cent among the various samples.

Jehle (J3) employed radioactive Na<sup>24</sup> and Cl<sup>38</sup> in measurements of the rate of self-diffusion of sodium and chloride ions in aqueous solution over a wide range of concentrations. His results at low concentrations are consistent with the known data on the diffusion coefficients of sodium chloride. Katzin (K13) is using radioactive Na<sup>24</sup>,  $K^{42}$ , and Br<sup>82</sup> to measure the rate of diffusion of sodium, potassium, and bromide ions through various membranes in the absence of concentration gradients.

## *8. Experiments with radioactive carbon*

This section will describe the experiments which have been done with radiocarbon. Because of the special importance of carbon in chemistry, it seems desirable to discuss these experiments, in spite of the fact that most of them have been of a biochemical nature and this review has not included a discussion of the applications of artificial radioactivity to biochemistry. The experiments have been done with the  $21$ -min.  $C<sup>11</sup>$ , usually made by the deuteron bombardment of boron. The short half-life is an inconvenience, but the production of very intense activities by means of the cyclotron has made it possible to carry out experiments lasting as long as  $5 \text{ hr}$ . The production of the long-lived  $C^{14}$  of Ruben and Kamen (R17) will make it possible to perform experiments whose duration is not  $\lim_{x \to 0}$  by the decay of the sample. However, radioactive  $C<sup>14</sup>$ , which seems to be best prepared by the deuteron bombardment of the separated (or enriched) isotope C<sup>13</sup>, has such a very long half-life that exceedingly long and intense bombardments are needed in order to obtain a radioactivity of sufficient intensity for useful work.

Ruben, Kamen, and Hassid (R33, R28) have been using radioactive C 11 in a study of photosynthesis. Radioactive carbon dioxide was fed to the unicellar green algae *Chlorella* and also to higher plants under various controlled conditions in the light as well as in the dark. The results obtained so far have been rather surprising. The plants and the algae reduce carbon dioxide in the dark. The dark reduction of carbon dioxide

is very likely the first step in photosynthesis and can be represented by the equation RH +  $CO<sub>2</sub> \rightleftharpoons$  RCOOH. Decarboxylation experiments have shown that the bulk of the radioactive carbon is in the carboxyl group. Attempts to identify the radioactive substances formed in the dark and in the light have been thus far unsuccessful. It is of considerable interest to note that formaldehyde, which has played a prominent r61e in many proposed mechanisms, was not formed from the radioactive carbon dioxide introduced. Experiments with the ultracentrifuge and diffusion methods indicate the average molecular weight of the radioactive molecules to be  $\sim$  1000, which explains the failure to identify chemically these molecules with any small molecules.

Smith and Cowie (S41) have also studied the photosynthesis mechanism. They observed the reduction of carbon dioxide in the dark, confirming the results of Ruben, Kamen, and Hassid. They also used radioactive carbon dioxide to show that carbon dioxide reacts with the calcium carbonate, magnesium carbonate, etc. present in plants to form  $HCO<sub>3</sub>$ .

Barker, Ruben, and Kamen (B43) have used radioactive carbon dioxide in a study of the methane bacteria. In the presence of various alcohols, acids, etc. these bacteria produce methane. The radiocarbon experiments have clearly shown that the methane arises from a complete reduction of carbon dioxide and not from a reduction of the organic substrate.

Similarly it was found by Carson and Ruben (C35) that carbon dioxide is reduced exclusively to two acids, propionic and succinic, by the propionic acid bacteria. They suggest that these results may be of general interest in connection with a major problem encountered in tracer experiments with labelled carbon,—namely, the synthesis of radioactive molecules starting with radioactive carbon dioxide. In many instances the appropriate microörganisms may offer the best method for the desired synthesis. For example, the propionic acid bacteria converted in 30 min. over 80 per cent of the radioactive carbon dioxide into propionic and succinic acids, which were thus made readily available with a very high specific activity for further tracer experiments.

Ruben and Kamen (R29) have found that a number of heterotrophic systems, previously thought only to produce carbon dioxide in their oxidation reactions, reduce carbon dioxide. In the case of yeast, for example, one carbon dioxide molecule is reduced for every fifty carbon dioxide molecules liberated in respiration. It would seem that carbon dioxide reduction is more widespread in living systems than has been hitherto suspected. This is of considerable interest, since it has been recently established that many microorganisms require small traces of carbon dioxide.

Mention should be made in this section of the experiments of Ruben,

Hassid, and Kamen  $(R34)$ , who have used radioactive  $N^{13}$  to show that barley plants fix small amounts of gaseous nitrogen, thus confirming the ideas of Lipman and Taylor (L49).

Hastings, Kistiakowsky, Cramer, Klemperer, Solomon, and Vennesland  $(H47)$  have made studies with lactic acid containing radioactive  $C<sup>11</sup>$  in the carboxyl group. The purpose of the experiments, performed with rats, was to determine whether the increase of liver glycogen is or is not accompanied by radioactivity proportional to the amount of lactate converted to glycogen. The radioactivity of the glycogen, corresponding in weight to 33 per cent of the administered lactate, was only from  $\lt 1$  to 3.6 per cent of that of the administered radioactive material. During the same time, the expired carbon dioxide contained more than 10 per cent of the radioactive carbon administered as lactate. They conclude that these results suggest either  $(1)$  that the lactate molecule may undergo a stage of decarboxylation before conversion to glycogen or (2) that the increase in liver glycogen may have arisen in these experiments from some precursor other than the radioactive lactate.

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