The ($4n+1$) Radioactive Series*

A. C. ENGLISH, T. E. CRANSHAW, P. DEMERS, J. A. HARVEY, E. P. HINCKS, J. V. JELLEY, AND A. N. MAY
Division of Atomic Energy, National Research Council of Canada, Chalk River, Ontario, Canada
June 20, 1947

THE three radioactive series known to occur naturally are characterized by members whose mass numbers are represented by the integers $4n$ (thorium series), ($4n+2$) (uranium series), and ($4n+3$) (actinium series). While the existence of a fourth series comprising the "missing" radioactive ($4n+1$)-isotopes has been postulated at various times,[1–4] early searches which were made for its members in natural minerals gave negative results.[2,3] More recently Wahl[5] has reported the observation of a line at mass 237 in mass spectrograms from certain minerals, and attributes it to a member of the new series. Since 1935 α- and β-active ($4n+1$)-species have been produced by artificial transmutation, and among the heavy elements almost a dozen have been reported in the literature. (See for example Seaborg's Table of Isotopes[6] and his recent review.[7]) In 1942 Seaborg, Gofman, and Stoughton[8] discovered and investigated the properties of U^{233}, the long-lived α-emitting product of the β-decay of Pa^{233} and the third member of a ($4n+1$)-chain which starts with Th^{233}.

The investigation of the ensuing route taken by the chain which was undertaken by the authors started with a search for the decay products of milligram amounts of U^{233}. A chemical extraction of radium resulted in an α-activity which decayed in a complex manner with periods of the order of days. Direct evidence for a chain including at least four α-emitters was first provided by the observation of 4-track stars in a photographic emulsion which had been soaked in the radium solution. Subsequent measurement of decay periods and α-particle energies showed that these formed part of a new disintegration series, distinct from the thorium, the uranium, and the actinium series. Making use of both chemical and physical evidence it has been possible to deduce the main route of the series, which, after the α-decay of U^{233}, passes through seven new nuclear species, and ends with the known[9] 3.3-hr. β-decay of Pb^{209} to stable Bi^{209}.

The path of the series which passes through isotopes of the type ($4n+1$) is shown in Fig. 1. Both U^{233} and its daughter Th^{229} are fairly long-lived, and, as far as this work was concerned, it has been convenient to treat the former as the parent of the series. Since the radium isotope is β-active, the main chain does not include an emanation followed by the usual "A" and "B" products. Instead, the main α-sequence starts with Ac^{225}, and the series thus includes isotopes of the recently named elements, francium ($Z=87$) and astatine ($Z=85$). The end product is Bi^{209} and not a lead isotope as in the case of the other three series. (It is interesting to note that the route is almost exactly that predicted by Turner[4] in 1940.) A 2 percent α-branching of Bi^{213} was detected, but the disintegration of Tl^{209} could not be investigated because of the low activity. A search was made for α-branching of Ra^{225} but none could be found. it is probably less than one percent.

The disintegration properties of the members of the series are summarized in Table I. Previously published data included for completeness are shown in light-faced type, while results obtained by the authors are in heavy type. The α-particle energies were measured with a grid-type ionization chamber, biased amplifier, and pulse analyzer. The 21-millisecond half-life of At^{217} was measured by photographing pulses on an oscilloscope with a triggered time base, and the 3.2-microsecond half-life of Po^{213} by a coincidence experiment using a variable resolving time.

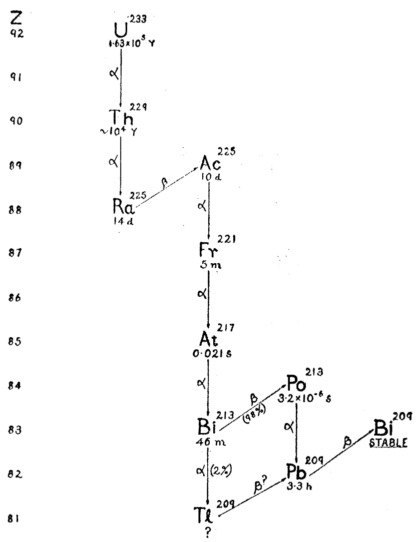

FIG. 1. The path of the ($4n+1$)-series following U^{233}.

The α-emitters of the series are found to lie on a good straight line in a Geiger-Nuttall plot (logarithm of disintegration constant against logarithm of α-particle energy), with the exception of Po^{213} which falls well below the line in the manner of the very short-lived C' bodies. This line lies between and is parallel to those representing the $4n$ and ($4n+3$)-series.

The authors feel that the name "Neptunium Series," after the longest-lived known ($4n+1$)-member, Np^{237},[7] should be adopted.

We wish to thank those members of the Montreal Laboratory who gave us assistance during the above investigation, and especially Professor F. A. Paneth and Dr. B. L. Goldschmidt whose interest and advice were invaluable.

TABLE I. The ($4n+1$) radioactive series.

Species	Type of radiation	Half-life	Energy of particles in Mev α	β
$_{92}U^{233}$	α^{10}	1.63×10^5 y[10]	**4.825±0.003**	
$_{90}$**Th**^{229}	**α**	{5×10^3 y[10]; **~10⁴ y**}	**~5**	
$_{88}$**Ra**^{225}	**β**	**14 d**		**<0.05**
$_{89}$**Ac**^{225}	**α**	**10 d**	**5.801±0.010**	
$_{87}$**Fr**^{221}	**α**	**5 m**	**6.31 ±0.02**	
$_{85}$**At**^{217}	**α**	**2.1×10^{-2} s**	**7.023±0.010**	
$_{83}$**Bi**^{213}	**α(2%), β(98%)**	**46 m**	**5.86 ±0.03**	**~1.3**
$_{84}$**Po**^{213}	**α**	**3.2×10^{-6} s**	**8.336±0.005**	
$_{82}Pb^{209}$	β^9	3.3 h[9]		0.70[9]
$_{83}Bi^{209}$	stable			

A detailed report of the work will be published later.

Note added in proof: A recalculation of the results from the measurement of the half-life of polonium 213 gives a better value of 4.4×10^{-6} seconds.

* The work reported here was carried out in the Montreal Laboratory of the Division of Atomic Energy, National Research Council of Canada, between November 1944 and May 1946.

[1] A. S. Russell, Phil. Mag. **46**, 642 (1923).
[2] W. P. Widdowson and A. S. Russell, Phil. Mag. **48**, 293 (1924).
[3] I. Curie, H. von Halban, and P. Preiswerk, J. de phys. et rad. **6**, 361 (1935).
[4] L. A. Turner, Phys. Rev. **57**, 950 (1940).
[5] W. Wahl, Science **93**, 16 (1941).
[6] G. T. Seaborg, Rev. Mod. Phys. **16**, 1 (1944).
[7] G. T. Seaborg, Science **104**, 379 (1946).
[8] G. T. Seaborg, J. W. Gofman, and R. W. Stoughton, Phys. Rev. **71**, 378 (1947).
[9] R. L. Thornton and J. M. Cork, Phys. Rev. **51**, 383 (1937); K Fajans and A. F. Voigt, Phys. Rev. **60**, 619 (1941).
[10] J. M. Cork, *Radioactivity and Nuclear Physics* (D. Van Nostrand Company, Inc., New York, 1947), information attributed to G. T. Seaborg.

Thresholds for Creation of Particles

H. FESHBACH
Massachusetts Institute of Technology, Cambridge, Massachusetts
AND
L. I. SCHIFF*
University of Pennsylvania, Philadelphia, Pennsylvania

June 9, 1947

IN the design of very high energy particle accelerators, it is of interest to know the smallest energy that a bombarding particle need have in order that a given combination of particles can be created in a collision. The calculation of such a threshold on the basis of purely energetic considerations, under the assumption that a mechanism of creation exists, was pointed out by Dr. M. S. Livingston to be a straightforward application of relativistic dynamics. This note presents the pertinent formulas, and discusses some limitations on their application.

We consider a collision in which a particle of rest mass, m_1, and kinetic energy, E, collides with an initially stationary particle of rest mass, m_2, and new particles of aggregate rest mass M (in addition to m_1 and m_2) are created. E is smallest when all particles are at rest after the collision in the center of mass-coordinate system (in which the center of mass of m_1 and m_2 is at rest before the collision). Application to this problem of the relativistic relation between energy and momentum leads to an expression for the threshold kinetic energy:

$$E=Mc^2(m_1+m_2+\tfrac{1}{2}M)/m_2. \qquad (1)$$

Equation (1) can be solved for Mc^2, which is the energy that is available in the center of mass system for particle creation, reactions, etc.:

$$Mc^2=[2m_2c^2E+(m_1+m_2)^2c^4]^{\frac{1}{2}}-(m_1+m_2)c^2.$$

In the non-relativistic limit ($E\ll m_1c^2$ and m_2c^2), this reduces to the usual expression $m_2E/(m_1+m_2)$.

If the colliding particles are protons ($m_1=m_2\equiv m$) and a pair of neutrons or a positive and negative proton are created ($M=2m$), Eq. (1) shows that the threshold energy is 6 $mc^2\cong5.6$ Bev. If an electron or a photon ($m_1\cong0$) collides with a proton, the threshold for nucleon-pair production is 4 $mc^2\cong3.7$ Bev. The corresponding thresholds for meson-pair production are higher than would be expected from non-relativistic theory by about 5 percent in the case of protons and about 10 percent in the case of electrons or photons.

Equation (1) indicates that a considerable reduction in the thresholds for nucleon-pair production would occur if the target particle were a heavy nucleus rather than a proton. This is actually true only if the target nucleus remains intact after the collision. We now show that the transfer of a large amount of momentum to the nucleus is very likely to break it up. The recoil momentum q of m_2 is given by:

$$(q/c)^2=\frac{2M(m_1+\frac{1}{2}M)(m_2+\frac{1}{2}M)(m_1+m_2+\frac{1}{2}M)}{(m_1+m_2+M)^2}.$$

In the case of a heavy nucleus ($m_2\ll m_1$, $m_2\ll M$), $q\sim mc$ for nucleon production either by protons or electrons. The region in which the creation process takes place has dimensions of the order of $\hbar/mc\sim10^{-14}$ cm. This is so small that only one of the constituent nucleons of the target nucleus is likely to be involved. Thus the nucleus remains intact only if this nucleon transfers the momentum q to the remainder of the nucleus.

The fraction of the events in which this occurs is of the order of the fraction of all collisions between a nucleon of momentum q, and a nucleus in which the nucleon is scattered at a large angle and the nucleus absorbs most of the momentum. Such a scattering process can be treated by the Born approximation. The cross section for back scattering is roughly equal to

$$(m^2/\hbar^4)\left|\int e^{i\mathbf{k}\cdot\mathbf{r}}V(\mathbf{r})d\mathbf{r}\right|^2, \quad \text{where} \quad k\sim(q/\hbar), \qquad (2)$$

and the total scattering cross section is approximately the geometrical nuclear cross section R^2, where R is the nuclear radius; here, $V(\mathbf{r})$ is the interaction potential between nucleon and nucleus. Equation (2) is roughly equal to $(m\bar{V}/\hbar^2)^2(\hbar/q)^6\sim(1/R_0{}^4)(\hbar/q)^6$, where $\bar{V}$ is the average nuclear potential, and R_0 is the range of nuclear forces. We are thus led to expect that the target nucleus remains intact in about $(\hbar/qR_0)^4(\hbar/qR)^2\sim10^{-7}$ of those collisions in which nucleon creation occurs, and is energetically possible with a proton target (the corresponding fraction for meson creation is considerably larger). This indicates that the effect is extremely improbable, and especially difficult to observe since newly created neutrons (although perhaps not negative protons) would tend to be masked by fragments of nuclei broken up by ordinary collisions.

McMillan and Teller[1] have pointed out that the kinetic energy of the nucleons inside a nucleus lowers the threshold. If this energy is assumed to be not greater than 30 Mev, it can be shown that the thresholds for nucleon-pair production are reduced to 4.1 Bev for protons and 2.6 Bev for electrons or photons. Since very few of the nucleons have the maximum kinetic energy, the cross section will be small just above the threshold. A simple estimate based on the Thomas-Fermi model indicates that the effective threshold is half-way between the figures given here and those given in the third paragraph. However, the difficulty of observing the effect in the presence of a background of nuclear fragments might make it worth while to work with a hydrogen target at the higher thresholds.

* Now at Stanford University.

[1] W. G. McMillan and E. Teller, Phys. Rev. **72**, 155(A) (1947).