

The Uranium 234 Content of Natural Uranium and the Specific Alpha-Activities of the Isotopes

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(Received August 1, 1949)

The uranium 234 content of natural uranium was determined, by a combination of alpha-counting and mass spectrometer techniques, to be 0.005481 ± 0.000012 weight percent, the precision being expressed on the basis of 95 percent probability. This corresponds to a half-life of $(2.522 \pm 0.008) \times 10^5$ years. Considering possibilities of bias, these figures are believed to lie within one percent of the true values. In conjunction with this determination, the half-lives of the other natural uranium isotopes were also measured.

PREVIOUS determinations of the weight percent of uranium 234 in natural or "normal" uranium have all involved measurement by the mass spectrometer. Nier first used such an instrument to determine the relative abundance of uranium 238 and uranium 234 in normal uranium.¹ Chamberlain, Williams, and Yuster used two related methods.² The first method was identical in principle to Nier's method in that they used the mass spectrometer for a direct measurement of normal uranium. In the second method the specific activity of uranium 234 was determined from the total alpha-activity and relative isotopic abundance of enriched uranium samples, assuming the specific activities of uranium 238 and 235 from the data of others. From their data on the alpha-activity of enriched samples, together with a specific activity figure for uranium 238,³ it is possible to deduce a value for the content of uranium 234 in natural uranium by computation of the uranium 234 alpha-emission enrichment factor.

By using pure samples of uranium 234 grown from UX_1 , Knight, Goldin, P. A. Macklin, and R. L. Macklin of this laboratory have determined the specific activity of uranium 234 from which they calculated, using the specific activity of uranium 238 given in this report, the weight percent of uranium 234 in normal uranium. Their normal abundance figures of 0.005805 ± 0.00008 weight percent is significantly different from that reported in this paper, but the cause of the difference is not yet known.

This report is concerned with a re-determination of the weight percent of uranium 234 in normal uranium by a method similar to the second method used by Chamberlain, Williams, and Yuster, and also an independent re-determination of the specific alpha-activities of the three isotopes. The uranium 234 content of uranium highly enriched in the 234 isotope can be determined with greater accuracy by the use of the mass spectrometer than that achieved in measurements on normal uranium, since the ratio of the uranium 234 to the sum of uranium 235 and 238 in greatly enriched uranium has been increased many times. The alpha-

activities of the enriched and normal uranium can then be measured by the use of counting equipment.

EXPERIMENTAL

Apparatus

The counting apparatus consisted of a parallel plate ionization chamber, utilizing electron collection in an atmosphere of nitrogen. The chamber was connected to an amplifier, discriminator, and scaler. The correction for coincident losses on this equipment was of the order of 0.8 percent at 100,000 random counts per minute.

The mass spectrometers used in the present work were of the modified Nier design, as commercially produced by the General Electric Company. The resolution between the ion beams from the 235 and 234 isotopes was such that the height of the valley was less than twenty percent of the height of the 234 voltage peak resulting from ion collection.

Measurement of Alpha-Activity by Alpha-Counting

Uranium samples were prepared for alpha-counting by electroplating a thin film of uranium oxide on an electro-polished nickel disk according to the method described by Cohen and Hull.⁴

The alpha-emissions were counted from films of various weights of uranium 238 and normal uranium, according to a random schedule. The counting rates were corrected for background and expressed as counts per minute per milligram of oxide per one-half solid angle.

Least squares parabolas were placed through each set of data, which permitted the extrapolation to zero weight film thickness for the specific activities of uranium 238 and normal uranium. These specific activities are expressed as oxides with an estimated geometry factor of 50 percent. In order to minimize the bias that might be introduced by a single counter, the specific activities were determined on eight counters.

A correction of 0.4 ± 0.2 count per minute was made on the specific activity of uranium 238 for the 0.05 ± 0.03 p.p.m. of uranium 234 contained in it. All precisions are

¹ A. O. Nier, *Phys. Rev.* **55**, 150 (1939).

² Chamberlain, Williams, and Yuster, *Phys. Rev.* **70**, 580 (1946).

³ G. T. Seaborg and I. Perlman, *Rev. Mod. Phys.* **20**, 637 (1948).

⁴ B. Cohen and D. E. Hull, MDDC-387 (October 18, 1946).

TABLE I. Weight percent of uranium 234 in normal.

Sample number	Natural abundance of U ²³⁴
1	0.005487
2	0.005468
3	0.005487
4	0.005475
5	0.005488
Av.	0.005481±0.000012(±0.2%)

expressed on the basis of 95 percent confidence belts. By placing a visual straight line through a plot of Crawford's back-scattering values against atomic numbers,⁵ a correction factor for back-scattering of alpha-particles by the nickel was estimated to be 1.26 ± 0.06 percent. Since the counting chamber detects the alpha-particles within an angle of 180°, a counting efficiency value of 50.63 percent was applied to all specific activities. The alpha-disintegration rate of natural uranium metal was then calculated to be 1502 ± 1.5 disintegrations per minute per milligram. The specific activity of uranium 238 was calculated to be 742.7 ± 1.6. Following the assumption of radioactive equilibrium between uranium 238 and 234 in normal uranium, the alpha-activity of uranium 234 in normal was equal to that of uranium 238 in normal, or 737.4 ± 1.6 disintegrations per minute per milligram of normal. The remaining 27.2 ± 3.5 disintegrations per minute were assumed to be due wholly to the alpha-activity of uranium 235. The alpha-activity of uranium 235 in normal was then divided by the weight fraction of 235 in normal and a specific activity of 3824 ± 490 disintegrations per minute thus calculated.

Uniform weight films of uranium oxide, prepared from each of four enriched samples and from a fifth sample which was a gravimetric dilution of one of the four, were alpha-counted, and the rates corrected for coincident losses and self absorption in the films. The alpha-counting enrichment factor, A_c , was determined by the formulation of a ratio of the counting rates of enriched uranium to normal uranium as R_c/R_n .

Measurement of the Uranium 234 Content of Uranium Highly Enriched in the 234 Isotope

Using two or more spectrometers, at least forty measurements for uranium 234 and 235 were made of each of the four uranium samples highly enriched in the isotope 234. All weight percents of uranium 234, or e values, were determined to precisions better than one percent.

The Relation of Isotopic Weight Percents and Specific Activities

The following equation relates the alpha-activities of normal and enriched uranium to their isotopic con-

TABLE II. Specific activities and half-lives of uranium isotopes.

Isotope	Alpha-activity in normal per mg per min.	Alpha-activity of isotope per mg per min.	Half-life (expressed in years)
U ²³⁴	737.4 ± 1.6	(1.345 ± 0.004) × 10 ⁷	(2.522 ± 0.008) × 10 ⁵
U ²³⁵	27.2 ± 3.5	3824 ± 490	(8.8 ± 1.1) × 10 ⁸
U ²³⁸	737.4 ± 1.6	742.7 ± 1.6	(4.49 ± 0.01) × 10 ⁹
Normal	1502.0 ± 1.5		

tents:

$$\frac{e}{e_n} = \frac{R_c - R_n^{235}(f/f_n) - R_n^{238}(100 - f - e)/(100 - f_n - e_n)}{R_n^{234}}, \quad (1)$$

where e = weight percent of uranium 234, f = weight percent of uranium 235, R = alpha-disintegration rate per mg of uranium metal, A_c = relative enrichment in alpha-counting rate over normal or R_c/R_n , subscript c = enriched uranium, subscript n = normal uranium.

Using this expression and the data obtained from measurements on the five enriched samples, the natural abundance of uranium 234 is calculated and summarized in Table I.

DISCUSSION OF RESULTS

The value of 0.005481 ± 0.000012 weight percent of uranium 234 in normal cannot be shown to be significantly different from Nier's value of 0.00574 ± 0.00057. Taking into consideration sources of possible bias in spectrometer and counting measurements, it is estimated that the value and the half-life calculated from it are accurate to at least one percent.

The half-life for uranium 234 of (2.522 ± 0.008) × 10⁵ years is somewhat less than the (2.69 ± 0.27) × 10⁵ value of Nier, and the (2.67 ± 0.04) × 10⁵ value of Knight, *et al.*, but is greater than the two values of (2.29 ± 0.14) and (2.35 × 0.14) × 10⁵ years as published by Chamberlain, Williams, and Yuster.² Since these values have been calculated using approximately the same alpha-activity, the main discrepancy is then in the weight percent of uranium 234 in normal. The half-life found for uranium 238, given in Table II, is in close agreement with Seaborg's and Perlman's values of 4.498 and 4.51 × 10⁹ years.³

The half-life of uranium 235, given in the preceding table, is not in good agreement with Nier's value of 7.07 × 10⁸ years¹ but is in good agreement with Clark, Spencer-Palmer, and Woodward's value of 8.91 × 10⁸ years as quoted by Seaborg and Perlman.³

Since uranium belongs to the actinium series of radio-activity, a comparison in normal uranium of the alpha-activity of the uranium 235 to the alpha-activity of uranium 238 gives the ratio of the actinium series to the uranium series. The value of 3.69 ± 0.48 percent obtained was somewhat lower than that of Nier's value of 4.6 percent but in good agreement with that of

⁵ J. A. Crawford, Report AEC-D 2034 (June 8, 1948).

Clark, Spencer-Palmer, and Woodward's value of 3.63 percent, obtained with an alpha-ray analyzer. While there are some discrepancies between alpha-activity values, the author feels that since the values determined are dependent on the alpha-activities of both uranium 238 and normal uranium, which are in very close agree-

ment with accepted values,^{3,6} the precisions quoted are reasonable.

This document is based on work performed for the Atomic Energy Commission by Carbide and Carbon Chemicals Corporation, at Oak Ridge, Tennessee.

⁶ A. F. Kovarik and N. I. Adams, *J. App. Phys.* **12**, 296 (1941).

Comparison of Calculations on Cascade Theory*

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(Received July 25, 1949)

The diffusion equations of the cascade theory of electron showers are solved using improved mathematical methods. The results obtained agree essentially with those obtained earlier by Carlson and Oppenheimer, by Snyder, and by Serber, but do not agree with those obtained by Bhabha and Chakrabarty. Solutions are given corresponding to a single incident electron, a single incident γ -ray, and a $1/E$ spectrum of γ -rays. The total number of particles expected under a given thickness of material are given in tables for various incident energies and each of the above initial conditions. Other tables and formulas are included which enable one to calculate the spectrum of the particles and γ -rays for various initial conditions.

I. INTRODUCTION

SINCE the original papers on the theory of cascade showers by Bhabha and Heitler¹ and by Carlson and Oppenheimer² other contributions have been made by Snyder,³ Landau and Rumer,⁴ Serber,⁵ Iyengar,⁶ and Bhabha and Chakrabarty.⁷ The work of Bhabha and Heitler was carried out without including ionization loss and is thus limited to the high energy portion of the spectrum. The calculations of Carlson and Oppenheimer did include ionization loss, but used simplified asymptotic forms for the high energy cross sections. They also replaced the integral equations of cascade theory by a simplifying differential equation. Their solution does not satisfy boundary conditions exactly, but provided the energy of the incident particle is sufficiently large, the error is insignificant. The work of Snyder was an extension of that of Carlson and Oppenheimer, using the same cross sections, but using the integral equations. This solution did not satisfy exact boundary conditions. A difficulty in this work which has been emphasized by Bhabha and Chakrabarty was that a certain function was computed only for integral values of its argument, the non-integral values being obtained by graphical interpolation. Soon after, Landau and Rumer showed

that the exact forms for the asymptotic cross sections produced no essential complication. Serber then continued the series of calculations as they were begun by Carlson and Oppenheimer and extended by Snyder, but also with the reservation that the incident energy be large enough so that the errors in the boundary conditions are small. Still later, Iyengar gave a complete solution in which the exact Bethe-Heitler cross sections were used; a solution from which it is not easy to obtain numerical values according to Bhabha and Chakrabarty. At about this same time, Bhabha and Chakrabarty also gave in series forms, an exact solution of the diffusion equations using asymptotic forms for the cross sections.

One of the major difficulties in this work has been to express the solution in such a form that numerical values can be obtained with reasonable ease. This has been particularly true for the total number of particles present at a given thickness and for the energy spectrum of the particles and γ -rays at low energies. Although Bhabha and Chakrabarty have given a series solution which converges even for zero energy, the first two terms in their expansion account for only about seventy-five percent of the total energy dissipated in a shower. On the other hand, the forms of solution as given by Carlson and Oppenheimer, by Snyder and by Serber, give in a single term the total energy dissipated in a shower. The objective of the work being reported in this paper is: (a) to give exact representations for all values of its arguments of the function which had previously been computed only for integral values of its argument, (b) to give a solution of the diffusion equations which satisfies the correct boundary conditions,

* Research carried out at Brookhaven National Laboratory under the auspices of the AEC.

¹ H. J. Bhabha and Heitler, *Proc. Roy. Soc.* **159**, 432 (1937).

² J. F. Carlson and J. R. Oppenheimer, *Phys. Rev.* **51**, 220 (1937).

³ H. Snyder, *Phys. Rev.* **53**, 960 (1938).

⁴ L. Landau and Rumer, *Proc. Roy. Soc.* **166**, 277 (1938).

⁵ R. Serber, *Phys. Rev.* **54**, 317 (1938).

⁶ K. S. K. Iyengar, *Proc. Ind. Acad. Sci.* **A15**, 195 (1942).

⁷ H. J. Bhabha and S. K. Chakrabarty, *Phys. Rev.* **74**, 1352 (1948).