B using U series

$$B(z, y) = \int_{0}^{w} e^{-\mu(w-x)} U dx = \sum_{n=-\infty}^{+\infty} b_{un}, \qquad (A17)$$

$$b_{un} = e^{(2n-1)y+z} \left[P\left(-\sqrt{z} - n\frac{y}{\sqrt{z}} + \frac{1}{2}\frac{y}{\sqrt{z}} \right) - P\left(-\sqrt{z} - n\frac{y}{\sqrt{z}} \right) \right]. \quad (A18)$$

B using V series

$$B(z, y) = \sum_{n=0}^{\infty} b_{vn}, \qquad (A19)$$

$$b_{v0} = (1 - e^{-y})/y, \qquad (A20)$$

$$b_{vn} = \frac{2y((-1)^n - e^{-y})}{y^2 + \pi^2 n^2} e^{-\pi^2 n^2 z/y^2}.$$
 (A21)

These series have been used in calculating the curves of Figs. 2 and 3; the U series used for $z < y^2$ and the V series for $z > y^2/9$ the region of overlap serves as a check on the calculations.

References of Tables

¹¹ W. Seith and E. A. Peretti, Zeits. f. Elektrochem. 42,

¹² G. v. Hevesy, W. Seith and A. Keil, Zeits. f. Physik
⁷⁹ (1932).
¹³ W. Seith, E. Hofer and H. Etzold, Zeits. f. Elektrochem. 40, 322 (1934).

14 W. Seith and J. G. Laird, Zeits. f. Metallkde. 24, 193 (1932).

¹⁵ W. Seith and H. Etzold, Zeits. f. Elektrochem. 40, 829 (1934).

¹⁶ W. Seith, Zeits. f. Elektrochem. 41, 872 (1935).

 ¹⁷ B. V. Rollin, Phys. Rev. 55, 231 (1939).
 ¹⁸ A. Sagrubskij, Physik. Zeits. Sowjetunion 12, 118 (1937).

¹⁹ H. A. C. McKay, Trans. Faraday Soc. 34, 845 (1938).
 ²⁰ W. Seith, Zeits. f. Elektrochem. 39, 538 (1933).

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PHYSICAL REVIEW

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Natural Radioactivity of Lutecium

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The half-life of the natural radioactivity of lutecium reported by Heyden and Wefelmeier is $2.9\pm0.5\times10^{12}$ years for the element or $7.3\pm2\times10^{10}$ years for Lu¹⁷⁶, which probably is the active isotope. Negative electrons are emitted. The upper energy limit of the spectrum is $215,000 \pm 15,000$ electron volts.

`HE bearing of the natural radioactivities of the ordinary elements on nuclear physics and geophysics makes it desirable to have data on their characteristics which are as accurate as those for the shorter-lived artificial radioactivities. It is the purpose of this paper to present data obtained on the natural radioactivity of lutecium (cassiopeium) reported by Heyden and Wefelmeier.¹

The work of Gollnow² on the hyperfine structure of lutecium had shown the presence of an unreported isotope of about 2.5 percent abundance. Recently Mattauch and Lichtblau³ investigated the element with a mass spectrograph and found Lu¹⁷⁶ present to 2.5 ± 0.07 percent of the element. Since both ytterbium and hafnium have isotopes of mass 176, this makes the Lu¹⁷⁶ case somewhat similar to that of K⁴⁰ and makes a determination of the sign of the beta-radiation of considerable interest.

The lutecium used was kindly furnished by Professor B. S. Hopkins of the University of Illinois. It was pure except for traces of thulium and ytterbium.

PROOF THAT THE ACTIVITY BELONGS TO A RARE EARTH ELEMENT

A consideration of the possible impurities (other than elements of low atomic number which would have to be present in large amounts to affect the data) reveals that all of those emitting beta-radiation of approximately the same

¹ M. Heyden and W. Wefelmeier, Naturwiss. 26, 612 (1938).

² H. Gollnow, Zeits. f. Physik 103, 443 (1936).

³ J. Mattauch and H. Lichtblau, Zeits. f. Physik 111, 514 (1939).



FIG. 1. Data obtained with proportional counter.

energy as that from lutecium either will come to equilibrium with alpha-emitting products or will be precipitated with PbS. The activity of the original Lu₂O₃ was compared with that of the purified compound and found to differ less than five percent. The purification was conducted by dissolving the Lu₂O₃ in dilute HNO₃, adding $Pb(NO_3)_2$, and saturating the solution with H_2S . The Lu_2O_3 was prepared from the filtrate from the PbS separation.

The search for alpha-radiation was made by using a screen wall counter^{4, 5} with a wire 0.22millimeter in diameter which allowed proportional counting to be done by lowering the voltage applied across the counter. The points in Fig. 1 show the data obtained. The ordinates are the counts observed when a Lu₂O₃ sample is placed over the counter. The abscissae are voltages below a standard value at which both alpha- and beta-radiation counted. The upper curve for alpha-radiation was obtained by means of a polonium source and a mica window in the counter chamber and the lower curve shows the effect due to gamma-radiation from a thorium source. This is characteristic of beta-radiation,

legatives Pac

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| 4 | W. | F. | Libby, | Phys. | Rev. | 46, | 196 | (1934). |
|---|----|----|--------|-------|------|-----|-----|---------|
| 5 | W. | F. | Libby, | Phys. | Rev. | 55, | 245 | (1939). |

Positives Pass.

10

RATE (NO PER MIN)

30 L

since the gamma-radiation ejects photoelectrons which count. Evidently any alpha-radiation present must be less than about ten percent of the total radiation. This fact, together with the PbS test shows that the activity must belong to a member of the rare earth group.

PROOF THAT THE ELECTRONS ARE NEGATIVELY CHARGED

Flat vanes running parallel to the counter axis and inclined so their surfaces made an angle of about 30° with the plane through the center of the vane and the counter wire (cf. reference 4) were inserted between the sample cylinder and screen wall counter. This device allows the sign of the beta-radiation to be determined by observation of the variation of the counting rate when the direction of the magnetic field is changed. Fig. 2 presents the results. The upper curve is drawn through the data obtained when the field direction was chosen to help negative electrons pass through the vanes and the lower curve is that obtained when the field was reversed to pass



positive electrons. The results show that the radiation is predominantly negative in character, the uncertainty amounting to a possible twenty percent. The conclusion is that the process occurring is

$$Lu^{176} \rightarrow \beta + Hf^{176}$$
.

ENERGY OF RADIATION

Figure 3 presents data on the aluminum absorption curve for the radiation. The Lu₂O₃ sample was placed around the screen wall counter and various thicknesses of foil were placed around the screen wall. Correction was made for the absorption by the gas in the counter (0.20 mg/cm^2) . Apparently the end-point is at 50 ± 5 mg/cm² of aluminum. From the data of Eddy⁶ and Varder⁷ on the ranges of cathode rays of various energies in aluminum, this gives $215,000 \pm 15,000$ electron volts as the upper energy limit of the lutecium spectrum. This procedure seems to be the most reliable method for interpreting absorption data on beta-spectra with upper limits below about 700,000 ev (cf. reference 5).

A determination of the upper limit by use of the screen wall counter in a magnetic field was made also. Fig. 4 gives the data representing the counting rate as a function of the current in the magnet coils. The end-point is at 50 ± 5 amperes which corresponds to 2000 ± 200 gauss and gives 1725 ± 173 as the H_{ρ} value for the upper limit of the beta-spectrum (cf. reference 5). This sets $212,000\pm18,000$ ev as the upper energy limit. Since the magnetic and absorption methods agreed, the end-point appears to be 215,000 $\pm15,000$ ev. The value agrees well with the value



⁶ C. E. Eddy, Proc. Camb. Phil. Soc. **25**, 50 (1929). ⁷ R. W. Varder, Phil. Mag. **29**, 726 (1915).



of 45μ of Al for the absorption half-thickness given by Heyden and Wefelmeier.¹

HALF-LIFE DETERMINATION

The specific activity was determined by using thin layers of Lu₂O₃ (obtained by moistening with ethyl alcohol, spreading smoothly on the sample cylinder, and evaporating the alcohol). The number of counts per minute per unit weight of Lu_2O_3 was plotted against the average thickness of the samples. Fig. 5 shows the data. A curve of the same shape as that found experimentally for potassium and rubidium⁸ (corrected for differences in the penetrating powers of the radiations) was drawn through these points and used as the specific activity without self-absorption. The factor for geometrical arrangement and counting efficiency was determined by substituting potassium chloride and calculating from the known weight of potassium used and the known decay constant. The result (8.3 percent counted) agreed within the limits of measurement with that calculated for the geometrical factor alone, showing that the counting efficiency was near one hundred percent. The specific activity of 34 ± 6 counts per minute per 296 mg of Lu₂O₃ obtained gives

$$34 \times (199/0.296) \times (1/0.083) = 2.75 \times 10^{5}$$

per mole per minute as the activity of lutecium element. Since Lu¹⁷⁶ (2.5 percent) is probably the isotope responsible this corresponds to 1.1×10^7 per minute per mole of Lu¹⁷⁶ which gives 7.3 ± 2 $\times 10^{10}$ years as the half-life of Lu¹⁷⁶. This figure agrees well enough with that of 4×10^{10} years given by Heyden and Wefelmeier.

The author wishes to thank Professor B. S. Hopkins for the Lu_2O_3 which he so generously furnished for this research.

⁸ W. F. Libby, Phys. Rev. 46, 745 (1934).