

## The Half-Life of Actinouranium and the Problem of Geologic Time<sup>1</sup>

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The actinium series arises from an isotope  $U^{235}$ , or from two such isotopes  $U^{239}$  and  $U^{235}$ , genetically connected. From work of Hahn and Meitner, the first possibility is almost certainly right. Assuming as a working hypothesis that  $U^{235}$  is the only long-lived uranium isotope, equations are developed for determining its decay constant  $\lambda_4$  from data concerning radioactive minerals and substances.

Branching ratio:	0.03	0.04
$\lambda_4$ in $yr.^{-1}$	$2.28 \times 10^{-9}$	$1.79 \times 10^{-9}$
$\lambda_1$ in $yr.^{-1}$	$0.1514 \times 10^{-9}$	$0.1509 \times 10^{-9}$

The best value for the half-life of  $U^{238}$  is  $(4.58 \pm 0.09) 10^9$  yr. If  $U^{239}$  exists and has a half-life long compared with that of  $U^{235}$ , the values for  $\lambda_4$  would apply to  $U^{239}$ . The ages, insensitive to the value of the branching ratio, are: Karlshus bröggerite,  $0.81 \times 10^9$  yr.; Wilberforce uraninite,  $1.04 \times 10^9$  yr. Since quantitative study of chemical alteration may throw light on the process and prove helpful in

These equations also give the decay constant  $\lambda_1$  of  $U^{238}$  and mineral ages. The results depend on the value of the actinium "branching" ratio, which lies between 0.03 and 0.04. Computations are carried out using both of these extreme values. Four minerals are discussed but only two, Karlshus bröggerite and Wilberforce uraninite, yield reliable results. The mean values from these are:

Branching ratio:	00.3	00.4
Half life of $U^{235}$ , yr.	$3.10 \times 10^8$	$3.96 \times 10^8$
Half life of $U^{238}$ , yr.	$4.58 \times 10^9$	$4.60 \times 10^9$

determining decay constants and mineral ages, equations are developed to show the effect of uniform leaching. Rates of removal or addition of Pb, U and Th are assumed different, but each of them is supposed constant in time. The resulting equations are applied to Katanga pitchblende, to illustrate the method.

### I. THE PRESENT STATUS OF THE ACTINOURLANIUM HYPOTHESIS

IT is generally accepted that the actinium series contains at least one member which is an isotope of uranium. Actinium has always been found in uranium minerals when search for it has been made. Further, the ratio of actinium to uranium is not far from constant and its deviation from constancy may be reasonably explained as a result of alteration of these minerals. Hahn and Meitner<sup>2</sup> found that protoactinium slowly accumulates in pure uranium compounds. Their observations show that UY is derived from uranium; the problem which remains is to determine the details of their relationship.

The earlier view that the actinium series originates by branching of the uranium series was discredited by Aston's<sup>3</sup> mass-spectrograph

studies of common and radiogenic leads in which he showed that some uranium leads contain amounts of  $Pb^{207}$  too large to come from any source other than the actinium series.

This means we must adopt the hypothesis of Piccard,<sup>4</sup> that the actinium series contains one or more uranium isotopes, genetically independent of  $U^{238}$  and  $U^{234}$ . Piccard supported a scheme of disintegration involving two so-called actinouranium isotopes, related through intermediate  $\beta$ -rayers in the same way as the isotopes  $U^{238}$  and  $U^{234}$  (Fig. 1). Similar schemes involving two genetically connected isotopes,  $U^{239}$  and  $U^{235}$ , have been suggested by several workers but have never been supported by strong evidence. Hahn<sup>5</sup> has made extensive experiments to detect  $\beta$ -radiations which would be expected to occur in the hypothetical sequence of disintegrations connecting  $U^{239}$  and  $U^{235}$ . He found no evidence of such radiations. It is fair to conclude from his

<sup>1</sup> Preliminary notes: Phys. Rev. **42**, 903 (1932); **43**, 205 (1933) and **43**, 781 (1933).

<sup>2</sup> Hahn and Meitner, *Berichte deutschen Chem. Ges.* **54**, 69 (1923).

<sup>3</sup> Aston, *Nature* **120**, 224 (1927); **123**, 313 (1929); **129**, 649 (1932); *Proc. Roy. Soc.* **A140**, 535 (1933).

<sup>4</sup> Piccard, *Arch. Science Phys. et Nat.* **44**, 161 (1917). See also T. R. Wilkins, *Nature* **117**, 719 (1926); and *Phys. Rev.* **29**, 352 (1928).

<sup>5</sup> Hahn, *Zeits. anorg. Chem.* **147**, 16 (1925).

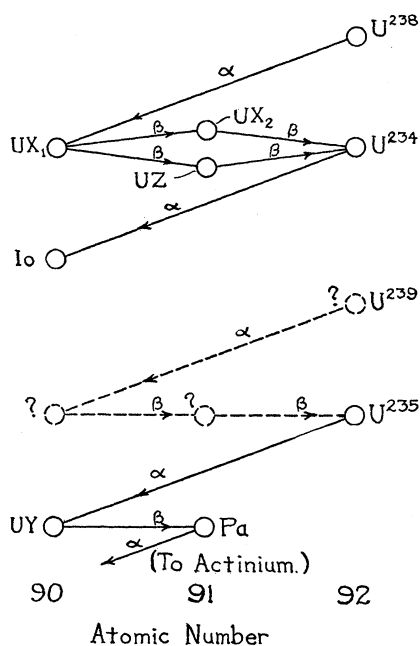


FIG. 1. Connection of the uranium and actinium series.

results that (1) either  $U^{239}$  does not exist; or (2) if it does exist, and if its immediate products are  $\beta$ -rayers, isotopic with  $UX_1$  and  $UX_2$ , these products must be so short-lived as to escape detection in spite of careful search.

In the light of our knowledge of the half-lives of  $UX_1$ ,  $UX_2$ , and  $UZ$ , the first-mentioned possibility appears much more probable than the second. If it is correct, then  $U^{235}$  is the parent of the actinium series. Let us assume, as a working hypothesis, that this is true. Then from existing data, it is possible to obtain an approximate value of the decay constant of  $U^{235}$  and thereby improve our knowledge of that of  $U^{238}$ ; the ages of certain minerals can also be obtained.<sup>6</sup> This done, we may consider how the results would be modified in the improbable event that both  $U^{239}$  and  $U^{235}$  exist. Decay constants of both isotopes cannot be rigorously determined at present because there are more unknowns than equations, but *if* one of the isotopes has a much longer half-life than the other, the previous analysis will furnish a good approximation to the decay constant of the more stable isotope. There are indications

<sup>6</sup> The calculations are based on the orthodox radioactive families (Fig. 1).

that *if* both isotopes exist,  $U^{239}$  will have a much longer life than  $U^{235}$ . Fajans<sup>7</sup> has pointed out that for isotopic  $\alpha$ -rayers the half-life increases with atomic weight, RaF being the sole exception.

We shall often speak, for convenience, as though the existence of a single actinouranium isotope,  $U^{235}$ , were an established fact. The reader is asked, therefore, to bear the above remarks in mind.

## II. NOTATION; DISCUSSION OF THE ACTINIUM BRANCHING RATIO

The subscripts 1, 2, 3 and 4 refer to the uranium isotopes  $U^{238}$ ,  $U^{234}$ , the hypothetical  $U^{239}$  and  $U^{235}$ , respectively, while the subscripts Pa, Ac, Th, and Pb refer to the elements of which they are the symbols. Our notation is:

$N_1, N_2$ , etc.—numbers of atoms of the respective isotopes of U, per gram of U now present.

$N$ —the sum of  $N_1, N_2, N_3$  and  $N_4$ .

$N'_{206}, N'_{207}, N'_{208}$ —numbers of atoms of the respective isotopes of Pb, per gram of U now present.

$N_{206}, N_{207}, N_{208}$ —numbers of atoms of these isotopes produced by radioactive disintegration.

$N_{Pb}, N_{Th}, N_L$ —numbers of atoms of the respective elements, and of common lead, per gram of U now present.

$\lambda_1, \lambda_2$ , etc.,  $\lambda_{Pa}, \lambda_{Ac}, \lambda_{Th}$ —decay constants of uranium isotopes, etc.

$f_{206}, f_{207}, f_{208}$ —fractions of  $N_{Pb}$  (total lead) constituted by  $Pb^{206}$ , etc.

$a_{206}, a_{207}, a_{208}$ —fractions of  $N_L$  (common lead) constituted by  $Pb^{206}$ , etc.

U/Pb, U/Th—ratio of mass of U to mass of Pb, etc.

$W_1, W_2$ , etc.—atomic weights on the chemical scale, 0 = 16.

The actinium branching ratio,  $R$ , is defined by

$$R \equiv \lambda_{Ac} N_{Ac} / E, \quad (1)$$

where  $E$  represents one-half the number of  $\alpha$  particles emitted per second by all the isotopes in one gram of uranium. Kovarik and Adams<sup>8</sup> found that  $E = 12,385$   $\alpha$ -particles/sec. per gram of U, with an uncertainty of 1 percent. The use of the ratio

$$B \equiv \frac{\lambda_4 N_4}{\lambda_1 N_1} \equiv \frac{\lambda_{Ac} N_{Ac}}{\lambda_1 N_1} \equiv \frac{\lambda_{Pa} N_{Pa}}{\lambda_1 N_1}, \quad (2)$$

<sup>7</sup> Fajans, *Le Radium* 10, 171 (1913); *Radioelements and Isotopes*, p. 31, McGraw-Hill Book Co., N. Y., 1931.

<sup>8</sup> Kovarik and Adams, *Phys. Rev.* 40, 718 (1932).

rather than  $R$ , simplifies many equations, so we shall adopt it. The available values show a considerable spread and we can only state<sup>9</sup> that with high probability  $B$  lies between 0.03 and 0.04. Calculations will be made using both these limiting values.

III. EQUATIONS FOR DETERMINING HALF-LIVES OF URANIUM ISOTOPES, AND MINERAL AGES

On the working hypothesis adopted here, that the actinium series contains only one long-lived uranium isotope, we have just enough data to calculate its decay constant and the ages of certain minerals. The relations involved are as follows.

Since the intermediate members of the uranium and thorium series are so short-lived as to be present in negligible amounts, we may write

$$N_{206} = N_1(e^{\lambda_1 t} - 1); \quad (3) \quad N_{208} = N_{Th}(e^{\lambda_{Th} t} - 1). \quad (4)$$

If the actinium series originates in  $U^{235}$ ,<sup>10</sup>

$$N_{207} = N_4(e^{\lambda_4 t} - 1). \quad (5)$$

Also,

$$\lambda_2 N_2 = \lambda_1 N_1; \quad (6) \quad \lambda_4 N_4 = B \lambda_1 N_1. \quad (7)$$

The observed quantities of the lead isotopes consist of original and radiogenic lead, as indicated by the equation,

$$N'_{206} = a_{206} N_L + N_{206}, \quad (8)$$

and two similar relations, (9) and (10), giving  $N'_{207}$  and  $N'_{208}$ . From the definition of Avogadro's number,

$$N_1 W_1 + N_2 W_2 + N_4 W_4 = 6.064 \cdot 10^{23}, \quad (11)$$

and from the experiments of Kovarik and Adams,

$$\lambda_1 N_1 + \lambda_2 N_2 + \lambda_4 N_4 = 2(3.9083 \times 10^{11}) \text{ per year.} \quad (12)$$

These equations contain ten unknowns:  $N_1$ ,  $N_2$ ,  $N_4$ ,  $\lambda_1$ ,  $\lambda_4$ ,  $N_{206}$ ,  $N_{207}$ ,  $N_{208}$ ,  $N_L$ , and  $t$ .<sup>11</sup> It is easy to eliminate all unknowns except  $\lambda_1$ ,  $\lambda_4$  and  $t$ . With times in years, and decay constants in  $\text{yr.}^{-1}$ , the result is,

$$\lambda_1 = \frac{3.9083 \cdot 10^{11}}{6.064 \cdot 10^{23}(1+B/2)} W_1 \left( 1 + \frac{W_4}{W_1} B \frac{\lambda_1}{\lambda_4} \right); \quad (13)$$

$$(e^{\lambda_1 t} - 1) - B \frac{a_{206} \lambda_1}{a_{207} \lambda_4} (e^{\lambda_4 t} - 1) = \frac{N_{Pb}}{N} \left( f_{206} - \frac{a_{206}}{a_{207}} \cdot f_{207} \right) \left( 1 + B \frac{\lambda_1}{\lambda_4} \right); \quad (14)$$

$$(e^{\lambda_4 t} - 1) - \frac{f_{206}}{f_{208}} \frac{W_U}{W_{Th}} \frac{Th}{U} \left( 1 + B \frac{\lambda_1}{\lambda_4} \right) (e^{\lambda_{Th} t} - 1) = \frac{N_{Pb}}{N} \left( f_{206} - \frac{a_{206}}{a_{208}} \cdot f_{208} \right) \left( 1 + B \frac{\lambda_1}{\lambda_4} \right). \quad (15)$$

<sup>9</sup> Western and Ruark, Phys. Rev. **42**, 903 (1932); **43**, 205 (1933).

<sup>10</sup> If the series originates in  $U^{239}$  and if  $U^{235}$  is short-lived compared to  $U^{239}$ , we would write

$$N_{207} = N_5(e^{\lambda_5 t} - 1);$$

and Eqs. (9) and (10) would contain additional terms.

<sup>11</sup> Walling found the half-life of  $U^{234}$  to be 342,500 years [Zeits. f. physik. Chemie **B10**, 467 (1930)]. Collie [Proc.

Roy. Soc. **A131**, 541 (1931)], concluded that this half-life is not less than  $10^6$  years, but it seems unlikely that it is much more than  $10^6$  years. The mass of  $U^{234}$  in uranium can be neglected in comparison with the masses of  $U^{238}$  and  $U^{235}$ , although its activity, of course, cannot be neglected. For our purpose, it is unnecessary to know  $\lambda_2$  exactly. The value of  $\lambda_{Th}$  was redetermined by Kirsch, who finds the half-life to be  $1.8 \cdot 10^{10}$  yr. [Phys. Zeits. **31**, 1718 (1930)]; also Kirsch and Lane, Proc. A. A. S. **66**, 357 (1931)].

In (14) and (15) we use the very close approximation,  $N/N_1 = 1 + B\lambda_1/\lambda_4$ , obtained by neglecting  $N_2$  as compared with  $N_1$  and  $N_4$ . It is convenient to write  $T = 10^{-9}t$ ,  $L_1 = 10^9\lambda_1/2.3026$ , etc., so that  $T$  is the mineral age in billions of years, and to rewrite the equations in a form adapted to solution by approximations. Inserting the values of coefficients of a general nature and replacing exponentials by powers of 10, we have<sup>12</sup>

$$L_1 = [1/(1+B/2)]0.06663(1+0.9874B(L_1/L_4)); \quad (16)$$

$$10^{L_1T} - 1 = (N_{\text{Pb}}/N)(f_{206} - 0.5600f_{208})(1+B(L_1/L_4)) + 0.5746(\text{Th}/\text{U})(10^{0.1672T} - 1)(1+B(L_1/L_4)); \quad (17)$$

$$\frac{L_4T}{10^{L_4T} - 1} = \frac{1.374BL_1T}{(10^{L_1T} - 1) - (N_{\text{Pb}}/N)(f_{206} - 1.374f_{207})(1+B(L_1/L_4))}. \quad (18)$$

#### IV. APPLICATION OF EQUATIONS TO MINERAL DATA

Table I gives the best values now obtainable for the isotopic and atomic weights of lead, uranium, and thorium.<sup>13</sup> The uncertainty of these figures is 0.03 atomic weight units, with the exception of that for common lead, where the uncertainty is 0.007 unit. Table II shows relevant portions of Aston's<sup>14</sup> mass analyses of leads, and Table III gives analyses of three of the minerals from which these leads were derived. The analysis of Katanga pitchblende is not available.

TABLE I. *Isotopic and atomic weights.*

Element or isotope	Atomic weight Chemical scale
Lead, 206	205.98
207	206.98
208	207.98
Ordinary lead, chemically	207.22
Thorium, calculated	232.03
Uranium, 234	234.04
235	235.04
238	238.04
239?	239.05
Uranium, natural mixture <sup>15</sup>	238.04

<sup>12</sup> To effect a solution, one neglects at first the terms containing  $B$ . By using a rough value of  $T$  in the thorium term, an approximate value for  $10^{L_1T} - 1$  is obtained from (17); this and the corresponding value of  $L_1T$  are substituted in (18), which gives  $L_4T$ .  $L_1/L_4$  is computed and is used in (16) to obtain a fair value of  $L_1$ .  $T$  is calculated from  $L_1T$  and the process is repeated. The convergence is good for uranium minerals and little would be saved by using approximate equations. A graph of  $x/(10^x - 1)$  facilitates the work.

<sup>13</sup> Western and Ruark, *J. Chem. Phys.* **1**, 717 (1933).

<sup>14</sup> *Proc. Roy. Soc. A140*, 535 (1933).

<sup>15</sup> Calculated on the hypothesis that  $\text{U}^{235}$  is the only actinouranium isotope by using an approximate value of the half-life of this isotope.

Of the minerals described, the thorite is not satisfactory for determining uranium decay constants because of the susceptibility of thorium

TABLE II. *Isotopic constitution of ordinary and radiogenic leads.*

Ordinary lead	Norwegian bröggerite	Katanga pitchblende	Wilberforce uraninite	Norwegian thorite
<i>Chemical atomic weight:</i>				
207.22	206.14 <sup>a</sup>	206.048 <sup>b</sup>	206.195 <sup>c</sup>	207.90 <sup>d</sup>
<i>Percentage 206:</i>				
27.75	86.8	93.3	85.9	4.6
<i>Percentage 207:</i>				
20.20	9.3	6.7	8.3	1.3
<i>Percentage 208:</i>				
49.55	3.9	(0.02)	5.8	94.1
<i>Other isotopes:</i>				
2.5	0	0	0	0

<sup>a</sup> Raade, Norway. Calculated from isotope-percentages, and isotopic weights of Table I.

<sup>b</sup> Hönigschmid and Birckenbach, *Ber.* **56**, 1837 (1923); von Grosse and Kurbatow, quoted by Aston, *Proc. Roy. Soc. A140*, 535 (1933).

<sup>c</sup> Baxter and Bliss, *J. Am. Chem. Soc.* **52**, 4851 (1930).

<sup>d</sup> Fajans, *Sitz. Heidelb. Akad. Wiss.* **3** (1918); Hönigschmid, *Phys. Zeits.* **19**, 437 (1918); *Zeits. f. Elektrochemie* **25**, 91 (1919).

TABLE III. *Mineral analyses.*

	Norwegian bröggerite <sup>a</sup>	Wilberforce uraninite <sup>b</sup>	Norwegian thorite <sup>c</sup>
Percentage U	61.16	53.52	0.45
Percentage Th	4.38	10.37	30.10
Percentage Pb	8.02	9.26	0.35
Th/U	0.0716	0.194	66.1
Pb/U	0.131	0.173	0.78

<sup>a</sup> Fenner and Piggot, *Nature* **123**, 793 (1929).

<sup>b</sup> Wells, reported by Baxter and Bliss, reference c to Table II.

<sup>c</sup> Fajans, *Zeits. f. Elektrochemie* **24**, 163 (1918); *Phys. Zeits.* **19**, 438 (1918).

minerals to leaching and because of the small amounts of  $Pb^{206}$  and  $Pb^{207}$  in the mineral. Decay constants computed from this mineral are listed in Table IV as a matter of record but are devoid

TABLE IV. *Calculated decay constants and mineral ages.*

Thorite $B=0.03$	Bröggerite		Uraninite	
	$B=0.03$	$B=0.04$	$B=0.03$	$B=0.04$
0.34	0.805	Age (yrs. $\times 10^{-9}$ ) 0.809	1.036	1.041
0.1512	0.1514	$\lambda_1$ (per yr. $\times 10^9$ ) 0.1508	0.1515	0.1510
4.58	4.578	Half-life of $U^{238}$ (yrs. $\times 10^{-9}$ ) 4.595	4.574	4.590
12 $\pm$ 1	2.62	$\lambda_4$ (per yr. $\times 10^9$ ) 2.07	1.94	1.51
0.056	0.264	Half-life of $U^{235}$ (yrs. $\times 10^{-9}$ ) 0.334	0.357	0.459

of significance. The data on the uraninite and the bröggerite appear fairly trustworthy. It is well known that alteration of a uranium mineral increases the lead-ratio,<sup>16</sup>  $Pb/(U+kTh)$ . The uraninite, described in detail by Kirsch and Lane,<sup>17</sup> has a lead-ratio slightly higher than that of some samples from the same vicinity but the difference is not great enough to affect our calculations materially. Fenner and Piggot<sup>18</sup> described the bröggerite as being without evidence of alteration. Comparison of its analysis with Gleditsch's<sup>19</sup> analyses of five samples from the same mine shows that, while its uranium content is in agreement with the lowest value of uranium from these, both the thorium content and the lead content are only about ninety percent of the lowest values found for these quantities in the Gleditsch samples. The low thorium content may indicate alteration. If so, calculations based on the Gleditsch samples show that the maximum effect of leaching on the values calculated for  $\lambda_4$  and  $t$  from the Fenner-Piggot sample would be to raise the one, and lower the other about five percent.  $\lambda_1$  is insensitive to such alteration. We assume, accordingly, that the mineral is unal-

tered and base our computations on the Fenner-Piggot analysis.

Values calculated from the three minerals discussed above are given in Table IV. The agreement between the results from the bröggerite and uraninite is satisfactory. It is unfortunate that the required data are not available for more good uranium minerals, to indicate whether this agreement is genuine.<sup>20</sup>

Kirsch and Lane<sup>17</sup> have shown that the age of the Wilberforce uraninite is  $(1.03 \pm 0.10)10^9$  years.

## V. THE EFFECT OF LEACHING ON RADIOACTIVE AGE DETERMINATIONS

In recent years techniques have been developed for judging whether or not a mineral specimen is essentially unaltered.<sup>21</sup> The principal aims of such work have been the selection of unaltered specimens, the establishment of reliable lead-ratios and the attempt to understand why leaching increases the lead-uranium ratio in uranium minerals, while it decreases the lead-thorium ratio in thorium minerals.<sup>22</sup>

It is desirable to begin a quantitative study of leaching which, as our knowledge increases, may throw additional light on the process and prove an aid in determining the ages of certain minerals.

<sup>20</sup> The kolm of Grällhöggen, Sweden has attracted much attention. The atomic weight of the lead is 206.013. Aston (Proc. Roy. Soc. A140, 535 (1933)) obtained a single mass-spectrum which showed  $Pb^{206}$  and  $Pb^{207}$  but he states that this spectrum was unsuitable for photometry. Previously several writers calculated the composition of the lead on the assumption that it contains only  $Pb^{206}$  and  $Pb^{207}$ . The results depend markedly on the values used for the isotopic weights. By using the weights in Table I, such a calculation gives 96.7 percent and 3.3 percent, respectively, for these two isotopes. However, these values are so sensitive to the isotopic weights that with the limits of error placed on the latter, the value computed for  $\lambda_4$  can vary from about  $0.015 \times 10^{-9}$  yr.<sup>-1</sup> to  $3.6 \times 10^{-9}$  yr.<sup>-1</sup>. Without a more complete mass analysis, the kolm cannot give information on the decay constant of actinouranium. The mineral age is  $0.42 \times 10^9$  yr., with an uncertainty of 4 percent.

<sup>21</sup> Kirsch, *Geologie und Radioaktivität*, pp. 132-180. Work of Ellsworth, Todd, Davis, and Hecht and Körner is described by Holmes on pp. 324-8 and 365-6 of *The Age of the Earth*, Bulletin 80 of the National Research Council, Washington, D. C. (1931).

<sup>22</sup> Holmes, *Phil. Mag.* 1, 1055 (1926).

<sup>16</sup> Here  $k = \lambda_{Th} W_{208} W_U / \lambda_U W_{206} W_{Th}$ . By using Kirsch's value of  $\lambda_{Th}$ ,  $k = 0.26$ .

<sup>17</sup> Kirsch and Lane, Proc. A. A. S. 66, 357 (1931).

<sup>18</sup> Fenner and Piggot, Nature 123, 793 (1929).

<sup>19</sup> Gleditsch, Ark. Mat. Og Naturv. Kristiania 36 (1), 184 (1919); Norske Videns. Akad. Oslo I, Mat. Nat. Kl. No. 3 (1925).

It is possible to improve our understanding of the effects of alteration by considering three hypothetical cases:

(1) *Initial alteration.* In this case practically no radiogenic lead is present when leaching occurs so the effect of alteration is the same as that of a decrease in the initial concentrations of U and Th.

(2) *Recent alteration.* Here the composition of the lead is almost unaffected. With a knowledge of the value of  $\lambda_4$ , isotopic analysis would make possible a correct age determination by equations (3), (5), (7), (8) and (9), even though the ratios Pb/U and Th/U were untrustworthy. This case may be realized in a rough way by some uraninites; for Ellsworth,<sup>23</sup> discussing uraninites of Villeneuve, says, "The lead ratio fortunately increases only slightly so long as the mineral remains black due to an appreciable UO<sub>2</sub> content, but once the UO<sub>2</sub> is entirely oxidized, uranium is lost much faster than lead, and the lead ratio rises to a large and entirely misleading value."

(3) *Uniform alteration.* It may be that for some minerals, the net effect of alteration is nearly the same as if it had taken place at a uniform rate. The simplicity of this assumption recommends it as a first approximation. If, in a radioactive series, the subscripts  $p$  and  $d$  refer to the long-lived parent element and the end-product, respectively, and if the intermediate elements can be neglected, uniform leaching is defined by

$$\begin{aligned} dN_p/dt &= -(\lambda_p + k_p)N_p; \\ dN_d/dt &= \lambda_p N_p - k_d N_d, \end{aligned} \quad (19)$$

where  $k_p$  and  $k_d$  may be called alteration constants. If we put  $k = k_p - k_d$ ,

$$N_d = [\lambda_p / (\lambda_p + k)] N_p (e^{(\lambda_p + k)t} - 1). \quad (20)$$

Application of (20) to each of the radioactive series gives three equations containing  $t$ ,  $k_U - k_{Pb}$  and  $k_{Th} - k_{Pb}$  as unknowns. Thus  $t$  can be obtained directly but in order to use these equations in finding alteration constants, we need the analysis of the mineral, the isotopic composition of the lead it contains and the analysis of an

unaltered specimen from the same deposit, if there be such.

As an illustration, the method may be applied to the Katanga pitchblende of Table II, by making some assumptions of uncertain validity. The lead was separated from a composite sample of altered material by Hönigschmid and Birckenbach.<sup>24</sup> They made no mineral analysis, feeling that the lead-ratio would be without significance. However, data on pp. 367-368 of *The Age of the Earth*, show that for three altered specimens designated as *I*, *J* and *K*, the mean ratio of lead to uranium is 0.096, while for several unaltered specimens it is close to 0.080. We write (Pb/U)' for the latter ratio to distinguish it from the value for the leached specimen. Thorium is practically absent. Assuming these data are appropriate to the material of Hönigschmid and Birckenbach, we can write equations which determine  $\lambda_1$ ,  $\lambda_4$ ,  $k_U - k_{Pb}$  and  $t$ . Eq. (13) applies directly. For an unleached thorium-free mineral,

$$\frac{205.98 N_{206} + 206.98 N_{207}}{238.04 N} = \left(\frac{\text{Pb}}{\text{U}}\right)',$$

which may be written

$$\begin{aligned} e^{\lambda_1 t} - 1 &= \frac{238.04}{205.98} \left(1 + B \frac{\lambda_1}{\lambda_4}\right) \left(\frac{\text{Pb}}{\text{U}}\right)' \\ &\quad - \frac{206.98}{205.98} B \frac{\lambda_1}{\lambda_4} (e^{\lambda_4 t} - 1). \end{aligned} \quad (21)$$

Further, we have two equations obtained by applying (20) to the uranium and actinium series. The results are:  $\lambda_1 = 0.1514 \times 10^{-9}$  per year,  $\lambda_4 = 2.8 \times 10^{-9}$  per year,  $k_U - k_{Pb} = 0.64 \times 10^{-9}$  per year and  $t = 0.55 \times 10^9$  years. Only the value obtained for  $\lambda_4$  can yield any test of the assumption of uniform alteration. In this case the character of the data is such that, even if  $\lambda_4$  were accurately known, no definite conclusion could be drawn. It is believed that when  $\lambda_4$  is more accurately known and further mineral data are available, similar investigations may throw much light on alteration processes.

<sup>23</sup> H. V. Ellsworth, *Am. Mineral.* **15**, 455 (1930).

<sup>24</sup> Hönigschmid and Birckenbach, *Ber. d. D. chem. Gesellschaft*, p. 1837 (1923).

## VI. CONCLUSIONS

In view of our present knowledge, a value of three or four hundred million years for the half-life of the principal actinouranium isotope cannot be far from correct. Further study of the actinium branching ratio and complete data on additional minerals are needed to yield a more accurate value.

The value calculated for the half-life of  $U^{238}$  is relatively insensitive to that of actinouranium. Consequently, the uncertainty in the former is no larger than the uncertainty in our knowledge of the rate at which uranium gives off  $\alpha$  particles. Judging from the small mean deviation in the counting experiments of Kovarik and Adams, a liberal estimate of all uncertainties allows us to state the half-life of  $U^{238}$  as  $(4.58_4 \pm 0.09) \times 10^9$  years, assuming that  $U^{239}$  does not exist. Kovarik and Adams<sup>8</sup> gave  $4.52_4 \times 10^9$  years. This was computed on the branching hypothesis and the value used for the atomic weight of uranium was 238.17, which is too

high. Our value should be considered as replacing that of Kovarik and Adams, since we have employed their data on the rate of emission of  $\alpha$  particles by uranium, and have taken account of  $U^{235}$ . The value of the International Radium-Standards Commission,<sup>25</sup>  $4.4 \times 10^9$  years, is not comparable with ours. The Commission states that no account was taken of the actinium series, because the half-life of actinouranium was not sufficiently well known.

The  $U^{235}$  content of uranium lies between 0.15 and 0.35 percent, provided  $U^{239}$  does not exist. The wide variation given is largely due to uncertainty in our knowledge of the branching ratio. On the other hand, if  $U^{239}$  exists and is long-lived in comparison to  $U^{235}$ , the same figures would apply to the former. A content of  $U^{239}$  ten times higher than the larger value given here would not explain the present atomic weight, 238.14, assigned to uranium.

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<sup>25</sup> Rev. Mod. Phys. 3, 427 (1930).