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FIVE FOLD CHECK OF URANITE AGE?

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The object of this paper is to show that with sufficient physical and chemical data it will be possible to have more than a five fold check on the age of a mineral disintegrating like uraninite, and that even now the range of possible age consistent with the data is limited.

That atoms of uranium and thorium disintegrate is now well known. The general theory has been treated in a popular way by A. Holmes¹ and by myself,² in scientific detail by Hevesy, Paneth, and Lawson,³ and in Bulletins 51 and 80 of the National Research Council, so that I need only briefly to outline certain facts to which I wish especially to refer.⁴

If one places a glass plate slightly sticky with castor oil, having a film of a sensitive zinc sulphide on its underside, over a mineral containing uranium and thorium, one will see in the dark with a pocket lense flashes like fireflies.⁵ These are due to particles of helium (less an electron), known as alpha particles, thrown off by the explosion of atoms of the radioactive elements, which strike the film of zinc sulphide and make it glow. One milligram of uranium, which weighs as much as 3.7 mm. square of fair weight letter paper will throw off 94 flashes a second. Of these 23.7 seem to be due to the explosion of uranium I and II, and most of the others to the successive changes (Bulletin 80, pp. 133-136) through ionium,

¹ Age of the Earth, Harper Bros.

² *Scientific Monthly*, April, 1931; *Scientific American*, Feb., 1930, p. 111.

³ Radioactivity. Oxford Press.

⁴ Since the presentation of this paper (December 29, 1933), I have had the advantage of an extensive mimeographed report by Prof. A. C. Ruark, and of seeing the recent papers and correspondence with O. B. Muench, J. P. Marble, H. S. Spence, T. R. Williams, H. R. Bishop, C. S. Piggot, W. D. Urry, and A. von Grosse; all of which has been used in the final revision.

⁵ Several minutes are needed to sensitize the eye in the evening, ten minutes or more in the daytime.

radium, etc., to a stable lead. But according to A. von Grosse⁶ for every 100 atoms which explode and pass through uranium, radium, etc., to lead, there are four which pass through protactinium and actinium. In the former case he gives final lead the atomic weight $205.96 \pm .02$,⁷ in the latter $207.010 \pm .01$. According to von Grosse this proportion is now the same for all minerals⁸ independent of geologic age, geographic origin, thorium or uranium concentration, but he assumes, following Rutherford, that Ur-actinium disintegrates about ten times more rapidly than the atoms which change into ionium so that of course, there was a larger proportion in times past.

The number of atoms of any radioactive element disintegrating seems to depend on the number present, and experiments through many degrees of temperature and atmospheres of pressure in various compounds show no variation. Thus this ratio generally designated as lambda (λ) is characteristic of an element. It may be represented, if dN indicates the number changing in a year and N the number present, as dN/N .

The lambda (λ) for uranium is⁹ about $1.535 (10)^{-10}$ if the year, according to Kovarik, is taken as the unit. Since in a gram of uranium there are about $1/(1.65(10)^{-24}.238)$ atoms, the dN is small enough relative to N to apply the integral calculus formula $\int dN/N = \log. N$, and if t_0 is the time of formation, and N_0 the number of atoms (then), and t the time of observation (at the present), and N the number of atoms at the present, and if U_0 is the weight of uranium at the beginning and U is the weight of uranium at the time of observation, then $U_0/U = N_0/N$. Also $\lambda t - \lambda t_0 = (\log. N - \log. N_0) = -\text{Nat. log. } N/N_0 = -\text{Nat. log. } U/U_0$. But the number of atoms N is $U/1/(1.65(10)^{-24}.238+)$.¹⁰

⁶ *Physical Review*, November 15, 1932, pp. 565-570, with references to previous literature. I wish heartily to thank him for the opportunity to see his paper in advance of its publication in preparing mine.

⁷ But Alter and Baxter find in the Bedford cyrtolite (*J. Am. Chem. Soc.*, 55, p. 1447, 1933) a lead with the atomic weight of but 205.92, and in the *Physical Review* for January, 16 isotopes of lead and of uranium are reported.

⁸ According to Wildish variable, but always small, and in the papers in the *Physical Review* just mentioned the order of abundance of the isotopes is 206 and 207 or 208.

⁹ 1.471 in National Research Council Bulletin 80, p. 86; 1.535 in the report of the Committee on the Measurement of Geological Time for 1932.

¹⁰ The weight of 1 hydrogen atom is $1.66(10)^{-24}$ grams, but its atomic weight is more than 1 depending on whether O is taken at 16, as usual, or allowance made for

The original number of atoms is:

$$N_0 = U(1/(1.65(10)^{-24} \cdot 238 +) + Pb(1/1.65(10)^{-24} \cdot 206 \pm)$$

thus:

$$N_0/N = 1 + Pb \cdot 238/U \cdot 206$$

If the Pb is all Pb produced from Ra then for 206 we should write, according to von Grosse, 205.96. Now the age of the mineral = $t_0 - t = 1/\lambda$ Nat. log. U_0/U , but in place of λ is often written the value of $t_0 - t$ when the uranium is half gone, i.e., $U_0/U = 2$, which is called T , therefore, $T = 1/\lambda$ (Nat. log. $2 = .692$); $T\lambda = .692$. If also, in place of the natural logarithms which are 2.3 as great, we use the decimal logarithms, we have a formula:

$$\text{Age of minerals} = 3.323 T_U \log.(1 + 1.156 \text{ RaG}/U).$$

For thorium there is a similar formula: Age = $3.323 T_{Th} \log.(1 + 1.115 \text{ ThD}/\text{Th})$.

There are two uncertainties in the calculations, the rate of disintegration of uranium and also what proportion of uranium gives that disintegration of atoms which will turn into radium, and ultimately into RaG (Pb_{206}), and what into other isotopes. Isotopes present in very small quantities may contribute a much larger proportion of the disintegration if they are exploding at a rapid rate.

That the rate of change and disintegration, however, has not widely changed in time is made clear by the halo rings around radioactive particles. The exploding atoms send off alpha particles, helium bullets in minerals which enclose small radioactive particles which are likely to smash up molecules and produce change of color, and do more smashing near the end of their flight, just as a bullet at muzzle velocity will go through a window pane and make a small hole, but toward the end of its flight is likely to shatter the pane.

Each element, moreover, sends its alpha particles off with a velocity which on the average is characteristic of that element. And the more rapidly it explodes, the higher the velocity and the wider the range affected.

Now it appears that the range of these halos in very old rocks is, as Kerr Lawson has shown, nearly identical with those today. However, the rings in halos around radioactive material are deeper and more frequently noted in the older rocks, though they are discharged by heating.

I have found it convenient to refer to the uranium isotope parent of radium as Ur-radium, U_{Ra} , and the uranium isotope parent of

the fact that oxygen itself has isotopes and therefore should be given an atomic weight of something like 16.00022.

actinium as Ur-actinium, U_{ac} . We may also have to refer to Ur-thorium and Ur-virginium. It seems certain that Ur-radium is the main isotope U_{238} of which Aston¹¹ says that uranium is composed to the extent of at least 97 per cent. The relative amounts of the isotopes of uranium are not yet certain.¹² The atomic weight found is 238.14 and Elsen¹³ estimated that there should be up to 18 per cent of U_{239} which he considered the parent of the actinium series, and Kirsch¹⁴ suggested similar figures. This is of course inconsistent with Aston's result. On the other hand, von Grosse¹⁵ and Rutherford and Kovarik all depend on Aston and deduce almost negligibly small amounts of Ur-actinium (U_{239}). One thing at least seems certain. The more rapidly Ur-actinium explodes the less of it there is to give the ratio of four actinium atoms to a hundred of radium. Assuming this to be true, and that the radium series ends with Pb_{206} which the work of Baxter and his students practically has proved, and that thorium forms the Pb_{208} , then the actinium series would be responsible for the Pb_{307} , which Aston reports as the next most abundant isotope; and we have five ways of estimating the age: by the formation of $Pb_{206,207,208}$, and helium, and if we can from the oxygen estimate the uranium originally present we may compare that with the uranium now present. We will illustrate with the Wilberforce uraninite.

(1) AGE OF WILBERFORCE URANINITE BY UR-RADIUM

Kirsch and I showed that the amount of RaG, i.e., Pb_{206} , can not vary greatly in the Wilberforce lead in view of the amount of lead and the atomic weight of the lead determined. Our Fig. 1, here repeated with additions, shows that our conclusion fits nicely with Aston's¹⁶ estimates of the proportions of lead present.

If, as von Grosse thinks, Ur-actinium disintegrates ten times as quickly as the Ur-radium (Kovarik makes it not quite as fast, $2.7(10)^8$ years), there can be but 0.4% of the Ur-actinium. This leaves the excess of uranium atomic weight over 238 unaccounted for as above mentioned.

The highest amount of Ur-actinium suggested, based on the

¹¹ *Nature*, **128**, p. 725, 1931.

¹² Wilkins, T. R., *Nature*, Oct. 1932, and the *Physical Review*, Jan. 1933.

¹³ *Loc. cit.*, p. 286.

¹⁴ Radioaktivität und Geologie, p. 133, 200. See also Western and Ruark.⁹

¹⁵ *Physical Review*, **42**, pp. 42-43, 1932.

¹⁶ *Nature*, **129**, p. 649, 1932.

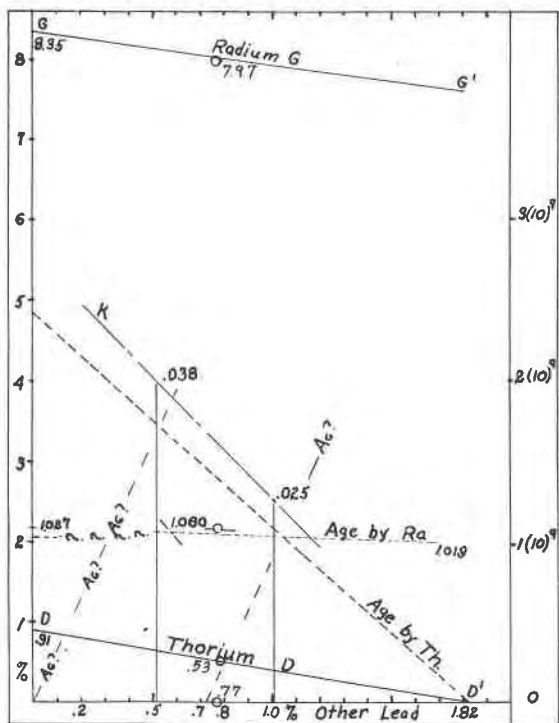


FIG. 1. (From p. 368, Vol. 66, *Proc. Am. Ac. of Arts and Sciences.*) Illustrates the computation of all possible ages of the Wilberforce uraninite consistent with the amount of uranium, thorium, and lead and the atomic weight of the latter. Possible amounts of lead not derived from radium and thorium are laid off as abscissas. Full lines join the tops of the ordinates which show the Radium G and Thorium D that correspond, and the scale is on the left. Lines with dashes show the ages corresponding. Their scale is on the right. In *Nature*, April 30, p. 649, 1932, Aston reports 85.9 of $Pb_{206} = RaG$, 83.0 of $Pb_{207} = AcD$, and 5.8 of Pb_{208} , which implies the percentages shown by the circles, and the age by the circle and dash which have been added to the original figure.

supposition that it is responsible for the excess of the atomic weight of U over 238, is by Elsen.¹⁷ A curve, labelled actinium Fig. 2, is plotted for all possible values of the amount of U_{ra} and the corresponding disintegration, and ages deduced. On the whole the curve shows that a short life and but little of Ur-actinium give an age that fits best the age given by U_{ra} . This latter age comes out much the same, whatever is assumed as to the Ur-actinium and is thus more likely to be correct.

¹⁷ *Receuil des Travaux chimiques des Pays Bas*, Feb. 1932, p. 286.

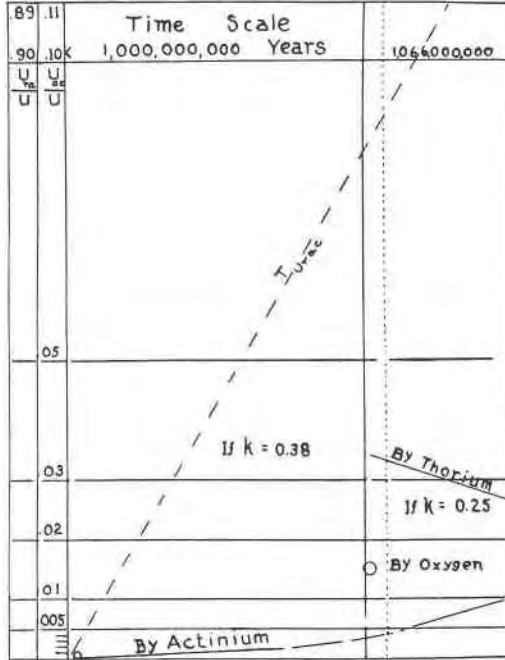


FIG. 2. Age of the Wilberforce Uraninite for various percentages of Ur-actinium: by radium G, short dashes (about 1075 million years); by thorium for various k factors from 0.25 to 0.38; by actinium, long dashes.

Also the half life of Ur-actinium T implied, on a scale reduced ten times. 427 million years fits best.

Obviously the more rapid the distintegration of the Ur-actinium the less will be needed to keep up the present supply of four atoms to 100. If the Ur-actinium decays at the same rate as the Ur-radium 4% would be enough. The rate of decay of Ur-radium is said to be such that its half life is¹⁸ something like $4.3(10)^9$ years. If Ur-actinium is so short lived as to be half gone in $4(10)^8$ years, there would be needed only $(4/43)$ a tenth as much.

If there is 0.004 of Ur-actinium as von Grosse says (p. 568) the Ur-radium is $53.52(1-0.004) = 53.31$. It can not be more than 53.52.

If von Grosse is right that the "activity ratio" (the ratio of distintegration) is four protactinium atoms to 100 radium atoms, then of the present distintegration studied by Kovarik only 96% is due to Ur-radium.

¹⁸ Kirsch and Lane, *Am. Ac. Sci.*, 66, p. 359, 1931.

Consequently, the number of particles thrown off by the .996 Ur-radium of the number noted by Kovarik will be

$$0.96 \times \frac{1.535(10)^{-10}}{.996} \text{ i.e., } 1.473(10)^{-10}$$

which means a half life $T_{\text{Urra}} = 4.7(10)^9$.

The age will then be $(3.323)4.7(10)^9 \log. (1 + 1.156 \times 7.97/53.31)$. In millions of years = $15600 \times 0.0689 = 1078$ million years. If we take a less rapid rate of disintegration of the Ur-actinium and a greater half life, we must assume T_{Urra} ¹⁹ almost proportionately shorter since the rate of disintegration of the whole U must be kept the same. But in that case there must be more Ur-actinium to make the proportions of atoms disintegrating still 4%, and therefore less Ur-radium in the denominator of the last term, and these two factors will about balance—will balance if $T_{\text{Urac}}:T_{\text{Urra}}$ is large enough.

The table appended shows ages for various assumed proportions of Ur-actinium. *We see from the table that on any hypothesis as to the Ur-actinium the age can not consistently with the physical and chemical data be far from 1070 million years.*

It is clear from the table that whatever the ratio of Ur-actinium assumed, from nothing to 11 per cent, there is only ten to fifteen million years difference in the age as figured by the radium. As we assume more of the Ur-actinium to be still present, its rate of decay must be assumed less and that of the Ur-radium increased so that this partly balances the greater ratio of Pb_{206} to the less amount of Ur-radium.

If there is, as appears (Fig. 2) most consistent, only a small amount of Ur-actinium present, then the rate of disintegration of the Ur-radium is about $1.475(10)^{-10}$ and the formula for age becomes in millions of years: Age = $15580 \log. (1 + 1.516 \text{ RaG/Urra}) = 1075$ million years.

(2) AGE OF WILBERFORCE URANINITE BY THORIUM LEAD—
 $\text{ThD} = \text{Pb}_{208}$

According to Aston's results and previous figures there is in the Pb 5.8 per cent of $\text{Pb}_{208} = 0.529 = \text{ThD}$.

¹⁹ T_{Urra} means the half life of that uranium isotope which is the parent of radium. T_{Urac} means the half life of that uranium isotope which is the parent of actinium.

TABLE OF COMPARATIVE AGES OF WILBERFORCE URANINITE BY RADIUM AND ACTINIUM FOR VARIOUS ASSUMED PROPORTIONS OF UR-
RADIUM TO UR-ACTINIUM BY SLIDE RULE. (Last figure not accurate. Base of Fig. 2.)

| 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 |
|------|-------|------|-------|------|-------|------|-------|--------|------|------|
| 1.00 | 1.474 | 4700 | 15600 | 1075 | 53.52 | 0.00 | ∞ | 0 | ∞ | 0 |
| .998 | 1.477 | 4691 | 15570 | 1075 | 53.4 | 0.12 | 29.5 | 0.0230 | 7.94 | 749 |
| .997 | 1.478 | 4680 | 15562 | 1075 | 53.36 | 0.16 | 19.7 | 0.420 | 5.56 | 946 |
| .996 | 1.480 | 4675 | 15550 | 1078 | 53.31 | 0.21 | 14.75 | .0469 | 4.23 | 1123 |
| .995 | 1.483 | 4670 | 15500 | 1080 | 53.2 | 0.32 | 11.8 | .0585 | 2.81 | 1135 |
| .99 | 1.491 | 4660 | 15430 | 1073 | 52.9 | 0.62 | 5.91 | .1170 | 1.45 | 1510 |
| .98 | 1.506 | 4600 | 15280 | 1073 | 52.4 | 1.12 | 2.95 | .2345 | .803 | 2000 |
| .97 | 1.521 | 4550 | 15140 | 1073 | 51.84 | 1.68 | 1.98 | .3500 | .535 | 2160 |
| .95 | 1.555 | 4450 | 14810 | 1072 | 50.8 | 2.72 | 1.18 | .5860 | .329 | 2410 |
| .90 | 1.640 | 4220 | 14050 | 1066 | 48.1 | 5.42 | .591 | 1.1700 | .165 | 2590 |
| .89 | 1.658 | 4175 | 13900 | 1066 | 47.6 | 5.92 | .536 | 1.2900 | .152 | 2640 |

- Column 1. Ratio of $\frac{U_{ra}}{U}$.
- Column 2. λU_{ra} required to supply 100 atoms of radium to 4 of actinium in equilibrium, λU being $1.535(10)^{-10}$ atoms per year.
- Column 3. $T_{Ura} = \text{half life} = \frac{0.692}{\lambda U_{ra}}$ in millions of years.
- Column 4. $\frac{2.3}{\lambda U_{ra}} = 3.323 T$.
- Column 5. Age in millions of years = $3.323 (T) \log \left(1 + 1.156 \frac{RaG}{U_{ra}} \right)$; $RaG = 7.97$; $1.156 RaG = 9.21$; $3.323T$ is given in Column 4.
- Column 6. Ur-radium, per cent of.
- Column 7. Ur-actinium, per cent of.
- Column 8. $\lambda U_{ra} = \frac{4}{104} \times 1.535(10)^{-10}$ (proportion of ur-actinium) in units of $(10)^{-10}$ atoms per year.
- Column 9. $T_{U_{ra}} = \frac{0.692}{\lambda U_{ra}}$ in billions of years.
- Column 10. $\frac{0.889}{U_{ra-actinium}}$. Actinium D = 0.77 (Aston).
- Column 11. Age by actinium from the formula: age in million of years = $3.323 \times T_{U_{ra}} \times \log \left(1 + 1.153 \frac{\text{Actinium D}}{U_{ra-actinium}} \right)$

Authorities differ on the rate of disintegration and of lead production and half life (T) of thorium.²⁰ Kirsch says it is a quarter of that of U. This would mean a life four times as long. Bulletin 80 (Kovarik) uses a factor 0.36; the International Critical Table 0.38. Thus, Age = 1020 (up to 1620) million years.

It must be remembered, however, that if uranium and thorium are composed of any isotopes of unequal rate of decay the rate will slow up as those more rapidly decaying disappear. So that the rate of production compared to that of uranium of lead by thorium (k) may not be the same now as it has averaged in the past. This will in part account for the discrepancy between Kovarik and Kirsch. If we take the age we found from Ur-radium from the previous section we can find what value of k will fit. To check perfectly with the age just obtained Ur-radium to lead, 1075^{±8} million years, we should have:

$$\text{Age} = 1075^{\pm 8} = 44,000 \log. \left(1 + 1.115 \frac{0.529}{10.37} \right) = 3.323 T_{\text{Th}}(.024-).$$

Therefore, $T_{\text{Th}} = 13,250(10)^6$, and $\lambda_{\text{Th}} = 0.522(10)^{-10}$. This indicates a $k = 0.36 = 0.522(10)^{-10} / 1.475(10)^{-10}$, just the value used in N.R.C. Bulletin 80.

(3) AGE OF WILBERFORCE URANINITE BY CHANGE FROM UR-ACTINIUM TO ACTINIUM—D = Pb₂₀₇

From Aston we have Pb₂₀₇ = 8.3 per cent of 9.26 = 0.77. This is about in the middle of the range of possibilities assigned by Kirsch and Lane (Fig. 1).

The age = 3.323 T_{Urac} log. (1 + 1.153 0.77/1.61 r) where r is proportional to the half life of the Ur-actinium and is defined as follows: $r = 4$ Urac/3 Ur_{ra}. If there was only 0.16 of Ur-actinium r would be 0.1 and T_{Urac} would be 0.075 of $T_{\text{Ur_{ra}}}$ and the age would be in millions of years 2.3/19.67(10)⁻¹⁰ log. (1 + 0.77/.161) = 956 million years. And if we take Ur-actinium as 0.2 = .00375 U

$$\lambda_{\text{Urac}} = \left(1.535(10)^{-10} \times \frac{4}{104} \right) / \left(\frac{0.2}{53.52} \right) = 15.79(10)^{-10}$$

and the age = 2.3/15.79(10)⁻¹⁰ log. (1 + 1.153(.077)/0.2) = 1073 million years. This checks much more closely with the age by Ur-radium and Thorium than the accuracy of the chemical analyses warrant, and may be taken as giving as accurate a value of the

²⁰ Int. Critical Tables I, p. 362, 1.31(10)⁻¹⁰ up to 2.37?

rate of disintegration of Ur-actinium as we have. The half life = $0.69/15.79(10)^{-10} = 4.3(10)^8$ years, which is near that derived by von Grosse from bröggerite.

The table and Fig. 2 show the results of assuming various proportions of Ur-actinium.

(4) AGE OF WILBERFORCE URANINITE BY OXYGEN

When Dr. Kirsch and I²¹ discussed the age of the Wilberforce uraninite, which had been described by Ellsworth and Spence and others, we did not attempt to compute the age by the oxygen method on account of uncertainties Ellsworth mentioned. Yet it seems as though it had a value in giving a maximum age, in freedom from uncertainties as to the atomic weights and relative properties of the different leads.

It is interesting to find that in spite of the large amount of lead J. T. Norton, Jr. finds the uraninite crystal lattice remains that of UO_2 . Therefore, we may naturally ask if the large atoms of oxygen have not retained their places, and the oxygen present, be that originally there.

We have therefore taken the analysis made by H. V. Ellsworth, one by F. Hecht on identically the same material as that used by Baxter and Bliss²² and by R. C. Wells²³ and another by the same analyst of a crystal from the same lot, obtained (by slide rule) the molecular proportions of all the U, Pb, Th, Ce, Y, Fe, and Al oxides, then found the proportions of the atoms of metal and of oxygen, Table 3. We find that the proportion of oxygen to metal is a little more than 2 to 1. But if we assume that the O of the PbO has really already been counted as O of the UO_3 and the summation of the analyses is high, then the ratio becomes as nearly 2 to 1, as different good analysts agree on the amount of U present.

It is conceivable then that from the O of the RO_3 one might infer the amount of UO_2 originally present = 7.44 O.

Yet it is probable that a little of the Fe_2O_3 and Al_2O_3 present were originally combined in some silicate like hornblende and some oxygen may be well be attributed to external oxidation. In his discussion of the present rate of disintegration of U, Kovarik²⁴ quotes

²¹ *American Acad. of Arts and Sciences*, Vol. 66, pp. 363-372.

²² *Jour. Am. Chem. Soc.*, 62, p. 4848, 1930.

²³ *Am. Acad. Arts and Sciences*, 66, p. 364.

²⁴ National Research Council Bull. 80, pp. 86 to 92. See also p. 208.

TABLE 3.
OXYGEN IN WILBERFORCE URANINITE

| | H.V.E. | Atom proportions | | Hecht & Wells | Atom proportion | | Hecht & Reich-Rohrwig | |
|---|--------|------------------|--------|--------------------|-----------------|--------|-----------------------|-------|
| UO ₂ | 39.10 | .145 U | .290 O | 62.12 ¹ | .2226 U | 69.10 | .3480 | .660 |
| UO ₃ | 32.40 | .1132 | .3396 | | 53.52 | | | |
| U | | | | 10.45 | .047 Pb | 10.80 | .049 | .049 |
| PbO | 10.95 | .049 | .1028 | 10.33 | .039 Th | 14.42 | .0554 | .1108 |
| ThO ₂ | 10.60 | .0514 | .03 | 2.04 | .01 ± | 2.60 | .01 | .03 |
| CeO ₃ | 1.88 | .01 | .03 | 1.64 | .02 ± | 1.49 | .02 | .03 |
| Y ₂ O ₃ | 2.14 | .02 | .008 | | | | | |
| Fe ₂ O ₃ | 0.43 | .005 | .003 | | | | | |
| Al ₂ O ₃ | 0.09 | .002 | | | | | | |
| Total metal | | .3956 | | | .33 (9) | | .3824 | |
| Total O | | | .8524 | | | | | .8798 |
| less for O of PbO | | | .8034 | | | | | .830 |
| for Fe ₂ O ₃ and Al ₂ O ₃ | | .3886 | .7924 | | .32 | | | |
| Ratio | | 2.07 | | | .65 | | .217 | |
| SiO ₂ | 0.19 | | | | 2.03 | | | |
| CaO | 1.01 | | | | | 0.36 | | |
| MgO | 0.08 | | | | | 0.85 | | |
| MnO | 0.03 | | | | | 0.25 | | |
| H ₂ O | 0.05 | | | | | 0.0 | | |
| H ₂ O | 0.65 | | | | | 0.0 | | |
| Insol. | 0.31 | | | | | | | |
| Sp. gr. | | 100.06 | | | | 100.60 | | |
| | | 9.062 | | | | | | |

¹ The UO₂ and UO₃ are not separated, but in the very same sample R. C. Wells found 53.52 U and von Grosse and Kurbatov 52.71, and Hecht's 62.14 of U₃O₈ gives 52.6. This sample was from a large 700 gram crystal used to obtain lead and uranium for atomic weights, and some fluorite was attached.

figures from 5.000 down and the International Critical Tables gives 4.670. The uncertainty is probably less than 5 per cent but shows that we can not, at present, expect an accuracy of one per cent in our calculations, and slide rule calculations are accurate enough.

The formula for age in millions of years becomes: (U_0 is uranium originally present and all isotopes are included)

$$\text{Age} = 4520 \times 3.323 \log. U_0/U$$

$$= 15,000 \log. U_0/U = 15,000 \log. 7.44 O/U$$

| | I | II | III | IV |
|----------|-----------------------|-----------------------|-----------------------|-----------------------|
| $UO_2 =$ | 39.10 | 45.18 | 13.55 | |
| $UO_3 =$ | $\frac{32.40}{71.50}$ | $\frac{24.90}{69.08}$ | $\frac{52.04}{65.59}$ | 61.96 |
| $U_0 =$ | $\frac{61.44}{10.06}$ | $\frac{60.56}{8.52}$ | $\frac{52.26}{10.33}$ | $\frac{53.52}{9.44}$ |
| $U_0 =$ | 74.6 | 63.5 | 77.0 | 70.4 |
| Age = | 1268 | 304 | 2460 | 1785 million years |

No. III is obviously oxidized and gives too high an age.

The result by No. I of Ellsworth checks fairly with that by Uranium Pb_{206} , the most reliable method (1070 million years). IV by Wells is not so far off.

In No. II one may suspect such troubles in the chemical method as Ellsworth describes. A small difference in the amount of oxygen and uranium found will make very large difference in the age computation. 1.3 per cent more O would make the results of Analysis II agree with I. As to the chemical work, Ellsworth writes (Nov. 30, 1932):

"When I first thought of using this method and Professor S. Beatty and I calculated the results for some of our analyzed uraninites we always found too much UO_3 (or not enough UO_2). This of course can be explained by assuming weathering, etc. Also it may be that the mathematics does not make proper allowance for an *acceleration* of oxidation that may be conceived to take place as time goes on due to U of UO_3 breaking up."

(5) AGE OF WILBERFORCE URANINITE BY HELIUM

W. R. Bennett found²⁵ in three 15-gram portions of the same sample of Wilberforce uraninite on which the atomic weight deter-

²⁵ *Proc. Am. Ac. Arts and Sciences*, 66, p. 372.

minations were made 15.605, 15.543, and 15.620 cc. of helium. This gives an average amount of 10.39 cc. of helium per gram of ore. The age, owing to the loss by diffusion, is always less in minerals with so much helium. The age Kirsch and Lane gave was, by an earlier approximate formula, $10.39/53.52 + 2.9 \times 9.10(10)^8 = 168$ million years. Bennett estimated 166.8 million years. But the ratios to the bröggerite which von Grosse studied are similar.

| | |
|--|----------------------------|
| The helium ratio for Wilberforce is | $\frac{.185}{.14} = 1.32$ |
| The helium ratio for Annerod bröggerite is | .14 |
| The lead ratio for Wilberforce is | $\frac{.165}{1.39} = 1.19$ |
| The lead ratio for bröggerite is | 1.39 |

Both indicate that the Wilberforce uraninite is older.

(6) AGE OF WILBERFORCE URANINITE BY PROPORTION OF ISOTOPES IN LEAD

If we assume that all uranium in the first place had the same proportion of isotopes and that their rates of disintegration varied, the older the lead the larger must be the proportion of the lead from the more rapidly disintegrating isotope, and as von Grosse points out we might find the age even if we did not have all the analytical figures to use the other methods, simply from the determination of the proportions of isotopes.

(7) AGE OF WILBERFORCE URANINITE BY SPECIFIC GRAVITY

Could we assume that spacing of the atomic network did not change from disintegration, loss of helium would show in the specific gravity which should decrease from 10.89 in a ratio $x270.14 + y238/270.14$ where x is the the proportion of UO_2 and y is the remaining Pb and O. It is true that the younger uraninites have greater specific weights up to that practically demanded by the crystal network. However, analytical methods do not suffice, since it is by no means certain that the network will remain unaltered.