THE HALF-LIFE OF Th^{232} AND THE BRANCHING RATIO OF Bi^{212}

By F. E. Senftle, T. A. Farley, and N. Lazar

Trace Elements Investigations Report 193

UNITED STATES DEPARTMENT OF THE INTERIOR
GEOLOGICAL SURVEY

IN REPLY REFER TO:

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WASHINGTON 25, D. C.

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June 1, 1956

Dr. T. H. Johnson, Director
Division of Research
U. S. Atomic Energy Commission
Washington 25, D. C.

Dear Dr. Johnson:

Transmitted herewith is one copy of TEI-193, "The half life of Th^{232} and the branching ratio of Bi^{212}," by F. E. Senftle, T. A. Farley, and N. Lazar, April 1956.

The authors plan to submit this report for publication in Physical Review.

Sincerely yours,

for W. H. Bradley
Chief Geologist

Nuclear Physics

This document consists of 12 pages.
Series A.

UNITED STATES DEPARTMENT OF THE INTERIOR

GEOLOGICAL SURVEY

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By

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ABSTRACT

The half-life of Th^{232} has been calculated by determining an absolute gamma disintegration rate for Tl^{208} in equilibrium with Th^{232} for three old thorium nitrate salts and one natural thorite sample. The branching ratio $\frac{\alpha}{\alpha + \beta}$, for Bi^{212}, a necessary parameter in the calculation, was also measured. The half-life of Th^{232} was found to be 1.42×10^{10} years which is essentially in agreement with the presently accepted value. The branching ratio, $\frac{\alpha}{\alpha + \beta}$, of Bi^{212} was found to be 0.362 ± 0.006, about 7.4 percent higher than the current value.

INTRODUCTION

It is well known that the geologic ages of rocks and minerals calculated from the Pb^{208}/Th^{232} ratio are generally lower by a considerable amount than the ages of the same specimens calculated from the Pb^{206}/U^{238} or Pb^{207}/U^{235} ratios. Although some of the discrepancies are probably due to inadequate techniques used to measure the ratios, recent age determinations of all three of the above ratios by several workers show that the Pb^{206}/U^{238} and Pb^{207}/U^{235} ages agree very well, whereas the Pb^{208}/Th^{232} ages are generally low.

For these reasons we have undertaken a redetermination of the half-life of thorium, whose currently accepted value of 1.39×10^{10} years was

measured by Kovarik and Adams 1/ in 1938. An absolute gamma-ray disintegration rate was determined for Tl^{208} in secular equilibrium with Th^{232}. Correction for the branching ratio gave the disintegration rate of thorium from which the half-life could be calculated. We have obtained a final result for the half-life of thorium, $T_1/_2 = 1.42 \times 10^{10}$ years and a branching ratio, $\frac{\alpha}{\alpha + \beta}$ for Bi^{212} of 0.362 ± 0.006. The half-life is essentially the same as that obtained by Kovarik and Adams, but the branching ratio is about 7.4 percent higher.

MATERIALS

Four different samples were used to make the determinations. Unlike the method of Kovarik and Adams, the methods used here do not require such a complete knowledge of the history of the sample. The only requirement is that the sample be in radioactive equilibrium with its disintegration products. The thorium series will be in equilibrium to better than 99 percent in 35 years. It was therefore necessary to procure thorium salts which were at least 35 years old for this method. The following samples were used in these experiments.

No. 2a Thorium nitrate (Eimer and Amend Inc.) purchased by the U. S. Geological Survey before 1905 and probably prepared in the early 1900's.

No. 3ab Thorite from Wet Mountain area, Custer County, Colorado. This material was extremely fresh and showed no signs of alteration. It is thought to be of early Tertiary age.

No. 4a Thorium nitrate (Messrs. Thorium, Ltd., London).
This salt was reported by C. F. Davidson 2/ of the Geological Survey and Museum

1/ A. Kovarik and N. Adams, Phys. Rev. 34, 413 (1938).
2/ Personal communication.

of Great Britain to have been prepared in Germany about 1906 or earlier.

No. 5a Thorium nitrate (Welsbach). This salt was purchased by the National Bureau of Standards in 1922, but it was probably prepared several years before this date.

For the purpose of this work, these four samples were considered to be in equilibrium; however, the results of the half-life determinations on the thorite sample, no. 3ab, did not compare with the other three samples. Subsequent radiochemical analysis of Ra^{224} in equilibrium with the Th^{232} indicated that 9.5 percent of the radium had been lost. The final half-life calculations have been corrected for this loss. Spectrographic analyses of the four samples are shown in Table 1.

The average thorium content of the nitrate samples was determined by weighing the samples after ignition to thorium oxide or by precipitation of thorium hydroxide from solutions of the salts followed by ignition to the oxide and weighing. The average analyses, in percent, for thorium of samples 2a, 4a and 5a are 41.78 $\pm$ 0.1 percent, 41.42 $\pm$ 0.2 percent, and 41.81 $\pm$ 0.1 percent, respectively. The thorite sample was analyzed after precipitation of thorium as the peroxynitrate 3/ followed by ignition to the oxide. The thorium content of sample 3ab was 40.8 $\pm$ 0.2 percent.

BRANCHING RATIO

As the method of measuring the half-life depends in a critical way on the fraction of transitions from Bi^{212} which decay through Tl^{208}, it was necessary to determine this fraction. A piece of platinum sheet was suspended over about 600 grams of shelf thorium nitrate for about 12 hours. The active

3/ F. S. Grimaldi and C. H. Warshaw, U. S. Geological Survey Bull. 1006, 165 (1954).

Table 1.--Spectrographic analyses of the thorium samples.

Percent	2a $Th(NO_3)_4$	3ab Thorite	4a 1/ $Th(NO_3)_4$	5a $Th(NO_3)_4$
Over 10	Th	Th, Si	Th	Th
5-10	--	--	--	--
1-5	--	Fe, Ca, Ba	--	--
0.5-1.0	--	--	--	--
0.1-0.5	--	Cu, Y, Ce, Nd	--	--
0.05-0.1	--	Al	--	--
0.01-0.05	La, Si, V	Sr, Sm, Dy, Pb, Mg, Mn, La, Zr, Gd	Fe, Si	Ca
0.005-0.01	Fe, B	--	La	Si
0.001-0.005	--	V, Er, Yb, Be	--	Fe
0.0005-0.001	Mg	--	Mg	Mg
0.0001-0.0005	--	--	--	--

1/ Radium analysis by the radon method for sample 4a is 1.65 + 0.05 x 10^{-10} gram/gram of thorium nitrate. Analyst, J. R. Dooley, U. S. Geological Survey.

deposit of Bi^{212} on the platinum was used as the source. This was placed in a 2π alpha proportional counter large enough that the entire path length of the highest energy particles was contained within the counting volume. The source was collimated with a 1/32 inch brass plate drilled with a large number of holes 0.06 inch in diameter. The chamber had a 3-mil wire center electrode operated at 1500 volts with 10 percent methane-argon counting gas, and the

output after amplification was fed into a multichannel pulse analyzer suitably modified to respond to "long" pulses. The two alpha peaks of Bi^{212} and Po^{212} corrected for background are shown in figure 1. The ratio $\frac{\alpha}{\alpha + \beta}$ in the decay of Bi^{212} was determined as 0.362 ± 0.006. This is about 7.4 percent larger than the value of 0.337 obtained by Kovarik and Adams.1/

EXPERIMENTAL METHOD

The intensity of the 2.62 Mev gamma ray (100 percent) from Tl^{208} was measured using a cylindrical crystal of NaI (Tl)--diameter 3 inches, height 3 inches. The sources were placed on the axis of the cylinder about 9.5 cm above the top face of the crystal. A polystyrene absorber, 1300 mg/cm^2 thick, was placed between source and crystal to insure absorption of all beta rays emitted in the thorium series.

The sources consisted of plastic cylindrical capsules, about 1 cm in diameter, filled with thorium salts, sealed, and stored several weeks to insure equilibrium in the thorium series. The maximum height of any of these sources, 1.3 cm, could be approximated by a point source, and the geometry was accurately defined by assuming the source was concentrated at the center.

The expression used for calculating the intensity of the 2.62 Mev gamma rays, I (2.62), from the measured peak areas, P (2.62), was

$$I(2.62) = \frac{P(2.62)}{T\epsilon_p(2.62)\Omega\left[1 - \epsilon_T(0.582)\Omega f_{0.582} - \epsilon_T(0.510)\Omega f_{0.510} - \epsilon_T(0.860)\Omega f_{0.860}\right]}$$

$\epsilon_T(E_\gamma)$ and $\epsilon_p(E_\gamma)$ are the total and peak efficiencies of the crystal, respectively, for gamma-ray energy E_γ. A complete description of the methods used for the determination of ϵ_T and ϵ_p as a function of energy is in preparation. In brief, the total efficiency for a given solid angle, Ω, subtended by the

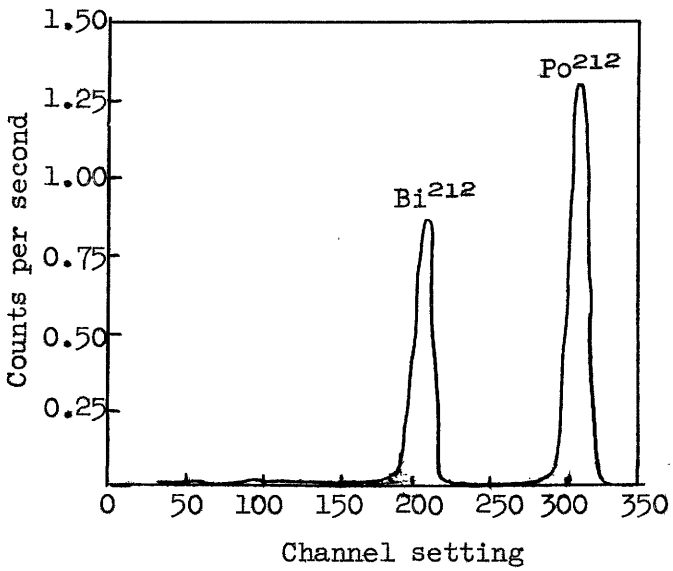

Figure 1.--Alpha spectrum of Bi^{212}-Po^{212} deposit.

crystal at the source is given by

$$\epsilon_T \Omega = \int_0^{\Omega} (1 - e^{-\tau x}) \, d\Omega$$

A numerical integration has been carried out with the assistance of the mathematics panel at Oak Ridge National Laboratory, and ϵ_T was determined for various energies and heights of the source above the crystal.

$\epsilon_p(E_\gamma)$, the probability that a gamma ray will be detected and give a full-sized pulse if it strikes the crystal, was obtained in the following manner: the pulse height spectrum from a number of radioactive sources which emit only one gamma ray was obtained with a cylindrical NaI crystal, and the areas under the full energy peak and total spectrum were evaluated. The ratio, R, of peak area to total area, when multiplied by ϵ_T is just ϵ_p. Care was taken, in the measurement of R, that extraneous effects which might distort the response of the crystal to the pure gamma rays, e.g., scattered radiation from the surroundings, bremsstrahlung, etc., were reduced to an absolute minimum. The curve of R vs energy was extended to higher energies by using sources which emit only two gamma rays. The contribution of the lower-energy radiation was subtracted from the total spectrum by measuring its peak area and making use of the value of R at its energy. A check on these values has been obtained by beta-gamma coincidence measurements and by determinations of R for single gamma rays obtainable from inelastic proton scattering in Si^{28} and B^{11}. The peak efficiency for 2.62-Mev gamma rays with the source placed 9.3 cm from the face of the crystal is 0.096.

The expression in the brackets in the denominator of equation (1) takes into account the coincident summing which occurs between cascade gamma rays. The factor, f_γ, is defined as the ratio of the intensity of the gamma rays with energy E_γ to the 2.62 Mev gamma-ray intensity. The values of f_γ used were

those given by Elliot and others.4/ Although other gamma rays cascade with the 2.62 Mev gamma ray, their intensities are too small to contribute appreciably to the intensity of the 2.62 Mev transition.

The factor, T, is inserted to take into account the interaction of the gamma rays with the polystyrene absorbers and any self-absorption in the source. For sources at distances large compared to the dimensions of the crystal, a gamma ray passing through this material is approximately directed parallel to the axis and a simple exponential absorption was assumed, i.e.,

$$T = e^{-\mu \bar{x}} + e^{-\mu' \frac{\ell}{2}}$$

where $\bar{x}$ is the thickness of the absorber in cm, and μ and μ' are the absorption coefficients in cm^{-1} of the absorber and sample, respectively. For the source absorption, it can be shown that for $\mu x \ll 1$ one may assume $\bar{x} = \frac{\ell}{2}$ where ℓ is the thickness of the source. Both of these corrections amounted to 5 percent of the intensity for the thickest sources.

Four samples were measured at heights -9.5 cm from the crystal and at a similar height above a crystal whose upper edge had been bevelled at 45° to the axis so that the upper diameter was 2 inches. The values for λ obtained with the bevelled crystal, table 2, agree among themselves and within 4 percent of the results with the unbevelled crystal; however, they are systematically high. It is felt that the efficiency of the unbevelled crystal is known with the higher accuracy and these values are quoted in our results.

Attempts were also made to obtain the intensity when the source was placed only 3.0 cm from the top face of the crystals. The peak areas in these measurements could not be obtained very accurately due to the inability to account

4/ L. G. Elliott and others, R. L. Graham, J. Walker, and J. L. Wolfson, Phys. Rev. 93, 356 (1954).

properly for the summing contribution under the peak itself. However, the results as given in table 2 are not in disagreement with the rough values obtained with the source at these closer distances.

From the intensities obtained in this way, the activity of the parent Th^{232} may be determined if the branching ratio of Bi^{212} is known. Using the new value for the branching ratio and the best values for I(2.62) we obtain $\lambda = 1.55 \times 10^{-18}$ sec^{-1}, or $T_{1/2} = 1.42 \times 10^{10}$ years with an expected uncertainty of about 5 percent.

Table 2.--Thorium decay constants as measured by bevelled and unbevelled crystals.

Sample no.	Thorium content of capsule (g)	Bevelled crystal I_γ	Bevelled crystal λ ($\times 10^{-18}$ sec)	Unbevelled crystal I_γ	Unbevelled crystal λ ($\times 10^{-18}$ sec)
2a	1.098_5	1693	1.64_0	1610	1.55_8
3ab	1.408_3	*	*	2155	1.54_7
4a	1.061_5	1613	1.61_7	1545	1.54_9
5a	0.445_0	679.1	1.62_4	646.0	1.54_8
		Average	1.62_7		1.55_0

*Not determined

The authors wish to acknowledge the assistance of their colleagues at the U. S. Geological Survey: Frank Cuttitta, who made the thorium analyses, J. N. Rosholt, Jr., for the radiochemical analysis, and C. L. Waring for spectrographic analyses. This work is part of a program being conducted by the Geological Survey on behalf of the Division of Research of the U. S. Atomic Energy Commission.