observed departures of the radius and surface parameters of the seven spherical nuclei by Hahn, Ravenhall, and Hofstadter, studied from smooth functions representing their general trends, are related to such fluctuations. If this is the case, it must be expected that the task of arriving at a general quantitative theory for predicting nuclear energy levels will be a truly formidable one, involving much greater complexities than those of self-consistent atomic field calculations. One might reasonably hope, however, that by starting with wave functions and potentials similar to those which underly this work, the convergence of such calculations would be rapid.

The writers would like to express their thanks to Mr. Prakash Sood, Mrs. Rose Nelson, Mr. Richard Gentry, Mr. William Stanley, and Mrs. Anna Lane for their assistance with the calculations and the preparation of the manuscript. One of us (A.G.) would also like to express his appreciation to the Oak Ridge National Laboratory for their hospitality during the summer of 1956 when the final parts of this work were completed.

PHYSICAL REVIEW VOLUME 104, NUMBER 6 DECEMBER 15, 1956

Half-Life of Th^{232} and the Branching Ratio of Bi^{212}†

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(Received July 9, 1956)

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, $\alpha/(\alpha+\beta)$, 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 within an estimated error of 5%, which is essentially in agreement with the presently accepted value. The branching ratio, $\alpha/(\alpha+\beta)$, of Bi^{212} was found to be 0.362 ± 0.006, about 7.4% higher than the currently accepted 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 using an absolute alpha-counting technique. In this work 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.

MATERIALS

Four different samples were used to make the determinations. Unlike the method of Kovarik and Adams, the methods used here do not require 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% in 35 years. It was therefore necessary to obtain thorium salts that were at least 35 years old for this method. The following samples were used in these experiments:

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

No. 3*ab*: 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. 4*a*: Thorium nitrate (Messrs. Thorium, Ltd., London). This salt was reported by Davidson[2] of the Geological Survey and Museum of Great Britain to have been prepared in Germany about 1906 or earlier.

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

For the purpose of this work, these four samples were considered to be in equilibrium; however, the results of

† Publication authorized by the Director, U. S. Geological Survey.

[1] A. Kovarik and N. Adams, Phys. Rev. 34, 413 (1938).

[2] C. F. Davidson (private communication).

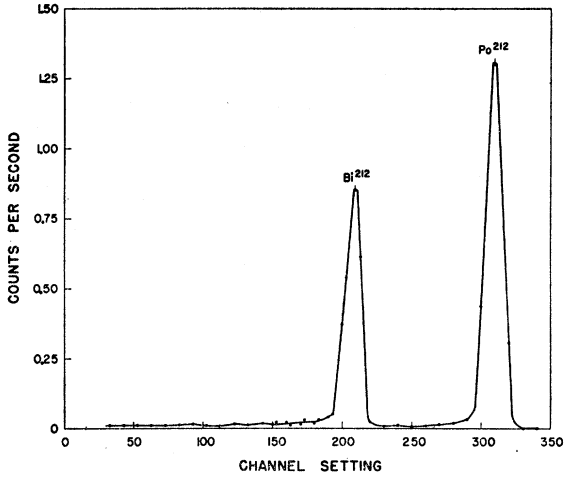

FIG. 1. Alpha spectrum of Bi^{212}-Po^{212} deposit.

the half-life determinations on the thorite sample, No. 3*ab*, did not compare with the other three samples. Subsequent radiochemical analysis of Ra^{224} in equilibrium with the Th^{232} indicated that 9.5% 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 I.

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 2*a*, 4*a*, and 5*a* were (41.78±0.1)%, (41.42±0.2)%, and (41.81±0.1)%, 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 3*ab* was (40.8±0.2)%.

BRANCHING RATIO

As the present 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 deposit of Bi^{212} on the platinum was used as the source. This was placed in a 2π alpha-proportional counter large enough so that the entire path length of the highest energy particles was contained within the counting volume. The source was collimated with a $\frac{1}{32}$-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% methane-argon counting gas, and the output after amplification was fed into a multichannel pulse analyzer suitably modified to respond to the "long" pulses from the chamber. The two alpha peaks of Bi^{212} and Po^{212} corrected for background are shown in Fig. 1. The ratio $\alpha/(\alpha+\beta)$ in the decay of Bi^{212} determined from the peak areas, is 0.362±0.006. This is about 7.4% 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%) from Tl^{208} was measured using a cylindrical crystal of NaI(Tl) three inches in diameter and three inches high. 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/cm² 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 was 1.3 cm, and the geometry was accurately defined by assuming that the source was concentrated at the center.

Figure 2 shows a typical pulse-height spectrum obtained from a sample of $Th(NO_3)_4$ placed at 3.0 cm from the crystal. 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

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

$\epsilon_T(E_\gamma)$ and $\epsilon_P(E_\gamma)$ are the total and peak efficiencies of the crystal, respectively, for gamma-ray energy E_γ. The methods used for the determination of ϵ_T and ϵ_P as a function of energy has been described elsewhere.[4] In brief, the total efficiency for a given solid angle, Ω, subtended by the crystal at the source is given by

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

A numerical integration has been carried out with the assistance of the Mathematics Panel at Oak Ridge National Laboratory, and ϵ_T has been determined for various energies and positions of the source above the crystal.

[3] F. S. Grimaldi and C. H. Warshaw, U. S. Geol. Survey Bull. **1006**, 165 (1954).

[4] Bell, Davis, and Lazar, Oak Ridge National Laboratory Report ORNL-1974, 1955 (unpublished); Nucleonics **14**, 52 (1956).

$\epsilon_P(E_\gamma)$, 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 that might distort the response of the crystal to the pure gamma rays, e.g., scattered radiation from the surroundings, bremsstrahlung, etc., was 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 the equation 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 Elliott *et al.*[5] 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.,

$$T=\exp(-\tau\bar{x})+\exp(-\tau'\bar{x}'),$$

where $\bar{x}$ and $\bar{x}'$ are the thickness in cm, and τ and τ' are the absorption coefficients in cm^{-1} of the absorber and sample, respectively. For the source absorption, it can be shown that for $\tau'\bar{x}'\ll 1$ one may assume $\bar{x}'=l/2$ where l is the thickness of the source. Both of these corrections amounted to 5% 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 beveled at 45° to the axis so that the upper diameter was 2 inches. The values for λ obtained with the beveled crystal, Table II, agree among themselves and within 5% of the results with the unbeveled crystal; however, they are systematically high. The efficiency of the unbeveled 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

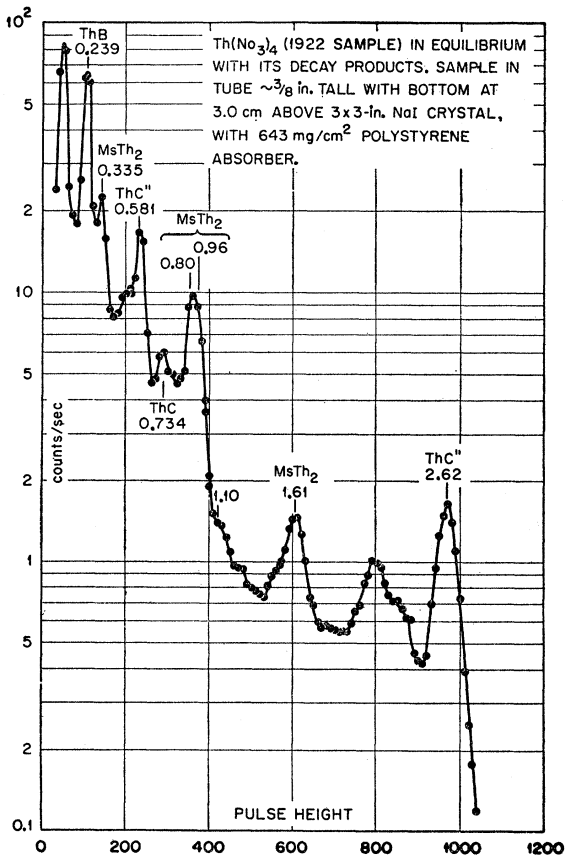

FIG. 2. Pulse-height spectrum of $Th(NO_3)_4$ in equilibrium with its decay products obtained with a 3×3 in. NaI(Tl) crystal.

TABLE I. Spectrographic analyses of the thorium samples.

Percent	2*a* $Th(NO_3)_4$	3*ab* Thorite	4*a*[a] $Th(NO_3)_4$	5*a* $Th(NO_3)_4$
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	···	···	···	···

[a] Radium analysis by the radon method for sample 4*a* is $(1.65\pm0.05)\times10^{-10}$ gram/gram of thorium nitrate. J. R. Dooley, U. S. Geological Survey, analyst.

[5] Elliott, Graham, Walker, and Wolfson, Phys. Rev. **93**, 356 (1954).

TABLE II. Thorium decay constants as measured by beveled and unbeveled crystals.

Sample No.	Thorium content of capsule (g)	Beveled crystal I	Beveled crystal $\lambda(\times10^{-18})$ sec	Unbeveled crystal I	Unbeveled crystal $\lambda(\times10^{-18})$ sec
2*a*	1.098_5	1693	1.64_0	1610	1.55_9
3*ab*	1.408_3	a	a	2155	1.54_7
4*a*	1.061_5	1613	1.61_7	1545	1.54_9
5*a*	0.445_0	679.1	1.62_4	646.0	1.54_6
	Average		1.62_7		1.55_0

[a] Not determined.

face of the crystals. The peak areas in these measurements could not be obtained very accurately due to the inability to account properly for the summing contribution under the peak itself. However, the results as given in Table II 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 $\lambda=1.55\times10^{-18}$ sec^{-1}, or $T_{1/2}=1.42\times10^{10}$ years with an expected uncertainty of about 5%. This is essentially in agreement with the presently accepted value.

CONCLUSIONS

The uncertainty in the value for the half-life of thorium must be estimated from the following factors: (a) the peak efficiency of the crystal, (b) the solid angle subtended by the crystal, (c) corrections for absorption in the source and beta-ray absorber, (d) statistical uncertainty and precision in the measurement of the peak area and (e) the branching ratio of Bi^{212}. As was discussed above, the scatter of points about the curve for peak efficiency *vs* energy is less than 3%. In view of the nearness of a point at 2.76-Mev (from Na^{24}), it is felt the peak efficiency for the 2.62-Mev gamma ray should be known to this order of uncertainty. The intensities of the gamma rays as obtained with the bevelled crystal seemed to be systematically higher than the values obtained with the cylindrical crystal, as mentioned above. In several previous cases in which the unbeveled crystal was used,[6] the intensity of gamma rays which were obtained were in excellent agreement with beta-ray intensity measurements. We have had no similar experience with the bevelled crystals. Thus, in view of our considerably greater experience with the cylindrical crystal, we have calculated the value of the half-life for thorium from its data.

The solid angle uncertainty may be estimated from the following considerations: an error of 1 mm in the measurement of the radius of the crystal yields an error in the solid angle subtended of $(\Delta r/r)^2\sim5\%$. The radius of the crystal which was used for the above measurements has been carefully measured, but it may be out of round by an amount which is difficult to estimate. The vertical positioning is also fairly critical but because of the relatively large spacing of source to crystal, the error in positioning is minimized. In this case, the source position is known well within a half-millimeter. It is felt that the solid angle uncertainty contributes an error of less than 2%.

The factors (c), (d), and (e) above may be evaluated with somewhat greater certainty. The absorption corrections, as discussed above, were all less than 5% of the intensity of the gamma ray and uncertainties in their value cannot be greater than 15%. A total of 10 000 counts or more were obtained for each of the peaks and the statistical uncertainty from this cause is of the order of a percent.

A measure of the precision of the measurement of the area of the peaks, the positioning of the source, etc., can be determined from the consistency of the results. These agree with each other within two percent and the average value of the four measurements was used for the half-life determination.

The uncertainty in the branching ratio, $\alpha/(\alpha+\beta)$, in Bi^{212} is due to the statistical uncertainty in the total number of counts under each peak. The over-all uncertainty in this measurement was certainly less than 2%.

From the values of I_γ indicated in Table II, and the new value for the branching ratio of Bi^{212}, we obtain $\lambda=1.55\times10^{-10}$ sec^{-1} or $T_{\frac{1}{2}}=1.42\times10^{10}$ years with an expected over-all uncertainty of 5%.

ACKNOWLEDGMENTS

The authors wish to acknowledge the assistance of their colleagues in the U. S. Geological Survey: Frank Cuttitta, who made the thorium analyses, J. N. Rosholt, Jr., for the radiochemical analyses, 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.

[6] W. S. Lyon and N. H. Lazar, Phys. Rev. **101**, 1524 (1956); O'Kelley, Lazar, and Eichler, Phys. Rev. **101**, 1059 (1956); N. H. Lazar and E. D. Klema, Phys. Rev. **98**, 710 (1955).