OAK RIDGE NATIONAL LABORATORY
Operated By
CARBIDE AND CARBON CHEMICALS COMPANY

UCC

POST OFFICE BOX P
OAK RIDGE, TENNESSEE

0

ORNL
CENTRAL FILES NUMBER

CF-55-6-43

DATE: June 8, 1955
SUBJECT: Calculations of Gamma Counting
Rate of Thorium
TO: Distribution Shown
FROM: S. A. Reynolds

Photostat Price \$ 3.30
Microfilm Price \$ 2.40
Available from the
Office of Technical Services
Department of Commerce
Washington 25, D. C.

This report was prepared as a scientific account of Government-sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights. The Commission assumes no liability with respect to the use of, or from damages resulting from the use of, any information, apparatus, method, or process disclosed in this report.

421-1

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

CALCULATIONS OF GAMMA COUNTING RATE OF THORIUM

S. A. Reynolds

Analytical Chemistry Division
Oak Ridge National Laboratory

A. Introduction

In the Thorex process, it is of some interest to know the magnitude of the natural gamma activity associated with thorium. This gamma activity is a complicated function of time after separation of thorium from its descendants, increasing from essentially zero (immediately after separation) to a "pseudo-equilibrium" level at about thirty days,¹ diminishing, then finally increasing to equilibrium at about sixty years. An excellent description of the thorium series is given by Fresco, et al.²

Most gamma-activity measurements for Thorex are made by use of well-type scintillation counters, equipped with cup-type lead absorbers about 5 g/cm² in thickness.^{3,4} All calculations refer to this method of measurement.

B. Available Data

The significant gamma emitters in the thorium series are Ac-228 (MsTh₂), Ra-224 (ThX), Pb-212 (ThB), Bi-212 (ThC), and Tl-208 (ThC'). Gamma radiation of energy below 0.2 Mev was ignored, since, as indicated below, it is counted with extremely low efficiency. Data on the decay scheme of Ac-228 were given by Kyles, et al.⁵ Information on the decay schemes of the other nuclides was taken from several sources.^{2,6,7,8} Most of the decay scheme data are in approximate agreement with those used by Arnold.⁹

The efficiency of a scintillation counter for gamma radiation is a function of energy. In making the calculations included in this paper,

CLASSIFICATION CANCELLED
DATE 12-20-55
For The Atomic Energy Commission
71-10000-10
Chief, Declassification Branch BG

it was assumed that the energy-efficiency curve for a counter used by E. I. Wyatt (Analytical Chemistry Division) is typical of all counters of the same type. The curve, Figure 1, was plotted from data given by Wyatt,¹⁰ and it was noted that the form of the curve was the same as that observed by Lyon¹¹ for another instrument.

C. Calculations

The specific activity of Th-232 is approximately 240 dpm/mg. In radioactive equilibrium the disintegration rates of all the daughters have the same value, with the exception of Po-212 and Tl-208, which have disintegration rates which are respectively $\sim 2/3$ and $\sim 1/3$ of the above quantity. From the decay schemes of each of the gamma-emitting daughters, the numbers of gamma photons of various energies were calculated for the equilibrium system, and these are given in the third column ("Intensity") of Table I. In the fourth column the efficiency of the scintillation counter for each gamma is given. The fifth column shows the product of intensity and efficiency, which is equivalent to the number of counts of a particular gamma per disintegration of Th-232. Finally, the sixth column (fifth column X 240) shows the number of counts per minute of each gamma per milligram of Th-232. It will be noted that the counting rate of 1 mg. of Th, in equilibrium with its daughters, is predicted to be 101 cpm, and that 54 cpm is associated with Ac-228, and 47 cpm with short-lived daughters of Th-228 (RdTh).

The equilibrium counting rates are not immediately useful, since no thorium encountered in Thorex is in equilibrium with its daughters. However, the numerical values in Table I may be used to calculate the activity of Th as a function of time after separation. Calculations are

Figure 1 - Efficiency of Scintillation Counter

421-4

Table I

Gamma Radiation from Thorium Series in Equilibrium

Source	E_γ	Intensity ^a	Efficiency	$I \times E^b$	Activity ^c
Ac-228	$\sim 1.62^d$	~ 0.43	~ 0.25	~ 0.10	24
	$\sim 0.94^d$	~ 0.45	~ 0.24	~ 0.11	26
	$\sim 0.43^d$	~ 0.07	0.17	~ 0.01	2
	0.33	~ 0.09	~ 0.12	~ 0.01	2
Ra-224	0.25	~ 0.04	~ 0.03	--	--
Pb-212	0.30	0.03	~ 0.07	--	--
	0.24	0.30	~ 0.02	~ 0.006	1
Bi-212	1.80	~ 0.04	0.25	~ 0.01	2
	1.61	~ 0.04	0.25	~ 0.01	2
	1.35	~ 0.03	0.25	~ 0.01	2
	1.03	~ 0.04	0.24	~ 0.01	2
	0.86	0.040	0.23	0.009	2
	0.73	0.040	0.22	0.009	2
Tl-208	2.62	0.33	~ 0.25	0.08	19
	0.86	0.04	0.23	0.009	2
	0.58	0.21	0.20	0.042	10
	0.51	0.07	0.19	0.013	3
	0.28	0.021	~ 0.055	--	--

54

47

^a Photons per disintegration of Th-232.

^b Counts per disintegration of Th-232.

^c cpm/mg of Th-232.

^d Average of two gammas.

421-5

greatly simplified by noting that the growth of Ac-228 (MsTh₂) is governed by the growth of its parent, Ra-228 (MsTh₁), after about two days of growth. The latter has a half-life of 6.7 years. Further, the growth of the daughters of Th-228 is governed by its growth after about thirty days.

The growth of Ac-228 is given by the equation¹²

$$A_1 = A_1^0 (1 - e^{-\lambda_1 t}),$$

where A_1 = Ac activity in Th at time t after purification of Th;

A_1^0 = equilibrium Ac activity;

λ_1 = decay constant of Ra-228 = $3.28 \times 10^{-9} \text{ sec}^{-1}$.

A_1^0 is 54 cpm, from Table I. Therefore

$$A_1 = 54 (1 - e^{-\lambda_1 t}) \quad (1)$$

The growth of the daughters of Th-228 is given, after ~30 days, by the equation

$$A_2 = A_2^0 e^{-\lambda_2 t} + A_2^0 [\lambda_2 (1 - e^{-\lambda_1 t}) - \lambda_1 (1 - e^{-\lambda_2 t})] / (\lambda_2 - \lambda_1),$$

where A_2 = Th-228 daughter activity in Th at time t after purification of Th;

A_2^0 = equilibrium Th-228 daughter activity;

λ_2 = decay constant of Th-228 = $1.16 \times 10^{-8} \text{ sec}^{-1}$.

The first term indicates the decay of the Th-228 daughters initially present, and the second term shows the build-up from Th-232 through MsTh₁ and MsTh₂.

This equation was derived from one given by Rutherford, et al.¹³ On substituting the value for A_2^0 given in Table I and other numerical quantities, the equation becomes

$$A_2 = 47 e^{-\lambda_2 t} + 5.7 [11.6 (1 - e^{-\lambda_1 t}) - 3.28 (1 - e^{-\lambda_2 t})] \quad (2)$$

A description of the growth of the Th-228 daughters in a short period of time has been given previously.¹

421-6

In Table II, equations (1) and (2) have been used to calculate the gamma activities of Th at times varying from 0.1 to 10 years.

If a separation of Th from the equilibrium system is made, and after some time, a second purification is performed, the value of A_2 corresponding to the time between purifications indicates the "pseudo-equilibrium" activity. Thus, suppose Th is separated from an ore, allowed to stand undisturbed for four years, then separated again. The gamma activity of the Th would be only ~19 cpm/mg about a month after the second purification, and would increase gradually, as shown in Table III. If a third purification were made, say at six years after the first, the pseudo-equilibrium activity would be only ~12 cpm/mg. The data of Tables II and III are shown in graphical form in Figure 2.

D. Experimental

No experiments have been performed on thorium samples of known history, but a few results have been obtained which show that the predictions made in part C are not grossly in error. A sample of Th of unknown history was purified and the growth of activity was followed to pseudo-equilibrium. The gamma activity was 14 ± 1 cpm/mg.¹ The data of Table III indicate that the original separation may have been made five years earlier, and this is possible. A sample of Th metal which had been in the author's possession almost four years showed an activity of 35.5 cpm/mg, in reasonable agreement with the data of Table II. A sample which probably had not been processed for eight years showed gamma activity of ~50 cpm/mg,¹⁴ again in agreement with the predicted value.

E. Conclusions

It is apparent that the gamma activity associated with natural thorium cannot be predicted unless the history of the material is known

421-7

Table II

Gamma Activity of Thorium as a Function
of Time after Purification

Time (years)	A ₁ (cpm/mg)	A ₂ (cpm/mg)	Total A (cpm/mg)
0.1	0.5	45.4	46
0.5	2.8	39.5	42
1	5.3	33.5	39
2	10.2	25.4	36
3	14.4	20.9	35
4	18.3	19.0	37
5	21.8	18.7	40
6	25.0	19.3	44
7	27.8	20.5	48
8	30.3	22.0	52
9	32.7	23.7	56
10	34.8	25.6	60
~60 ^a	54	47	101

^a equilibrium

421-8

Table III

Thorium Gamma Activity after Double Purification

t^a (years)	t^{lb} (years)	A_1 (cpm/mg)	A_2 (cpm/mg)	Total A (cpm/mg)
0.1-4	---		As in Table II	
4.0 ^c	0.0	0	0	~ 0
4.1	0.1	0.5	18.3	19
4.5	0.5	2.8	16.2	19
5	1	5.3	14.1	19
6	2	10.2	11.9	22
7	3	14.4	11.5	26
8	4	18.3	12.4	31
9	5	21.8	14.2	36
10	6	25.0	16.1	41

^a Time from original purification

^b Time from second purification

^c Time of second purification

421-9

9

Figure 2 - Gamma Activity of Thorium

-6-

421-10

from the time it was separated from the ore. If it is assumed that the information presented by the author in an earlier memo¹ was based on thorium which is representative of that being used in the Thorex process, the activity data therein may be useful. The uncertainty of the quantitative predictions made in the present memo is determined largely by the validity of the decay schemes assumed. It is therefore difficult to estimate, but it probably does not exceed 20%. If someone could furnish thorium samples of accurately known history, experimental data could be obtained which would be subject to much less doubt. Obviously, if the thorium-series daughters of U-232 are present in a Th product,⁹ its gamma activity could be calculated by similar methods.

F. References

1. S. A. Reynolds, "Preliminary Study of 'Cotton' and Thorium 'Backgrounds' in the Thorex Process," ORNL-CF-54-11-117 (November 22, 1954).
2. J. Fresco, E. Jetter, and J. Harley, Nucleonics 10, No. 8, 60 (1952).
3. S. A. Reynolds and W. S. Lyon, Anal. Chem. Quar. Prog. Rep. Sept. 26, 1952, ORNL-1423, p. 14.
4. ORNL Master Analytical Manual, Method No. 2 00365.
5. J. Kyles, C. G. Campbell, and W. J. Henderson, Proc. Phys. Soc. (London) 66A, 519 (1953).
6. J. M. Hollander, I. Perlman, and G. T. Seaborg, Revs. Mod. Phys. 25, 469 (1953).
7. D. E. Muller, H. C. Hoyt, D. J. Klein, and J. W. M. DuMond, Phys. Rev. 88, 787 (1952).
8. D. G. E. Martin and H. O. W. Richardson, Proc. Phys. Soc. (London) 63A, 223 (1950).

421-11

9. E. D. Arnold, "Formation of U^{232} and the Effects of Its Decay Chain Activity . . .," ORNL-1869 (April 4, 1955).
10. ORNL Master Analytical Manual, Method No. 9 0733002.
11. D. A. Brown and W. S. Lyon, Anal. Chem. Quar. Prog. Rep. March 26, 1952, ORNL-1276, p. 26.
12. G. Friedlander and J. W. Kennedy, Introduction to Radiochemistry, p. 112, Wiley, New York, 1949.
13. E. Rutherford, J. Chadwick, and C. D. Ellis, Radiations from Radioactive Substances, p. 17, Cambridge, 1930.
14. R. E. Leuze, personal communication, April, 1955.

421-12

0-11

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK