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Half-Life of Samarium-147[†]

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A liquid scintillation counting technique has been applied to the determination of the half-life of Sm^{147} by specific alpha activity measurement. A value of $(1.05\pm0.02)\times10^{11}$ years is obtained.

INTRODUCTION

The presence of natural alpha radioactivity in samarium was discovered independently by Hevesy and Pahl[1] and Libby and Latimer.[2] This activity was later shown[3,4] to be due to the isotope of mass 147. A number of investigations of the half-life of this radioactivity by the method of specific activity have been made and are summarized in Table I. The results of these determinations, using a variety of counting techniques, range over a factor of two.

The low α energy (2.18 Mev)[5] and low specific activity make the determination of the absolute disintegration rate difficult owing to uncertainties in sample self-absorption, the possibility of radioactive impurities, and poor counting statistics. The photographic emulsion technique offers some advantages for the measurement of low-intensity alpha radioactivity; however, problems of uniformity and precise area of impregnation are present.[6]

Liquid scintillation counting offers a convenient solution to many of the problems associated with the absolute counting of low specific activity and/or low energy radioactivities.[7–12] Large samples may be dissolved in the detecting medium to give 4π geometry with no sample self-absorption. Experiments comparing the results of absolute disintegration rate determinations by conventional alpha counting techniques with liquid scintillation counting have been carried out in this laboratory, and the results indicate a high degree of reliability for the latter technique.

The present investigation of the half-life of Sm^{147} is part of a general program of study of naturally occurring radioactivities with liquid scintillation counters. During the course of the present work, a value for the half-life of Sm^{147} based on liquid scintillation counting was published by Beard and Kelly (reference k, Table I). This measurement is in disagreement with our result, and the discrepancy is discussed below (see Discussion).

APPARATUS

A Tri-Carb (model 314) liquid scintillation spectrometer[13] was used for the determination of alpha disintegration rates. The samples were counted in cylindrical, screw-cap, glass vials (25×56 mm) with aluminum liners in the caps. The vials were placed in a well in a cylindrical Lucite light pipe containing silicone oil for optical coupling.[10] Exposed surfaces of the vials and light pipe were painted with TiO_2-Lucite reflecting paint. The light pipe was coupled with Dow-Corning grease to two DuMont 6292 (two inch) multiplier phototubes mounted horizontally in a refrigerated chest operated at $-10°C$. The pulses from the phototubes are fed through an amplifier, coincidence circuit, pulse-height analyzer, and scaler. In this apparatus[11]

Table I. Summary of previous measurements of the half-life of Sm^{147}.

Investigator	Counting method	Specific activity (α/sec g nat. Sm)	Half-life[a] (10^{11} yr)
G. Hevesy, M. Pahl, and R. Hosemann[b]	Geiger counter	75	1.8
M. Herzfinkiel and A. Wroncberg[c]	Ionization chamber	67	2.0
M. Mader[d]	Ionization chamber	89	1.5
W. F. Libby[e]	Screen-wall counter	...	0.95 ± 0.08
R. Hosemann[f]	Geiger counter	89 ± 5	$1.5\ \pm0.1$
C. M. G. Lattes, E. G. Samuel, and P. Cuer[g]	Nuclear emulsion	...	$1.4\ \pm0.1$
E. Picciotto[h]	Nuclear emulsion	133 ± 6	1.01 ± 0.05
G. Beard and M. L. Wiedenbeck[i]	4π Geiger counter	719 ± 36[m]	1.25 ± 0.06
G. E. Leslie[j]	Nuclear emulsion	115 ± 3	1.15 ± 0.03
G. Beard and W. H. Kelley[k]	Liquid scintillation counter	702 ± 20[m]	1.28 ± 0.04
R. D. Macfarlane[l]	Ionization chamber	113 ± 5	1.18 ± 0.05

[a] Based on 15.0% isotopic abundance for mass 147.
[b] Z. Physik **83**, 43 (1933).
[c] Compt. rend. **199**, 133 (1934).
[d] Z. Physik **88**, 601 (1934).
[e] Phys. Rev. **46**, 196 (1934).
[f] Z. Physik **99**, 405 (1936).
[g] Anais acad. brasil. cienc. **19**, 1 (1947); Nature **158**, 197 (1946).
[h] Compt. rend. **229**, 117 (1949).
[i] Phys. Rev. **95**, 1245 (1954).
[j] Thesis, North Carolina State College, AD-37749, 1954 (unpublished).
[k] Nuclear Physics **8**, 207 (1958).
[l] Thesis, Carnegie Institute of Technology, Atomic Energy Commission Report NYO-7687, 1959 (unpublished).
[m] Specific activity per gram Sm^{147}.

[†] Based on work performed under the auspices of the U. S. Atomic Energy Commission.

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[1] G. Hevesy and M. Pahl, Nature **130**, 846 (1932).

[2] W. F. Libby and W. M. Latimer, J. Am. Chem. Soc. **55**, 433 (1933).

[3] B. Weaver, Phys. Rev. **80**, 301 (1950).

[4] J. O. Rasmussen *et al.*, Phys. Rev. **80**, 475 (1950).

[5] W. P. Jesse and J. Sadauskis, Phys. Rev. **78**, 1 (1950).

[6] See reference h, Table I.

[7] J. Steyn, Proc. Phys. Soc. (London) **69A**, 865 (1956).

[8] J. K. Basson and J. Steyn, Proc. Phys. Soc. (London) **67A**, 297 (1954).

[9] J. K. Basson, Anal. Chem. **28**, 1472 (1956).

[10] D. L. Horrocks and M. H. Studier, Anal. Chem. **30**, 1747 (1958).

[11] K. F. Flynn and L. E. Glendenin, Phys. Rev. **116**, 744 (1959).

[12] H. H. Seliger, Int. J. Appl. Rad. and Isotopes, **8**, 29 (1960).

[13] Packard Instrument Company, P. O. Box 428, LaGrange, Illinois.

the low temperature and coincidence requirement both serve to reduce the phototube noise background.

When differential pulse height spectra were desired, a Lucite light pipe with one face covered with the reflecting paint was used, and the output of one multiplier phototube was fed through a separate amplifier to a 256-channel analyzer.

EXPERIMENTAL

Source Purity

A source of samarium oxide of very high purity (99.9%) was used for this work.[14] A complete spectrochemical analysis indicated the only impurities to be about 0.1% Ca and 0.01% Si. These were probably reduced further by the chemical preparation of samarium octoate described below.

The possible presence of radioactive impurities such as uranium or thorium was investigated by searching for gamma-ray activity in the Sm_2O_3 using a 3×3 in. NaI(Tl) crystal spectrometer. The gamma radiations from known weights of U_3O_8 and $Th(NO_3)_4$ were analyzed, and samples of Sm_2O_3 and background were determined under identical conditions. The pulse-height distribution for the Sm_2O_3 was essentially identical with the background. These data indicated an upper limit to the presence of uranium and thorium corresponding to an alpha contamination of less than 0.8% of the Sm^{147} activity.

A low-background proportional counter with a 1.0-mg/cm^2 Mylar window was used to detect the possible presence of beta activity. A 20-mg sample of Sm_2O_3 spread over an area of 7 cm^2 was counted through a 1.5-mg/cm^2 aluminum absorber (to stop the Sm^{147} alphas) with a geometric efficiency of 0.16. A net counting rate of 0.16±0.05 count/min was observed, corresponding to an upper limit of about 1% contamination of the Sm^{147} with beta radiation of maximum energy greater than 300 kev.

An alpha pulse-height analysis was carried out on a thin sample of the samarium with a Frisch-grid ionization chamber spectrometer. Although it was not possible to prepare a thin, uniform source of the high quality normally used in such analyses, it was possible to set an upper limit of 0.6% to the presence of alpha particles of higher energy than that of Sm^{147} (i.e., >2.2 Mev).

A more sensitive test for the presence of contaminating beta and alpha activity is provided in the liquid scintillator measurements described below.

SAMPLE PREPARATION

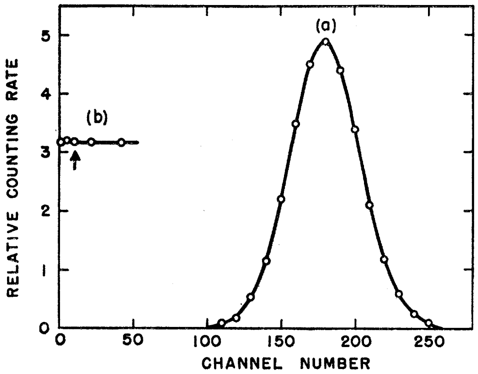

FIG. 1. Pulse-height analysis of Pu^{239}. (a) Differential curve. (b) Integral curve.

Ronzio[15] has shown that the 2-ethyl-hexanoic acid (octoic acid) salts of many metal ions are soluble in organic scintillator solutions and do not cause appreciable quenching. The usefulness of this reagent in liquid scintillation counting for alkali metals, alkaline earths, and rare earths, as well as several other metallic elements, has been verified in this laboratory, and excellent results have been obtained. Samarium octoate was prepared in the following manner. The Sm_2O_3 was dissolved in HNO_3. The hydroxide was precipitated with ammonia, washed free of NO_3^- ion with H_2O, and dissolved in glacial acetic acid. A slight excess of octoic acid was added, and the acetic acid was removed by evaporation under vacuum. The thick, oily samarium octoate that remained was dissolved in scintillator solution [4 g *p*-terphenyl and 0.1 g 1, 4-di-[2-(5-phenyl-oxazolyl)]-benzene (POPOP) per liter of toluene] and made up to 100 ml in volume.

ABSOLUTE ALPHA COUNTING

The liquid scintillation counter was established as an absolute alpha counter (100% efficiency) by comparison with a carefully standardized intermediate geometry alpha counter (IGAC).[16] Equal aliquots of a pure Pu^{239} solution were prepared (with the same 50 λ

TABLE II. Alpha counting efficiency of liquid scintillation counter.

Pu^{239} sample	IGAC[a] α dis/min	Liquid scintillation counter[b] α counts/min
1	32 142±58	32 293±56
2	32 164±58	31 833±56
3	31 956±58	32 187±25
Average	32 090±51	32 104±45

[a] Geometric efficiency =0.09633 ±0.00001; Background =13.3 counts/min.
[b] Background =56.8 counts/min.

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[14] Lindsay Chemical Division, American Potash and Chemical Company, West Chicago, Illinois.

[15] A. R. Ronzio, Int. J. Appl. Rad. and Isotopes 4, 196 (1959).

[16] A. H. Jaffey, *The Actinide Elements*, edited by G. T. Seaborg and J. J. Katz, (McGraw-Hill Book Company, New York, 1954), National Nuclear Energy Series, Plutonium Project Record, Vol. 14A, Div. IV, Chap. 16; A. H. Jaffey, C. T. Hibdon, and R. Sjoblom, J. Nuclear Energy, Part A **11**, 21 (1959); and later unpublished modifications.

pipet) for counting in the IGAC by evaporation on Pt disks and for counting in the Tri-Carb by dissolution in the liquid scintillator solution with di-*n*-butyl phosphate (DBP) as a complexing agent.

A differential pulse-height analysis of the Pu^{239} source in the liquid scintillator indicating a well-defined, symmetric alpha peak is shown in Fig. 1(a). Integral counts were taken on the Tri-Carb (coincidence mode) at several pulse-height discriminator settings below the position of the alpha peak to ensure the counting of all pulses due to alpha particles. Backgrounds were taken under identical conditions with blank DBP-scintillator solutions. The constancy of the net integral counting rate over this region of pulse height is illustrated in Fig. 1 (b). The counting data are summarized in Table II. Counting rates in the liquid scintillator were determined at the discriminator setting indicated by the arrow in Fig. 1 (b). All samples were counted to a minimum of 300 000 total counts to obtain a standard statistical error of $<0.2\%$. The ratio of the disintegration rate obtained with the liquid scintillation counter to that with the IGAC is 1.0004 ± 0.002. From these data it is evident that the efficiency of the liquid scintillation counter for alpha particles is 100%. A similar result has been obtained by Seliger[12] for Po^{210} alphas by comparison with a 4π gas counter.

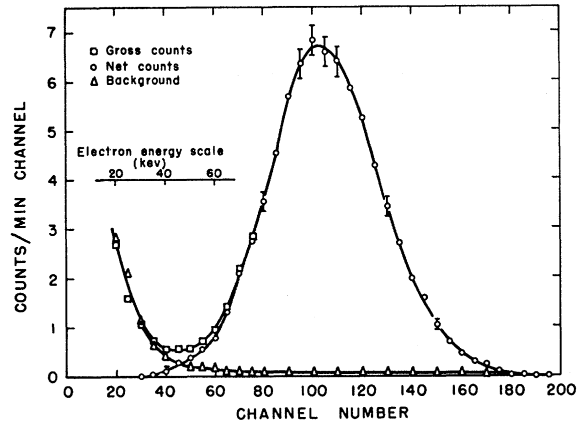

FIG. 2. Pulse-height analysis of Sm^{147}.

HALF-LIFE DETERMINATION

Six 15-ml aliquots of the samarium octoate-scintillator solution were counted in the same manner as described previously for Pu^{239}. Backgrounds were taken with blank octoic acid-scintillator solutions. Ethanol was added until the pulse-height output as measured by an external Cs^{137} source (Ba^{137m} conversion electron peak) was the same for the blank as for the samarium-loaded scintillator solution, i.e., until the degree of quenching in the two solutions was identical.

The differential pulse-height spectrum of Sm^{147} is shown in Fig. 2. A well-defined symmetric peak is observed with no evidence of contaminating activity. By comparison with the alpha spectra of Pu^{239} and Th^{230} the relative pulse height and resolution of the Sm^{147} alpha peak is consistent with the emission of monoenergetic alpha particles of about 2.2 Mev. An approximate electron energy scale is given in the figure for the region of low pulse heights. The absence of low-energy beta contamination in this region confirms the previously described observations made with an end-window proportional counter. In addition, no activity in excess of background was observed in the pulse-height region above the Sm^{147} alpha peak within a statistical uncertainty of 0.1% of the Sm^{147} activity.

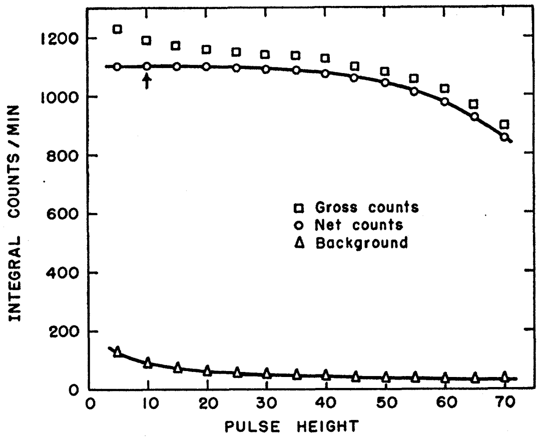

FIG. 3. Integral counting rate curve for Sm^{147}.

An integral counting rate versus pulse-height curve is given in Fig. 3. Integral counting data for the Sm^{147} half-life determination were recorded with the discriminator set below the alpha peak at the position indicated by the arrow.

Data for the determination of the Sm^{147} half-life are presented in Table III. A total of at least 10^6 counts was recorded for each aliquot to limit the statistical error to less than 0.1%. Background determinations were made at intervals throughout the time required to count the six aliquots. The background was observed to be constant within statistical error, and an average value of 1.427 ± 0.002 count/sec was used for the calculation of the net counting rates (column 2).

TABLE III. Data for Sm^{147} half-life determination.

Aliquot	Activity (dis/sec)	Weight Sm_2O_3 (g)	Specific activity (dis/sec g Sm)	Half-life[a] (10^{11} yr)
1	18.18	0.1686	125.0	1.054
2	18.17	0.1683	125.2	1.052
3	18.17	0.1685	125.1	1.053
4	18.28	0.1685	125.8	1.047
5	18.25	(0.1685)[b]	125.6	1.049
6	18.22	(0.1685)[b]	125.4	1.050
Average	18.21 ± 0.04	0.1685 ± 0.0001	125.3 ± 0.3	1.051 ± 0.01

[a] Based on $(14.97 \pm 0.10)\%$ isotopic abundance for mass 147.
[b] Aliquot not assayed; average value used.

After counting, four of the six aliquots of samarium octoate-scintillator solution were carefully wet-ashed with a mixture of conc. H_2SO_4 and $HClO_4$ to destroy all organic matter. The solutions were reduced to a small volume (about 3 ml), diluted to $1M$ H_2SO_4, and heated to boiling. Samarium oxalate was quantitatively precipitated by the addition of saturated $H_2C_2O_4$, allowed to stand overnight, filtered, washed with 1% $H_2C_2O_4$, ignited in Pt crucibles at 800°C, and weighed as Sm_2O_3 (column 3).

The half-life of Sm^{147} (column 5) was calculated from the relation, $T_{\frac{1}{2}}=0.693N/A$, where N is the number of atoms of Sm^{147} and A is the disintegration rate. The isotopic abundance for mass number 147 in natural samarium was taken as $14.97\pm0.10\%$.[17] It should be noted that the uncertainty in this value actually represents the largest source of error in the present investigation. The observed specific activity of 125.3 ± 0.3 α/sec g natural samarium (column 4) corresponds to a half-life of $(1.05_1\pm0.01)\times10^{11}$ years for Sm^{147}.

DISCUSSION

Most of the earlier measurements of the half-life of Sm^{147} suffer from uncertainties in purity and assay of the source and from possible errors in the correction for self-absorption of alpha particles in the source material. In the present work considerable care was taken to minimize errors associated with source purity and assay. The most sensitive test for possible radioactive contamination is the shape of the differential pulse-height spectrum (Fig. 2). The curve is typical of a pure alpha emitter, and after background subtraction, shows no residual activity above or below the peak. Any low-energy beta radioactivity would exhibit a rapidly rising spectrum with decreasing pulse height in the region of channel numbers 20–70. The maximum statistical error of the observed data in this region may be used to give an upper limit of 1% to the presence of any such activity. A summary of the assessed errors in the present measurement is given in Table IV. Application of these errors to the observed results gives a value for the half-life of Sm^{147} of $(1.05_1\pm0.02)\times10^{11}$ years.

TABLE IV. Evaluation of errors.

Type of error	Value
Chemical purity of source	$\leq0.1\%$
Radioactive purity of source	$<1\%$
Chemical assay	$\pm0.06\%$
Counting statistics	$\pm0.1\%$
Isotopic abundance of mass 147	$\pm0.7\%$

Beard and Kelly (reference k, Table I) also employed a liquid scintillation counting technique, but obtained a 20% larger value for the half-life. These authors kindly sent us their sample of a commercially-loaded samarium scintillator solution for examination. After correction for the presence of some alpha contamination of higher energy than the Sm^{147}, a counting rate in good agreement with that observed by Beard and Kelly was obtained. However, a chemical assay for samarium showed that the reported samarium content was considerably in error. The corrected data on their sample correspond to a half-life of $(1.06\pm0.04)\times10^{11}$ years, in good agreement with our value.

ACKNOWLEDGMENTS

The kind cooperation of Mr. D. Foster of the Lindsay Chemical Division, American Potash and Chemical Company in providing a selected source of Sm_2O_3, and of Professor G. B. Beard of Wayne State University in giving us the opportunity to resolve the discrepancy between our measurements is gratefully acknowledged. We also wish to express our appreciation to Mr. J. P. Faris who performed the spectrographic analyses, Mr. D. Henderson who carried out the ionization chamber spectrometer analyses, Dr. A. H. Jaffey who measured the absolute disintegration rates of the Pu^{239} samples in the IGAC, and Mr. K. F. Flynn for his general helpfulness in many phases of the work.

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[17] *Nuclear Data Tables*, edited by K. Way, National Academy of Science, National Research Council (U. S. Government Printing Office, Washington, D. C., 1959).