

5 keV and with an intensity greater than 50 percent of the 1.06-MeV line.

The *F* line of thorium *B* was used to recheck the earlier precision calibration⁷ of the spectrometer. The instrument is known to be linear to a very high degree.⁷

At the higher resolution the *K* photoconversion line of the 0.57-MeV gamma ray was ~ 0.6 count/sec above background. We conclude that two nearly equal energy gamma rays do not exist in the region of the 0.57-MeV gamma ray either.

Upon adding the 109.79-keV binding energy of thorium⁸ to the measured energy of the *K* photoconversion electrons, we obtain 1063.43 ± 0.50 keV and 568.85 ± 0.30 keV, respectively, for the energies of the two gamma rays. The former is to be compared with the measurement of Alburger,¹ *viz.*, 1063.9 ± 0.3 keV.

⁷ D. I. Meyer and F. H. Schmidt, Phys. Rev. **94**, 927 (1954).

⁸ Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).

Lazar and Klema³ have shown that the 1.06–0.57 MeV cascade have relative gamma intensities of 0.77 ± 0.06 and 1.00, respectively. Bi²⁰⁷ is thus particularly useful as a test source in gamma-ray coincidence spectroscopy. The 975.9-keV *K* conversion electrons¹ from the 1.06-MeV transition, together with the coincident 0.57 gamma ray, have proved to be of particular use in this laboratory for energy calibration of beta-gamma coincidence scintillation spectrometers.

ACKNOWLEDGMENTS

We wish to thank T. J. Morgan and the cyclotron crew for the long deuteron bombardment, J. R. Penning for help with the chemical separation, and H. R. Maltrud for assistance with the tedious hours of operation of the beta-ray spectrometer.

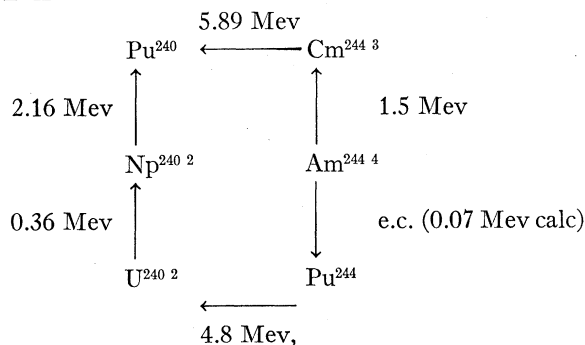
Electron Capture Decay of Am²⁴⁴ and the Spontaneous Fission Half-Life of Pu²⁴⁴†

P. R. FIELDS, J. E. GINDLER, A. L. HARKNESS, M. H. STUDIER, J. R. HUIZENGA, AND A. M. FRIEDMAN
Argonne National Laboratory, Lemont, Illinois

(Received June 30, 1955)

The branching ratio (electron captures/beta particles) of Am²⁴⁴ was determined as $0.039 \pm 0.003\%$. An enriched Pu²⁴⁴ sample was isolated from neutron irradiated Am²⁴³, and the spontaneous fission half-life of Pu²⁴⁴ was found to be $(2.5 \pm 0.8) \times 10^{10}$ years.

A CONSIDERATION of the closed energy cycle¹



using available data and an estimated value of 4.8 MeV for the alpha disintegration energy of Pu²⁴⁴, indicates

† Work performed under the auspices of the U. S. Atomic Energy Commission.

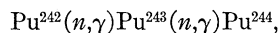
¹ Studier, Fields, Sellers, Friedman, Stevens, Mech, Diamond, Sedlet, and Huizenga, Phys. Rev. **93**, 1433 (1954).

² Knight, Bunker, Warren, and Starner, Phys. Rev. **91**, 889 (1953).

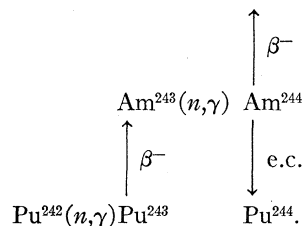
³ Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 611 (1953).

⁴ Ghiorso, Thompson, Choppin, and Harvey, Phys. Rev. **94**, 1081 (1954).

that Am²⁴⁴ may be slightly unstable toward electron capture decay. The fact that Pu²⁴⁴ is apparently beta stable^{1,5} suggests that Am²⁴⁴ is analogous to Am²⁴² and is probably unstable toward electron capture decay. Evidence of electron capture decay for Am²⁴⁴ can be found in the Materials Testing Reactor (MTR) irradiations of plutonium. The Pu²⁴² to Pu²⁴⁴ ratios in several MTR irradiations of plutonium (Table I) indicated that Pu²⁴⁴ was being produced more rapidly than would be predicted from the path



and that some Pu²⁴⁴ was probably being formed by the alternate path



⁵ Inghram, Hess, Fried, and Pyle, private communication.

TABLE I. The $\text{Pu}^{244}/\text{Pu}^{242}$ ratios in MTR irradiated plutonium.

Sample No.	Integrated flux	$\text{Pu}^{244}/\text{Pu}^{242}$
1	4×10^{21}	5.28×10^{-5}
2	1.1×10^{22}	3.84×10^{-4}
3	1.4×10^{22}	5.26×10^{-4}

In order to determine the electron capture branching ratio of Am^{244} and to prepare some enriched Pu^{244} samples, Am^{243} was irradiated in the Argonne heavy water reactor and in the MTR.

Approximately one milligram of Am^{243} was irradiated for two days in the heavy water reactor and later also irradiated for three weeks in the MTR. The plutonium formed in the irradiation was isolated from the Am^{243} with a Dowex A-1 anion resin column and TTA (thenoyltrifluoroacetone) extractions. Table II shows the amount of Pu^{244} and Cm^{244} formed in the irradiations and Table III gives the isotropic composition of the plutonium fractions. An average branching ratio (electron captures/beta particles) of $0.039 \pm 0.003\%$ was obtained from the two experiments. The largest source of error in the determination is the uncertainty in the Cm^{244} half-life (18.4 ± 0.5 yr).⁶ Complete recovery of plutonium was assumed, and in

TABLE II. Results of irradiating *ca* 1 mg of Am^{243} .

Reactor	$\mu\text{g Pu}^{244}$ formed	$\mu\text{g Cm}^{244}$ formed
CP-5	3.75×10^{-6}	8.95×10^{-2}
MTR	1.42×10^{-2}	39.15

view of the fact that the recovery of plutonium after the first step in the purification could be followed by the alpha pulse analyzer, this assumption is probably fairly reliable. From the branching ratio and the half-life for beta emission (25 min)⁴ an electron capture half-life of 44.5 days was calculated. A mass spectrometric analysis of the irradiated Am^{243} showed less than 0.001% Am^{244} . If Am^{244} has a long-lived ground state similar to Am^{242} , then the branching of Am^{244} to this

TABLE III. Isotopic composition of plutonium formed from irradiation of *ca* 1 mg of Am^{243} .

Plutonium isotope	CP-5 reactor	MTR
Pu^{238}	17.8 $\pm 0.2^a$	<2.1
Pu^{239}	13.0 $\pm 0.2^a$	8.1 ± 0.2
Pu^{240}	67.5 ± 0.3	70.4 ± 0.4
Pu^{241}	0.052 ± 0.004	1.7 ± 0.1
Pu^{242}	1.63 $\pm 0.09^a$	4.2 ± 0.2
Pu^{244}	0.058 ± 0.004	13.5 ± 0.3

^a The larger percentages of Pu^{238} , Pu^{239} , and Pu^{242} were due to the fact that the plutonium was contaminated with some of the original irradiated plutonium. The amount of Pu^{244} introduced by this contamination was negligible.

⁶ Friedman, Harkness, Fields, Studier, and Huizenga, Phys. Rev. **95**, 1501 (1954).

state is less than 0.002%. The figure 0.002% was calculated by also taking into consideration the fact that on the average approximately one-half of the Am^{243} was in the MTR for about one year before it was isolated and reirradiated for the present experiment.

In the MTR irradiation sufficient enriched plutonium was formed to measure the spontaneous fission half-life of Pu^{244} . A total of 110 fissions in 50 356 minutes was observed using a sample containing 0.0064 microgram of Pu^{244} . After correcting for the counter background (0.00033 counts/minute), and the spontaneous fission rates due to the Pu^{240} ^{7,8} and Pu^{242} ⁹ in the sample, a spontaneous fission half-life of $(2.5 \pm 0.8) \times 10^{10}$ years was calculated for Pu^{244} .

Figure 1 shows a plot of the logarithm of the spontaneous fission half-lives of even-even plutonium isotopes as a function of the mass number A as proposed by

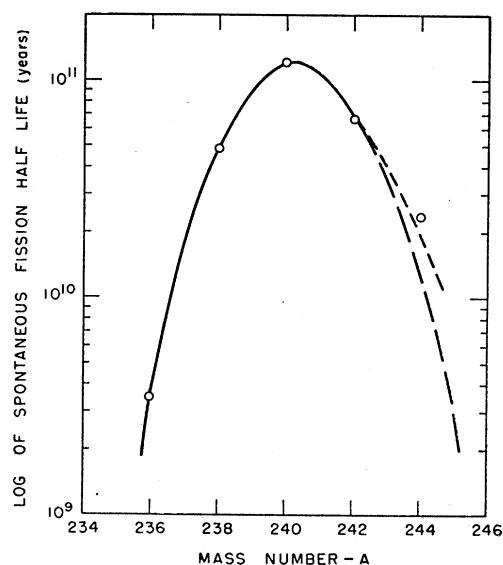


FIG. 1. A plot of the spontaneous fission half-lives of the even-even plutonium isotopes as a function of their mass number.

Huizenga.¹⁰ An extrapolation of this curve would predict a spontaneous fission half-life of $1.5-2 \times 10^{10}$ years of Pu^{244} , which is in fair agreement with the experimental value reported here.

We would like to thank Mr. D. J. Henderson of Argonne National Laboratory for the many alpha pulse analyses which made this work possible, and the staff of the Materials Testing Reactor for irradiating our sample. We would also like to acknowledge J. F. Mech of this Laboratory for his help in purifying the americium.

⁷ Chamberlain, Farwell, and Segrè, Phys. Rev. **94**, 156 (1954).

⁸ E. M. Kinderman, Atomic Energy Commission Declassified Report HW-27660, April, 1953 (unpublished). The more recent value of $1.51 \times 10^9 \pm 2\%$ fissions per gram per hour for the spontaneous fission rate of Pu^{240} as given in this reference was used in the calculations.

⁹ M. H. Studier and A. Hirsch (private communication).

¹⁰ J. R. Huizenga, Phys. Rev. **94**, 158 (1954).