Possible Existence of Cm^{247} or Its Daughters in Nature*

H. DIAMOND, A. M. FRIEDMAN, J. E. GINDLER, AND P. R. FIELDS
Argonne National Laboratory, Lemont, Illinois

(Received October 1, 1956)

The absence of any detectable Pu^{243} daughter activity in a sample of curium containing two micrograms of Cm^{247} indicates that the alpha half-life of Cm^{247} exceeds 4×10^7 years and probably exceeds 9×10^7 years. The possibilities of finding Cm^{247} in nature and of finding high U^{235}:U^{238} ratios in very old rare-earth minerals are discussed.

INTRODUCTION

INTENSIVE neutron irradiation of plutonium has produced curium enriched in the heavier curium isotopes.[1] Earlier attempts to measure the half-life of Cm^{247} were made with samples too small to yield significant results. A recent sample (isotopic composition in Table I) containing two micrograms of Cm^{247} led to a lower limit for the Cm^{247} half-life of 4×10^7 years. This implies a strong possibility of finding this nuclide or its effect in nature.

The elimination of other possible modes of decay, and arguments based on systematics indicate that the principal mode of Cm^{247} decay is alpha emission. Studies of thermonuclear test debris[2] have demonstrated that Cm^{247} is stable toward electron capture. The observed electron capture[3] by Bk^{247} is strong evidence for the beta stability of Cm^{247}. Spontaneous-fission systematics would predict the fission half-life of Cm^{247} to be greater than that of Fm^{255} (6×10^8 years).[4]

The decay of Cm^{247} leads to the following chain of nuclides:

$$Cm^{247} \xrightarrow{\alpha} Pu^{243} \xrightarrow[4.98\ hr]{\beta^-} Am^{243} \xrightarrow[8600\ yr]{\alpha} Np^{239} \xrightarrow[2.33\ days]{\beta^-}$$

$$Pu^{239} \xrightarrow[24\,400\ yr]{\alpha} U^{235} \xrightarrow[7.1\times10^8\ yr]{\alpha} \cdots.$$

The most sensitive measure of the alpha half-life would be a determination of the amount of Pu^{243} formed by the decay of Cm^{247} in a known time. Pu^{243} is readily detected by its (4.98±0.02)-hour half-life, its 560-kev beta energy, and its 84-kev gamma ray.[5,6]

EXPERIMENTAL

An accurate initial time for the growth of Pu^{243} was obtained by passing a $12M$ HCl, $0.05M$ HNO_3 solution of the curium through a Dowex A-1 anion exchange column. This removed all of the plutonium and many of the interfering fission products resulting from the spontaneous fission of Cm^{244}. Fifteen to seventeen hours after the initial purification, the plutonium produced by the curium decay was isolated by: an anion exchange column, fluoride and hydroxide precipitations, another anion exchange column, and finally a thenoyl trifluoroacetone (in benzene) extraction. The plutonium was mounted on a stainless steel disk for counting. The separation took approximately five hours. The ratio of the Pu^{240} alpha activity expected to grow into the plutonium fraction in the 15 to 17 hour decay period to the Pu^{240} activity on the final sample plate gave the chemical yield. This yield ranged from 20 to 60%.

The earliest runs showed that Pu^{243} could not be perceived above background and persisting radioactive impurities in a proportional flowing gas beta counter or in a NaI crystal used with a single-channel pulse analyzer. A much lower background and a more definitive identification of Pu^{243} activity was obtained by counting the coincidences between the beta emission and the 84-kev gamma rays that occur in 21% of the Pu^{243} disintegrations.[6] The gamma rays were detected with a NaI(Tl) crystal and their energy measured in a 20-channel pulse analyzer; the beta particles were detected with an anthracene crystal after passing through 8.3 mg/cm² of aluminum absorber. In the most sensitive run, all betas below about 120 kev were rejected. The counter's efficiency was determined by simultaneously counting aliquots of Pu^{243} (produced by neutron irradiation of Pu^{242}) in both the coincidence counter and in a 4π counter. The efficiency in the most

TABLE I. Mass spectrometric analysis of Cm.

Cm isotope	Mole %
242	0.76 ±0.03
243	0.054±0.005
244	95.6 ±0.1
245	1.04 ±0.02
246	2.1 ±0.1
247	0.024±0.003
248	0.010±0.001

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.

[1] Bentley, Diamond, Fields, Friedman, Gindler, Hess, Huizenga, Inghram, Jaffey, Magnusson, Manning, Mech, Pyle, Sjoblom, Stevens, and Studier, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. VII, Paper 809.

[2] Fields, Studier, Diamond, Mech, Inghram, Pyle, Stevens, Fried, Manning, Ghiorso, Thompson, Higgins, and Seaborg, Phys. Rev. **102**, 180 (1956).

[3] J. R. Huizenga (private communication from B. G. Harvey, September, 1955).

[4] A. Ghiorso, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. VII, Paper 718.

[5] Engelkemeir, Fields, and Huizenga, Phys. Rev. **90**, 6 (1953).

[6] Engelkemeir, Freedman, Porter, and Wagner (private communication, 1956).

sensitive run was 232 disintegrations per coincidence count in the three peak channels. The experimental error is estimated at 17%, arising mostly from the uncertainty in the mass spectrometric determination of Cm^{247}.

RESULTS AND CONCLUSIONS

No peak corresponding to the 84-kev gamma rays in coincidence with beta emission was observed. The activity in the three peak channels was analyzed to determine the maximum amount of Pu^{243} (4.98-hour component) activity that might have been present. The most probable maximum amount of Pu^{243} that might have been present was 0.05 count per minute, implying that the Cm^{247} half-life exceeds 9×10^7 years. A more conservative lower limit to the half-life is obtained by using 0.1 count/min, the upper limit to the amount of Pu^{243} activity consistent with the data, and compensating for the 17% experimental error. This gives a lower limit to the alpha emission half-life of Cm^{247} of 4×10^7 years. The validity of this limit rests on the assumption that there is no long-lived isomeric state of Pu^{243} to which Cm^{247} decays.

Terrestrial material is believed to have assumed its present nuclear form five to nine billion years ago.[7] The amount of Cm^{247} formed at that time, plus the amount of higher mass $4n+3$ series that quickly decayed into Cm^{247}, can be crudely estimated by extrapolating from the measured variation of lighter odd-mass nuclides' abundances with atomic weight.[8] The estimate of one Cm^{247} atom per thousand rare-earth atoms is probably within two orders of magnitude of being correct. An error of a factor of 100 would not seriously effect the estimate of the half-life necessary for Cm^{247} to be found in nature, but would be important to an estimate of the probability for finding an unusual U^{235}:U^{238} ratio in nature.

If we arbitrarily set the limit of detection of curium in nature by mass-spectrometric methods at one atom of Cm^{247} per 10^{15} atoms of rare earth, then Cm^{247} can be found if it has not decayed for more than 40 half-lives. Cm^{247}, then would require a half-life of 1.3×10^8 years to be found in nature if the age of terrestrial materials is five billion years, or 2.2×10^8 years if the age is nine billion years.

If the time between the formation of Cm^{247} and the separation of curium and uranium into separate mineral phases were less than seven times the Cm^{247} half-life, an identifiable increase (2%) in the U^{235}:U^{238} ratio would be found in rare-earth minerals having a rare-earth to uranium ratio of 1000 or more. Older minerals, and lower natural uranium content in the mineral would enhance the enrichment.

Cm^{247} and its daughters should have been an important source of radioactive heat[7,9] for at least the first half-billion years after the formation of the elements. The decay of one atom of Cm^{247} into U^{235} releases $(6.23\pm0.13)\times10^{-13}$ calories.[10]

Programs to look for Cm^{247} in nature, and for high U^{235}:U^{238} ratios in rare-earth ores, are currently in progress at Argonne.

ACKNOWLEDGMENTS

The authors are indebted to Donald Engelkemeir for extensive aid in counting problems, and to C. L. Stevens and A. L. Harkness for the mass spectrometric data.

[7] Reviewed by T. P. Kohman and N. Saito, *Annual Review of Nuclear Science* (Annual Reviews Inc., Stanford, California, 1954), Vol. 4, pp. 439–440, and by T. P. Kohman, Ann. N. Y. Acad. Sci. **62**, 503 (1956).

[8] H. E. Suess and H. C. Urey, Revs. Modern Phys. **28**, 53 (1956).

[9] H. C. Urey, Proc. Natl. Acad. Sci. U. S. **41**, 127 (1955).

[10] Calculated from: J. R. Huizenga, Physica **21**, 410 (1955); 65% of β^- decay energy is allowed for loss to neutrinos as in reference 9.