Possible Existence of Curium in Nature. II*

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A lower limit to the alpha half-life of Cm^{247} was previously reported as 4×10^7 yr, but the determination reported here, with a curium sample containing 30 times more Cm^{247}, yields a half-life of $(1.64\pm0.24)\times10^7$ yr. This result implies that no detectable amount of primordial Cm^{247} exists on earth and that Cm^{247} could not account for the high U^{235}:U^{238} ratios reported for certain minerals.

INTRODUCTION

IN an earlier publication[1] a lower limit to the alpha half-life of Cm^{247} was reported as 4×10^7 yr. The limit was based on the absence of any detectable Pu^{243} daughter activity in a sample of curium containing 2 μg of Cm^{247}. A consequence of this early measurement was the possibility that Cm^{247} might exist in nature. Wetherill *et al.*[2] reported that Cm^{247} had a concentration of $<10^{-14}$ g/g of rare earth. A similar measurement at this Laboratory set a limit on the concentration of Cm^{247} in nature as $<1.9\times10^{-16}$ g/g of rare earth. On the other hand, Cherdyntsev *et al.*[3] reported finding enriched U^{235} in gadolinite, magnetite, and molybdenite. Abnormally high U^{235} contents in certain select minerals containing small total amounts of uranium could arise from the decay of Cm^{247} by the path:

$$Cm^{247} \xrightarrow{\alpha} Pu^{243} \xrightarrow[4.98\text{ h}]{\beta} Am^{243} \xrightarrow[7650\text{ yr}]{\alpha} Np^{239} \xrightarrow[2.33\text{ day}]{\beta} Pu^{239}$$

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

Since the lower limit of the Cm^{247} half-life in the earlier work was dependent on a negative result, and in view of recent investigations of the presence of curium or its daughters in nature, a much better measurement of the Cm^{247} half-life was required. Hence, when a sample of Cm^{244} which had been irradiated for many years in the Materials Testing Reactor became available, the half-life determination was repeated. For comparison purpose Table I shows the isotopic composition of the curium used in this series of experiments and in the earlier work. The greatly enhanced Cm^{247} content yielded measurable amounts of Pu^{243} which was identified by its radiations and half-life. The assignment of the observed radioactivity to Pu^{243} was corroborated by separating two of the radiogenic plutonium samples in the Argonne mass separator and showing that mass-243 contained the characteristic radiations of Pu^{243}.

An alpha half-life of $(1.64\pm0.24)\times10^7$ yr was found for Cm^{247} as a result of the experiments reported in this paper.

TABLE I. Isotopic composition of curium samples.

Curium isotope	Earlier sample[a] (mole %)	Present sample (mole %)
242	0.76 ±0.03	0.0019[b]
243	0.054±0.005	···
244	95.6 ±0.1	75.9 ±0.2
245	1.04 ±0.02	0.838±0.016
246	2.1 ±0.1	21.48 ±0.2
247	0.024±0.003	0.684±0.014
248	0.010±0.001	1.059±0.02

[a] See Ref. 1.
[b] Measured by alpha-pulse analysis.

EXPERIMENTAL

9.5 mg of curium were purified and separated from plutonium by a Dowex-1 anion resin exchange column as described in the earlier paper.[1] This separation provided an accurate initial time for the period of growth of Pu^{243} from its parent Cm^{247}. This time interval varied from 22 to 70 h. Following the growth period, the plutonium was separated from the curium and fission products resulting from the spontaneous fission of Cm^{244} by an anion resin exchange column, a thenoyl trifluoroacetone (in benzene) extraction, and finally another anion exchange column. The amount of Pu^{240} in the purified plutonium was determined by a multichannel alpha-pulse analyzer. The chemical yield was determined from the ratio of Pu^{240} alpha activity obtained in the final plutonium solution to the amount of Pu^{240} activity expected from the decay of Cm^{244} during the growth period of each experiment. Pu^{238}, formed by the alpha decay of the small amount of Cm^{242} in the original curium, was the only other alpha emitting plutonium isotope present. The over-all yield of plutonium varied between 1 and 15%. The low value, 1%, resulted from the separation of the plutonium isotopes in the mass separator.

In contradistinction to the earlier work,[1] the increased amount of Cm^{247} in the present experiments formed sufficient Pu^{243} that it could be characterized by its half-life and absolute yields could be determined. To be certain that the observed radioactivity was due to Pu^{243} and not some interfering fission product which

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[1] H. Diamond, A. M. Friedman, J. E. Gindler, and P. R. Fields, Phys. Rev. **105**, 679 (1957).
[2] G. W. Wetherill, W. J. Libby, and G. W. Barton, *Abstracts of 43rd Annual Meeting, American Geophysical Union* (American Geophysical Union, Washington, D. C., 1962), p. 68.
[3] V. V. Cherdyntsev, D. P. Orlov, E. A. Isabaev, and V. I. Ivanov, Geokhimiya **10**, 840 (1961).

TABLE II. Experimental results.

Run No.	Type of separations	Plutonium yield (%)	Pu^{243} (dis/min)	Plutonium growth time (h)	Cm^{247} half-life (yr)
I	Chemical	14.6	2.06×10^3	22.3	$(1.42\pm0.12)\times10^7$
II	Chemical	8.3	933	44.2	$(1.75\pm0.13)\times10^7$
III	Mass separated	1.0	124	22.5	$(1.63\pm0.12)\times10^7$
IV	Mass separated	3.1	347	69.2	$(1.75\pm0.11)\times10^7$
	Average value (standard deviation)				$(1.64\pm0.16)\times10^7$
	Average value (95% confidence level)				$(1.64\pm0.24)\times10^7$

might have persisted through the chemical separation, in two of the experiments the plutonium fractions from the curium decay were chemically separated, put into the Argonne mass separator, and the 242-, 243-, and 244-mass fractions collected. The three mass fractions were counted in a low-background (1.5 counts/min) proportional beta counter surrounded by anticoincidence tubes. The 242 and 244 fractions showed no decay whereas the 243 fraction decayed with the characteristic 5 h half-life of Pu^{243}.

Table II shows the results of four different experiments. In the first two experiments the amount of Pu^{243} was determined by counting the purified plutonium in a proportional beta counter. The third and fourth experiments give the results of the mass separated samples. The results of several experiments were rejected because the half-life of the beta activity in the plutonium fraction deviated more than 10% from the accepted 5.0 h half-life of Pu^{243} or the yield of plutonium was very low resulting in a large error in the Pu^{243} beta counting. All the counters were standardized by counting aliquots of a Pu^{243} (produced by neutron irradiation of Pu^{242}) solution whose absolute disintegration rate had been determined by 4π counting.

RESULTS AND CONCLUSION

The lower limit of 4×10^7 yr for the alpha half-life of Cm^{247} obtained earlier[1] was based on the absence of Pu^{243} in a sample containing 2 μg of Cm^{247}. In this case, the search for the Pu^{243} daughter activity was severely complicated by the presence of relatively large amounts of Pu^{238} resulting from the decay of Cm^{242}. To overcome the large Pu^{238} background, a beta-gamma coincidence counting technique was employed to identify Pu^{243}. The result was a decrease in sensitivity for detection of Pu^{243}. One beta-gamma coincidence was found to be equivalent to 232 disintegrations as determined by a standard Pu^{243} sample. Based on many experiments and using the best data, the limit of detection above background by the beta-gamma technique was set at 0.1 count/min. No detectable decay of Pu^{243} could be observed in the activity assigned as background resulting from other plutonium isotopes. In the light of the present experiments, it appears that some of the activity ascribed to background was probably Pu^{243}, and a better limit of detection was 0.2 count/min. The increased amount of Cm^{247} in the present experiments resulted in 30 times as much Pu^{243} being formed. In addition, the greatly reduced background due to the much smaller amounts of Pu^{238} actually made the present determinations approximately 600 times more sensitive than the earlier determinations.

The half-life of 1.64×10^7 yr makes it very unlikely that any primordial Cm^{247} could still exist on earth. As a result of the earlier work it appeared possible that enriched U^{235} samples might conceivably have resulted from the decay of Cm^{247}, but in view of the present work it is unlikely that Cm^{247} could be the source of U^{235} in natural ores such as reported by Cherdyntsev.

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