Isotopes of Berkelium and Californium Produced by Neutron Irradiation of Plutonium

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Samples of berkelium and californium have been separated from neutron-irradiated plutonium and carefully purified. Pure Cf^{249} daughter has been separated from the Bk^{249}. The californium sample and its curium decay products have been mass analyzed. The berkelium fraction contained 314±8 day Bk^{249} emitting 0.114±0.015 Mev beta particles and 5.417±0.015 Mev and 5.03±0.03 Mev alpha particles. The α-β ratio was found to be $(2.2\pm0.3)\times10^{-5}$. The californium sample contained: 360±40 year Cf^{249} emitting 5.808±0.010, 5.93±0.02, and 6.20±0.02 Mev alpha particles; 10.9±0.8 year Cf^{250} emitting 6.020±0.010 Mev alpha particles; 800-year Cf^{251}; 2.55±0.15 year Cf^{252} emitting 6.112±0.015 Mev alpha particles; and 17±1 day Cf^{253}.

INTRODUCTION

ISOTOPES of the elements berkelium and californium were first made by cyclotron irradiations of Am^{241} and Cm^{242} at the University of California.[1,2] More recently, heavier isotopes of these elements, Bk^{249}, Cf^{249}, Cf^{250}, Cf^{251}, Cf^{252}, Cf^{253}, and Cf^{254}, have been produced by successive neutron capture by plutonium irradiated in the Materials Testing Reactor at the National Reactor Testing Station in Idaho[3–5] or in a thermonuclear explosion.[6] To confirm and extend measurements of the nuclear properties of these isotopes, berkelium and californium have been extracted from two 348-mg samples of plutonium which had been irradiated to an integrated neutron flux of 7.5×10^{21} and 1.46×10^{22} neutrons/cm^2 in the MTR. By using the berkelium fraction, the half-life, α-branching ratio, and α- and β-particle energies of Bk^{249} have been measured. Also the half-life and decay scheme of its daughter, Cf^{249}, have been studied. By using the californium fraction, the half-life of Cf^{250}, Cf^{251}, Cf^{252}, and Cf^{253} and the α-particle energies of Cf^{250} and Cf^{252} have been measured.

EXPERIMENTAL

The chemical steps used to isolate and purify the actinide elements from the fission products present in irradiated plutonium have been described in an earlier paper.[7] Individual actinide elements were separated from each other by cation-exchange using Dowex-50 resin columns eluted with either 0.4*M* ammonium lactate at *p*H 4.2[8] or 0.4*M* ammonium α-hydroxyisobutyrate at *p*H 3.9.[9]

Both the routine α and β counting were done with windowless, methane-flow proportional counters. Spontaneous fission counting was done with a methane-filled ionization chamber.[7]

Alpha-particle energy measurements were made by comparison with the α particles from the standards Po^{214}, Po^{218}, Em^{222}, Cm^{242}, and Cm^{244} in a gridded ionization chamber.[10] Sources for this purpose were either vacuum sublimed or electroplated onto polished platinum disks. The α-γ coincidence counter used to study the γ rays from Cf^{249} was similar in principle to that described by Engelkemeir and Magnusson.[11] By using a gridded ionization chamber, pulses from α particles of the desired energy were selected and gated the γ pulses from a 1 in.×1¼ in. cylindrical NaI(Tl) crystal coupled to an RCA 5819 photomultiplier tube. The apparatus was calibrated with Ra^{226} and Am^{241} sources.

The isotopic analysis of the californium was made with a double-focusing mass spectrometer with a surface ionization source and electron multiplier detector.[12]

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[1] Thompson, Ghiorso, and Seaborg, Phys. Rev. **77**, 803 (1950); **80**, 781 (1950).

[2] Thompson, Street, Ghiorso, and Seaborg, Phys. Rev. **78**, 298 (1950); **80**, 790 (1950).

[3] Hulet, Thompson, and Ghiorso, Phys. Rev. **95**, 1703 (1954).

[4] Magnusson, Studier, Fields, Stevens, Mech, Friedman, Diamond, and Huizenga, Phys. Rev. **96**, 1576 (1954).

[5] Diamond, Magnusson, Mech, Stevens, Friedman, Studier, Fields, and Huizenga, Phys. Rev. **94**, 1083 (1954).

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

[7] Jones, Schuman, Butler, Cowper, Eastwood, and Jackson, Phys. Rev. **102**, 203 (1956).

[8] Thompson, Harvey, Choppin, and Seaborg, J. Am. Chem. Soc. **76**, 6229 (1954).

[9] Chopin, Harvey, and Thompson, J. Inorg. & Nuclear Chem. **2**, 66 (1956).

[10] Harvey, Jackson, Eastwood, and Hanna, Can. J. Phys. **35**, 258 (1957).

[11] D. W. Engelkemeir and L. B. Magnusson, Rev. Sci. Instr. **26**, 295 (1955).

[12] F. A. White and T. L. Collins, Appl. Spectroscopy **8**, 17 (1954); **8**, 169 (1954).

TABLE I. Nuclear properties of californium and berkelium.

Nuclide	Half-life	Mode of decay	Particle energies (Mev)	Photon energies (kev)	Spontaneous fission half-life
Bk^{249}	314±8 days	β^-	0.114±0.015		$\geqslant 1.4\times10^9$ yr
		α	5.417±0.015 (96%)		
		$\alpha/\beta=(2.2\pm0.3)\times10^{-3}\%$	5.03 ±0.03 (4%)		
Cf^{249}	360±40 yr	α	5.808±0.010 (92%)	406	$\geqslant 4.5\times10^8$ yr
			5.93 ±0.02 (5%)	107	
			6.20 ±0.02 (3%)	50	
Cf^{250}	10.9±0.8 yr	α	6.020±0.010		
Cf^{251}	~800 yr	α			
Cf^{252}	2.55±0.15 yr	α	6.112±0.015		82±6 yr
Cf^{253}	17±1 days	β^-			

TABLE II. Previously reported values of californium and berkelium properties.

Nuclide	Half-life	Mode of decay	Particle energies (Mev)	Photon energies (kev)	Spontaneous fission half-life
Bk^{249}	290±20 days[a]	β^-	0.08 ±0.02[a]		$\geqslant 2\times10^8$ yr[a]
		α ($10^{-3}\%$)[a]	5.40 ±0.05[a]		
Cf^{249}	470±100 yr[a]	α	5.82[b]	394[c]	$\geqslant 5\times10^6$ yr[a]
			5.91	341	
			6.19	105	
				60	
Cf^{250}	10±2.4 yr[a]	α	6.024±0.005[d]		$(1.5\pm0.5)\times10^4$ yr[a]
			5.980±0.005		
Cf^{252}	2.2±0.2 yr[a]	α	6.112±0.005[d]	100[d]	66±10 yr[a]
			6.069±0.005	40	
Cf^{253}	18±3 days[a]	β^-			

[a] See references 4 and 5.
[b] See reference 16.
[c] See reference 15.
[d] Asaro, Stephens, Harvey, and Perlman, Phys. Rev. **100**, 137 (1955).

RESULTS

A summary of the results of this investigation is given in Table I and for comparison previously reported values are shown in Table II. Comments about individual values in Table I follow.

Berkelium-249.—The half-life was determined by least squares analysis of decay data from two sources prepared from the longer irradiated Pu. Both were observed for at least one half-life. The individual values with their probable errors were 309±3 and 318±2 days; the value given in Table I is the average and the error is about twice the probable error of the mean.

The maximum β-particle energy was estimated from the aluminum absorption curve by Feather's method using C^{14} and S^{35} as references. By pulse analysis two groups were observed in the α decay of Bk^{249}, one at 5.42 Mev and the other at 5.03 Mev.

For the measurement of the α-branching ratio a solution of Bk^{249} freshly separated from Cf^{249} was made. A source was prepared from one aliquot of the solution and the rate of growth of the Cf^{249} α activity into the Bk^{249} was observed for 17 days. The Bk^{249} α-disintegration rate was obtained by extrapolation of the growth curve back to the beginning of the growth period.

To determine the β-disintegration rate four more aliquots of the berkelium solution were taken for 4π β counting. These were prepared by evaporation of the solution onto gold-coated VYNS film.[13] One was treated with insulin solution to aid in the formation of a uniform source and thus minimize self-absorption.[14] To correct the observed rate for self-absorption three sources of different sizes spread over about the same area were made. Extrapolation of their rates to zero source weight gave the rate without self-absorption. The correction for self-absorption introduces the largest error in the measurement and accounts for most of the quoted error. The correction for absorption in the film was made by observing the effect of adding successive backing layers of VYNS film to a source and extrapolating to zero thickness.

Californium-249.—The half-life of Cf^{249} was obtained from the rate of growth of Cf^{249} into a Bk^{249} source of known disintegration rate as described in the previous section. The uncertainty in the β-disintegration rate of the parent accounts for the error in the Cf^{249} half-life.

The α-particle spectrum of Cf^{249} is shown in Fig. 1 and the spectrum of γ rays in coincidence with Cf^{249} α particles is shown in Fig. 2. In addition to the main α group (Fig. 1) two others separated from the main group by about 400 and 120 kev are clearly resolved. Since a 406-kev γ ray is observed in coincidence with Cf^{249} α particles (Fig. 2) it appears that the most abundant α group populates an energy level in Cm^{245} which subsequently decays to the level populated by the 6.20-Mev α group. The γ spectrum also shows small peaks at 50 and 107 kev. Whether these are γ

[13] B. D. Pate and L. Yaffe, Can. J. Chem. **33**, 15 (1955).

[14] B. D. Pate and L. Yaffe, Can. J. Chem. **34**, 265 (1956).

rays or K x-rays from Cm (at 109 kev) and the platinum source mount (at 66 kev) is not certain.

These results agree with earlier work described in the thesis presented by F. S. Stephens, Jr., at the University of California.[15] In addition to the γ rays shown in Fig. 2, he has also observed a peak at 341 kev. In the thesis, reference is also made to unpublished work[16] on the α-particle spectrum of Cf^{249} using an ionization chamber with a Lektromesh collimator which agrees with Fig. 1. Stephens proposed a partial decay scheme for Cf^{249}, essentially that shown in Fig. 3, on the basis of these data. In their study of the decay of Bk^{245}, Magnusson *et al.* refer to work on Cf^{249} decay and state

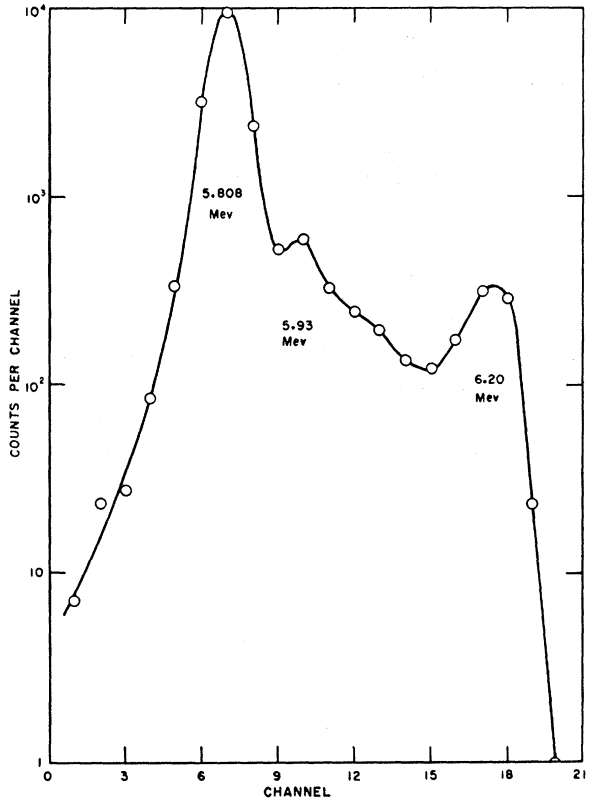

FIG. 1. The α-particle spectrum of Cf^{249}.

that, in addition to the levels shown in Fig. 3, α decay of Cf^{249} also populates a level of Cm^{245} at about 290 kev above the ground state.[17] However, in the absence of more detailed information the decay scheme in Fig. 3 seems to be the most satisfactory summary of the data at present.

Californium-250.—The half-life of Cf^{250} was calculated from the 250–252 mass ratio from mass spectrometric analysis and the 250–252 counting rate ratio from α-particle analysis.

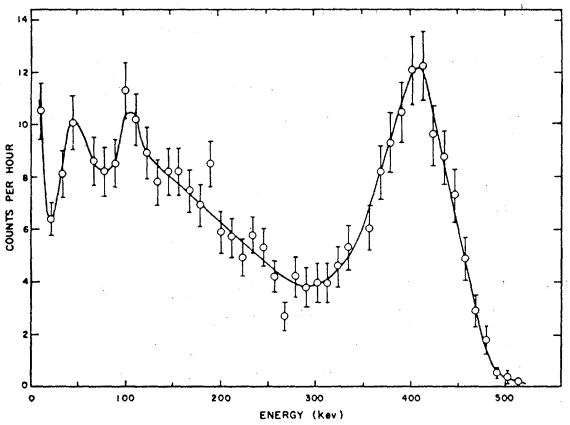

FIG. 2. Typical photon spectrum taken in coincidence with Cf^{249} α particles. The vertical lines on the points indicate the "reliable" or 9/10 errors and are large because the sample is small. The two low-energy peaks are almost within the errors on the individual points. However, these two peaks are observed in all Cf^{249} spectra and are probably real. The averages of several measurements of the energy of the photon peaks are 406, 107, and 50 kev.

Californium-251.—The half-life of Cf^{251} was obtained by comparing the californium 250–251 mass ratio with the curium 246–247 ratio. The curium daughters were allowed to grow into about 3×10^{-10} g of californium, initially free from curium. At the end of the growth period the sample was mass analyzed and both the mass 250–251 and mass 246–247 ratios were measured.

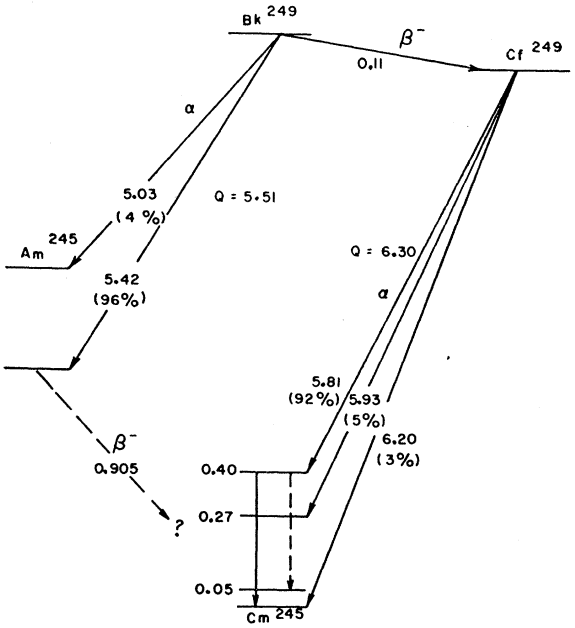

FIG. 3. Partial decay schemes of Cf^{249} and Bk^{249}. The energies are in Mev. Transitions reported in this paper are shown as solid lines; those reported elsewhere are broken lines. The position of the β transition of Am^{245} is uncertain.

[15] F. S. Stephens, Jr., University of California Radiation Laboratory Report UCRL-2970, 1955 (unpublished).
[16] Ghiorso, Harvey, Thompson, and Choppin (unpublished).
[17] Magnusson, Friedman, Engelkemeir, Fields, and Wagner, Phys. Rev. **102**, 1097 (1956).

TABLE III. Isotopic composition of californium from 1.46×10^{22} neutrons/cm² irradiation.

Isotope	Composition when mass analyzed (atom %)	Composition at end of neutron irradiation (atom %)
Cf^{249}	4.16±0.20	3.61
Cf^{250}	36.1 ±1.5	33.2
Cf^{251}	10.3 ±0.6	8.94
Cf^{252}	49.4 ±2.0	54.2

TABLE IV. Yield of californium and berkelium from 348 mg of Pu irradiated in MTR.

Isotope	Irradiation: 7.5×10^{21} neutrons/cm²	Irradiation: 1.46×10^{22} neutrons/cm²
Bk^{249}	2.7×10^{-12} g	3.2×10^{-9} g
Cf^{249}		0.2×10^{-9}
Cf^{250}	1.3×10^{-12}	2.8×10^{-9}
Cf^{251}		0.76×10^{-9}
Cf^{252}	4.1×10^{-13}	4.6×10^{-9}

The value for the former (column 2, Table III) was 3.50±0.24 and that for the latter was 260. Although the Cm-isotope mass ratio was constant over a wide range of filament temperatures, there might have been contributions from impurities since the abundance of the mass-247 peak is so low. The Cf^{251} half-life is thus only approximate.

Ions of ThO^+ from filament impurities contributed to the mass-248 peak and since the 245–246 mass ratio varied with filament temperature some impurity at mass 245 was also present, so that accurate half-lives of Cf^{252} and Cf^{249} could not be obtained from mass spectrometric measurements.

Californium-252.—The half-life of this nuclide was determined by following the decay of the spontaneous-fission counting rate in a Cf source. The decay was followed for 700 days and the data were analyzed by the method of least squares. A small correction was made for the spontaneous fissions contributed by Cf^{250}; using a spontaneous fission half-life of 1.5×10^4 yr[4] for this nuclide the correction was 0.3%. The half-life of Cf^{252} was also measured from the decay of the α-counting rate and the correction for Cf^{250} was made by α-pulse analysis. The half-life agreed with the value quoted, but the error was larger because the Cf^{250} contribution was large.

Californium-253.—The half-life of Cf^{253} was determined by following the formation and subsequent decay of E^{253} α activity in a Cf source by pulse analysis. A half-life of 20.0 days[7] for E^{253} was used in the Cf^{253} half-life calculations.

Yields of Berkelium and Californium.—The yields of isotopes of these elements produced by neutron irradiation of 348 mg of Pu, extrapolated to the end of the bombardment are summarized in Table IV. They have been calculated from the mass analysis of the Cf given in Table III and from the observed disintegration rates using the quoted half-lives. Only the yield of Bk^{249}, Cf^{250}, and Cf^{252} could be obtained for the sample of shorter irradiation because there was insufficient Cf for mass analysis. The values given in Table IV are approximate because chemical yield from the isolation procedure was not known accurately.