Berkelium and Californium Isotopes Produced in Neutron Irradiation of Plutonium

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Californium isotopes of mass numbers 249, 250, 251, and 252 have been identified by mass spectrometric analyses of californium samples produced from neutron-irradiated plutonium. The half-lives of Bk^{249}, Cf^{249}, Cf^{250}, Cf^{252}, and Cf^{253} are 290±20 days, 470±100 years, 10.0±2.4 years, 2.2±0.2 years, and 18±3 days, respectively. The alpha-particle energies of the most prominent alpha peaks for Cf^{249}, Cf^{250}, and Cf^{252} are 5.81±0.03, 6.033 ±0.010, and 6.117±0.010 Mev, respectively. The plot of alpha-particle energies *vs* mass number for the isotopes of californium shows a break (i.e., the alpha energy of Cf^{252} is larger than the alpha energy of Cf^{250}) indicating an irregularity in the nuclear energy surface. The alpha spectra of Cf^{250} and Cf^{252} exhibit characteristic even-even nuclide fine structure peaks. The beta-particle energy of Bk^{249} is 80±20 kev. The spontaneous fission half-lives of Cf^{250} and Cf^{252} are $1.5\pm0.5\times10^4$ and 66±10 years, respectively. Lower limits of 2×10^8 and 5×10^6 years have been placed on the partial spontaneous fission half-lives of Bk^{249} and Cf^{249}. The pile (Materials Testing Reactor at Arco, Idaho) neutron capture cross sections of Bk^{249}, Cf^{250}, Cf^{251}, and Cf^{252} are 350, 1500, 3000, and 25 barns, respectively.

INTRODUCTION

THE preliminary identification of new isotopes of berkelium and californium formed by multiple order neutron capture reactions in the MTR (Materials Testing Reactor at Arco, Idaho) has been reported in a recent communication.[1] These isotopes were found in three plutonium samples, designated henceforth as I, II, and III, which had been subjected to integrated fluxes of 4.2×10^{21}, 1.14×10^{22}, and 1.40×10^{22} neutrons, respectively. Relatively large amounts of transcurium elements were created in the plutonium samples; e.g., about 3×10^{-4} microgram of californium was separated from sample II. The information obtained from measurements of re-irradiated berkelium and californium fractions will be included in this report.

EXPERIMENTAL

The actinide elements produced in neutron irradiation of plutonium were chemically separated from fission products and other impurities by procedures briefly indicated in other publications.[2–4] The transplutonium elements were separated by elution from a hot Dowex-50 cation resin column.[4,5] The nonvolatile solid content of the eluate fractions was reduced for the preparation of radioactive assay plates by organic solvent extractions. Gross amounts of solid impurity were eliminated by extraction of the actinide elements into tributyl phosphate from salted nitric acid solution.[6] Essentially weightless deposits of the actinide elements were formed by evaporating toluene solutions of the thenoyltrifluoroacetone (TTA) chelate complexes of the +3 ions. The chelates are extracted by contacting aqueous solutions of the ions at a *p*H of about 4 with 0.2*M* TTA in toluene.[7] Plates prepared by the TTA method proved to be excellent for alpha-particle pulse analysis since the energy degradation by sample self-absorption is negligible.

Alpha-particle energies were measured with the ionization chamber and twenty-channel pulse-height analyzer described elsewhere.[8] The known alpha-group energies of Cm^{242} (6.110 Mev) and Bi^{212} (6.047 Mev) were used as reference standards. Spontaneous fission counting was done with conventional ionization chambers and scaling circuits.[9]

Isotopic compositions of the californium samples were measured in a 12-in., 60° mass spectrometer with multiple filament surface ionization source.[10] Samples containing as little as 10^{-11} g of californium were successfully analyzed in the mass spectrometer.

The origins and irradiation histories of the various californium samples described in this article are as follows:

I, II, and III were isolated from the initial irradiated plutonium samples I, II, and III, respectively;

IIa was a small fraction of californium sample II which had been left with the berkelium fraction from plutonium sample II for five weeks before chemical separation;

TABLE I. Mass spectrometric analyses of three californium samples giving isotopic abundances in mole percent.

Cf Isotope	Sample II	Sample IIa	Sample IIb
$_{98}Cf^{249}$	4.3±0.5	28±7	< 7
$_{98}Cf^{250}$	49 ±6	34±8	<10
$_{98}Cf^{251}$	11 ±3	8±1	< 6
$_{98}Cf^{252}$	36 ±5	30±3	>77

[1] Diamond, Magnusson, Mech, Stevens, Friedman, Studier, Fields, and Huizenga, Phys. Rev. **94**, 1083 (1954).
[2] Studier, Fields, Sellers, Friedman, Mech, Diamond, Sedlet, and Huizenga, Phys. Rev. **93**, 1428 (1954).
[3] Stevens, Studier, Fields, Mech, Sellers, Friedman, Diamond, and Huizenga, Phys. Rev. **94**, 974 (1954).
[4] Studier, Fields, Diamond, Mech, Friedman, Sellers, Pyle, Stevens, Magnusson, and Huizenga, Phys. Rev. **93**, 1433 (1954).
[5] Fields, Studier, Mech, Diamond, Friedman, Magnusson, and Huizenga, Phys. Rev. **94**, 207 (1954).
[6] Peppard, Gray, and Markus, J. Am. Chem. Soc. **75**, 6063 (1953).
[7] L. B. Magnusson and M. L. Anderson (unpublished results).
[8] D. W. Engelkemeir and L. B. Magnusson, Rev. Sci. Instr. (to be published).
[9] A. H. Jaffey and A. Hirsch (unpublished results).
[10] M. G. Inghram and W. A. Chupka, Rev. Sci. Instr. **24**, 518 (1953).

IIb was a fraction (*ca* 3×10^4 alpha-dis/min) of californium sample II which was reirradiated in the MTR for six weeks (*ca* 10^{21} neutrons);

IIc was separated from reirradiated berkelium (*ca* 6×10^{19} neutrons) which had been isolated from plutonium sample II.

The final chemical purification produced a californium sample with an alpha-disintegration rate of about 2.3×10^5/minute from the neutron-irradiated plutonium sample II. An aliquot, 2.3×10^4 alpha disintegrations/minute, was analyzed in the mass spectrometer. Californium isotopes of mass numbers 249, 250, 251, and 252 were detected in mole percentages given in column 2 of Table I. A small quantity of the californium, about 9×10^3 alpha disintegrations/minute, was left with the berkelium chemical fraction during the initial actinide element separation (californium sample IIa). After a five-week growth period, californium sample IIa was chemically separated from the berkelium and analyzed in the mass spectrometer. The isotopic analysis of this sample is given in Column 3 of Table I. The Cf^{250}/Cf^{252}, Cf^{251}/Cf^{250}, and Cf^{251}/Cf^{252} mole ratios of samples II and IIa are constant within statistical error (Table II). The Cf^{249} in sample IIa has, however, been enhanced

TABLE II. Isotopic enrichment of californium-249 in sample IIa.

Californium isotope ratio	Sample II	Sample IIa	Sample IIa/ sample II
250/252	1.36±0.25	1.13±0.29	0.83±0.27
251/250	0.22±0.07	0.24±0.06	1.05±0.42
251/252	0.31±0.09	0.27±0.04	0.87±0.30
249/250	0.09±0.015	0.82±0.29	9.1 ±3.5
249/251	0.39±0.12	3.50±0.98	9.0 ±3.8
249/252	0.12±0.02	0.93±0.25	7.8 ±2.6

by a factor of about 9. These data are evidence for assigning to Cf^{249} the 5.81-Mev alpha particles observed to grow into a purified berkelium fraction and to elute chemically in the californium position.

A portion of californium sample II containing approximately 3×10^4 alpha disintegrations/minute was reirradiated for six weeks (integrated flux 10^{21} neutrons) in the MTR. After irradiation this californium (sample IIb) was again chemically separated, and a major part of the sample was used for a mass spectrometric analysis. The quantity of this sample was much smaller than the quantities of the other californium samples examined in the mass spectrometer, therefore the results reported in column 4 of Table I are limits.

ALPHA-PARTICLE AND SPONTANEOUS FISSION INTENSITIES

Alpha-pulse analysis of californium sample II showed two prominent alpha groups of 6.03 and 6.12 Mev. The calibration of alpha-particle energies is presented in the next section of this paper. The 6.12-Mev alpha peak in californium sample II was 77±2 percent of the combined 6.03 and 6.12-Mev alpha peaks as

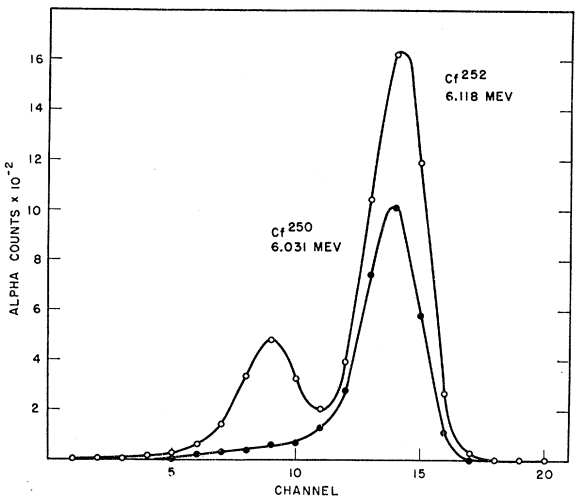

FIG. 1. Californium alpha spectra. Alpha counts per alpha-pulse analyzer channel *vs* channel number which is a linear function of energy. The open circles (○) and solid circles (●) represent the alpha-pulse analyses of californium samples II and IIb, respectively. The samples were deposited on 2-mil platinum by evaporating TTA-toluene solutions of californium. The alpha particles were collimated with a $\frac{1}{8}\times1$-inch copper ring.

shown in Fig. 1. Californium samples I and III contained 38±4 and 82±2 percent, respectively, of 6.12-Mev alpha particles. Californium sample II was reirradiated (integrated flux *ca* 10^{21} neutrons) for six weeks in the MTR. The alpha spectrum of the resulting californium sample (californium sample IIb) is shown in Fig. 1 and contains about 97±2 percent 6.12-Mev alpha particles.

The ratios of alpha disintegrations (6.03-plus 6.12-Mev) to spontaneous fission disintegrations for californium samples I, II, III, and IIb are 74±5, 39±1, 36 ±1, and 32±1, respectively. As the percentage of 6.12-Mev alpha particles increases, the alpha-to spontaneous fission disintegration ratios decrease, indicating that a major fraction of the spontaneous fission activity is associated with the isotope emitting the 6.12-Mev alpha particles, or other higher mass isotopes whose alpha particles were not detected.

A sample of berkelium-249, 3.6×10^4 beta disintegrations/minute, chemically separated from traces of californium impurity was irradiated in the "rabbit" of the MTR for 3.5 days. Prior to irradiation no spontaneous fission events were observed in this berkelium sample during one day of fission counting. The spontaneous fission counting rate is therefore less than 1.3×10^{-3} disintegration/minute (standard error), and the calculated californium alpha disintegrations/minute is less than 0.05. Approximately sixty californium alpha-disintegrations/minute were produced by the irradiation which were by activity greater than 98 percent 6.03-Mev alpha particles (californium sample IIc). The californium alpha disintegrations/minute to spontaneous fission disintegrations/minute ratio of this sample was 875±26.

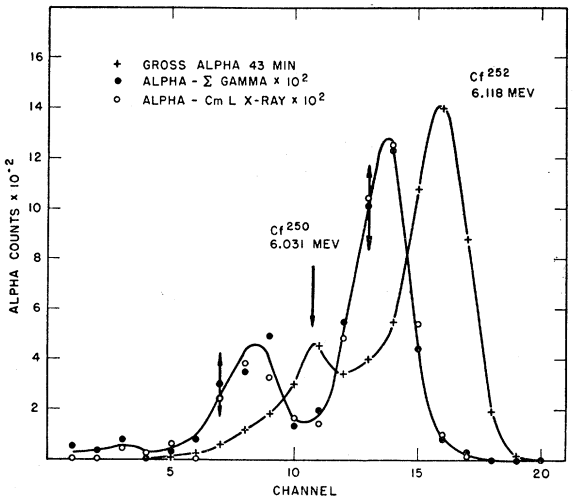

FIG. 2. Gamma-alpha coincidence spectra of Cf^{250} and Cf^{252}. The crosses (+) are the gross californium alpha counts. The solid circles (●) are the alpha-Σgamma (>10 kev) coincidence counts and the open circles (○) are the alpha—Cm *L* x-ray (14–23 kev) coincidence counts. The sample was deposited on 0.5-mil aluminum foil by evaporation of a tributyl phosphate solution. The alpha particles were collimated by covering the foil with 2-mil Lektromesh screen.

ALPHA- AND BETA-PARTICLE ENERGIES

The californium alpha-particle energies were calibrated by intercomparisons of samples II, IIa, IIb, and IIc and the 6.047-Mev and 6.110-Mev alpha-particle groups of Bi^{212} and Cm^{242}, respectively.

Pb^{212} recoil atoms from an emanating source of RdTh were caught on a sample of californium II deposited by evaporation and ignition of a drop of ammonium citrate solution from a column elution. The alpha particles were collimated by covering the platinum sample disk with 2-mil Lektromesh screen.[8] With this type collimator the 6.047-Mev alpha particle[11] of Bi^{212} can be used as an energy standard for ion pulse analysis since the conversion electron energy liberated in the deexcitation of the daughter Tl^{208} atom is not added to the alpha-particle ionization.[8] The intensity of the Bi^{212} alpha particles was adjusted by the length of recoil collection time to be several times greater than the lower intensity californium alpha particle (6.03 Mev). The position of the resultant pulse-analyzer peak was essentially governed by the 6.047-Mev Bi^{212} alpha-particles. The higher intensity californium alpha particle was found to be *ca* 60 kev more energetic than the 6.047-Mev Bi^{212} alpha particles. The energy separation of the prominent californium peaks was *ca* 80 kev (no Bi^{212} present). The californium alpha-particle energies obtained with this sample are known to be too small but are valuable supporting evidence for the energies measured with the Cm^{242} standard. The californium peak widths (full width at half the maximum height) in the ignited citrate sample were 50 kev, whereas the normal width is about 30 kev for a thin sample.

The alpha-particle energy of the californium (sample IIc) produced by neutron irradiation of berkelium was compared with the Cm^{242} 6.110-Mev alpha particle.[12] Both alpha emitters were mounted on pieces of 2-mil platinum by the TTA method. The platinum pieces were placed within a single collimating ring. Although both the californium and curium have branching decay to excited states, as will be shown in the next section only single ion pulse peaks appear for each of the two alpha-emitters. The peak energy is that for the alpha particle emitted in decay to the ground state. The californium alpha particle was found to be 77 kev less energetic than the Cm^{242} alpha particle. In other comparisons, the alpha-particle energy of sample IIc was identical with that of the lower energy alpha particle of samples II and IIa and less energetic than the higher energy alpha particle in samples II, IIa, and IIb by 84 kev. Relative to the Cm^{242} alpha-particle energy of 6.110 Mev, the energies of the californium alpha particles are 6.033 and 6.117 Mev. The uncertainty in the relative peak positions in the pulse spectra were estimated to be equivalent to less than 5 kev.

Californium sample IIa also contained a small alpha-particle peak at 5.81 Mev. A peak of this energy was observed to grow into a beta-emitting berkelium sample which had previously been separated from all detectable californium alpha activity. The 5.81-Mev peak appeared to be the only one present. The limit of detection of any other peak in the sample was estimated to be less than 5 percent of the 5.81-Mev peak intensity.

Absorption measurements showed the energy of the Bk^{249} beta particles to be 80 ± 20 kev. A Bk^{249} sample of 1.5×10^5 beta disintegrations/minute also contained an alpha-particle peak (*ca* 2 disintegrations/minute) at 5.40 ± 0.05 Mev. The alpha- to beta-branching ratio of Bk^{249} is therefore about 10^{-5}.

MASS ASSIGNMENTS OF THE 6.03- AND 6.12-MEV ALPHA-PARTICLE GROUPS

It has been shown (see Table I and reference 1) that californium-249 emits a prominent alpha group with energy of 5.81 Mev. The 6.03-Mev and 6.12-Mev alpha groups are assigned to Cf^{250} and Cf^{252}, respectively, from the following arguments.

(1) Californium produced by a short irradiation (3.5 days) of Bk^{249} showed an alpha activity consisting almost entirely (>98 percent) of 6.03-Mev particles.

(2) Since the 6.12/6.03-Mev alpha-activity ratio increased with the integrated neutron flux received by the plutonium, the 6.12-Mev activity is probably a higher order capture product than the 6.03-Mev activity.

(3) Re-irradiation of californium sample II for six weeks (californium sample IIb) depleted the 6.03-Mev

[11] A. Rytz, Compt. rend. **233**, 790 (1951).

[12] Asaro, Reynolds, and Perlman, Phys. Rev. **87**, 277 (1952).

activity relative to the 6.12-Mev activity. The mass spectrometric analysis of californium sample IIb also showed the mass 250 depleted relative to the 252.

(4) The alpha-particle spectrum of californium sample II was found to have characteristics to be expected for the decay of two even-even nuclides. Figure 2 shows the alpha-particle spectrum measured in coincidence with electromagnetic radiations from a sample collimated with Lektromesh screen. The spectrum in coincidence with radiation in the energy range of curium L x-rays (14 to 23 kev) was the same, within the statistical uncertainty, as that in coincidence with all radiations of energy greater than *ca* 10 kev. In a corollary measurement, no significant coincident rates were detected between alpha particles and electromagnetic radiation of energy greater than the curium L x-rays. The two alpha particles observed in the gross spectrum are not seen in the coincident spectrum. These alpha particles (6.033 and 6.117 Mev) must be emitted in decays to the ground states of two curium isotopes. The coincident spectrum also shows two alpha-particle peaks but with energies of *ca* 5.99 and 6.08 Mev. The ratio of the intensity of the 5.99-Mev particle to that of the 6.08-Mev particle is approximately the same as the intensity ratio of the 6.033-Mev and 6.117-Mev particles. The alpha particles coincident with L x-rays appear to be emitted in decays to the first excited states (40-45 kev) of the two curium isotopes. The 40–45 kev energies of the first excited states, which are deduced from the separations of the coincident and gross alpha-particle peaks, must be highly converted in the L shells of the curium isotopes. A first excited state energy of *ca* 40 kev and high conversion in the transition to ground appear to be universal properties of the known even-even nuclides with atomic number greater than 90.[13] The percentage of branching decay to the first excited state was measured by comparing the L x-ray—alpha-particle coincidence rates of californium sample II and a plutonium-238 sample under the same conditions. From the known branching of Pu^{238} (24 percent)[14] the branching in the decays of both californium isotopes was calculated to be about 10 percent, an unusually small percentage for the decay of even-even nuclides in this region.

The lack of evidence for alpha particles from Cf^{251} is not surprising. The mass of Cf^{251} is low relative to the masses of Cf^{250} and Cf^{252} in sample II and the odd neutron nuclide as a rule has a much longer half-life than the neighboring even-even nuclides. The resolution of the alpha-particle pulse analyzer is not high enough to detect very low intensity particles.

The mass assignments of Cf^{250} (6.03-Mev alpha particles) and Cf^{252} (6.12-Mev alpha particles) have important implications in that the plot of alpha-particle energies *vs* mass number for the isotopes of californium shows an irregularity in this region. Except for the large irregularities in the alpha energies near the closed shell $N=126$, the plots of alpha-particle energies *vs* mass number for other elements exhibit a monotonic decrease in alpha-particle energy with increasing mass.[15] Ghiorso, Thompson, Higgins, Harvey, and Seaborg[16] have used the break in the alpha energies of the californium isotopes to postulate a subshell at $N=152$.

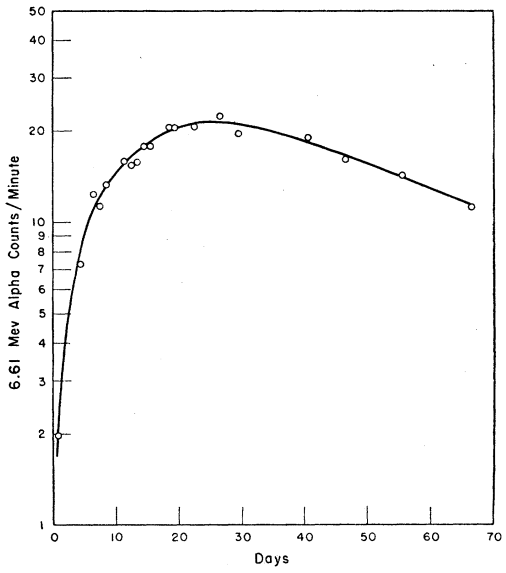

FIG. 3. Growth and decay of 6.61-Mev alpha particles in a chemically separated californium sample as a function of time.

$$Cf^{253} \xrightarrow{\beta^-} 99^{253} \xrightarrow[6.61\ \text{Mev}]{\alpha}.$$

Alpha particles of 6.61-Mev energy have been observed to grow into a chemically separated californium fraction (see Fig. 3). From the growth and decay of the 6.61-Mev alpha particles, a beta half-life of 18 ± 3 days has been calculated for the beta-emitting californium isotope. The mass of this isotope is greater than 252 since two mass spectrometer analyses of a californium sample separated by five weeks showed Cf^{251} to have a beta half-life much longer than 18 days or to be beta stable. The yield of the 18-day beta-emitting isotope was about one-hundredth that of Cf^{252} in californium sample II. Californium-254 would be predicted to be beta stable, and such a large yield of Cf^{255} is very unlikely. In addition, alpha-decay systematics favor mass 253 over 255 for the 6.61-Mev alpha particles of element 99. Thus the 18-day beta-emitting isotope of californium is almost certainly mass 253. An attempt to identify the Cf^{253} beta particles has not been successful owing to the large amount of spontaneous fission activity in the californium.

[13] F. Asaro and I. Perlman, Phys. Rev. **91**, 763 (1953).
[14] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).
[15] Perlman, Ghiorso, and Seaborg, Phys. Rev. **77**, 26 (1950).
[16] Ghiorso, Thompson, Higgins, Harvey, and Seaborg, Phys. Rev. **95**, 293 (1954).

TABLE III. [Alpha disintegrations/minute]/[spontaneous fission disintegrations/minute] ratios for five californium samples.

Californium sample	$\frac{(6.03+6.12)\text{-Mev } \alpha \text{ dis/min}}{\text{spon. fission dis/min}}$	$\frac{6.12\text{-Mev } \alpha \text{ dis/min}}{(6.03+6.12)\text{-Mev } \alpha \text{ dis/min}}$	$\frac{6.12\text{-Mev } \alpha \text{ dis/min}}{\text{spon. fission dis/min}}$
I (from plutonium which received 4.2×10^{21} neutrons)	74+5	0.38±0.04	28±4
II (from plutonium which received 1.14×10^{22} neutrons)	39±1	0.77±0.02	30±1
III (from plutonium which received 1.40×10^{22} neutrons)	36±1	0.82±0.02	30±1
IIb (sample II re-irradiated with 10^{21} neutrons)	32±1	0.97±0.02	31±1
IIc (berkelium re-irradiated with 6×10^{19} neutrons)	875±26	0.013±0.004	

HALF-LIVES

The half-life of berkelium-249 has been measured by following the decay of its beta activity. The best half-life value is 290±20 days. The californium-249 half-life has been calculated from the growth of alpha activity (0.6 disintegration/minute per day) into a berkelium-249 sample of 1.5×10^5 beta disintegrations/minute. A berkelium-249 counting efficiency in a Bradley PC-10 counter of 56 percent from a calibration curve of beta energy *vs* counting efficiency[17] was used. The half-life of Cf^{249} from the above data is 470±100 years.

An aliquot of the californium sample II was followed for the decay of the spontaneous fission activity. The spontaneous fission activity decayed with a 2.2±0.2-yr half-life as shown in Fig. 4. Data from Table III (compare samples II and IIc) and Table I show that the fraction of spontaneous fission events due to Cf^{250} in the above sample is less than 1.3 percent [$<(0.23/875)/0.77/39)=0.013$]. The spontaneous fission half-life of Cf^{249} is long, and it is not expected that the odd-neutron nuclides, Cf^{251} and Cf^{253}, would contribute to the spontaneous fission activity. It is also improbable that higher even A californium isotopes (e.g., Cf^{254}) are contributing to the spontaneous fission activity. The integrated neutron flux received by californium samples I, II, III, and IIb varied by a factor of about three. The spontaneous fission activities (Cf^{250} spontaneous fission contribution negligible in each of those samples) of each of the above samples are directly proportional to their 6.12-Mev alpha activities (Cf^{252}). One would not expect this relation with neutron flux if higher order capture products (Cf^{254}, etc.) were making a measurable spontaneous fission contribution. From these considerations the decay of the spontaneous fission activity may be taken as a measure of the Cf^{252} half-life, although the main mode of decay of Cf^{252} is by alpha-particle emission. The direct measurement by pulse analysis of decay of the Cf^{252} alpha activity was not attempted during the relatively short time of these experiments.

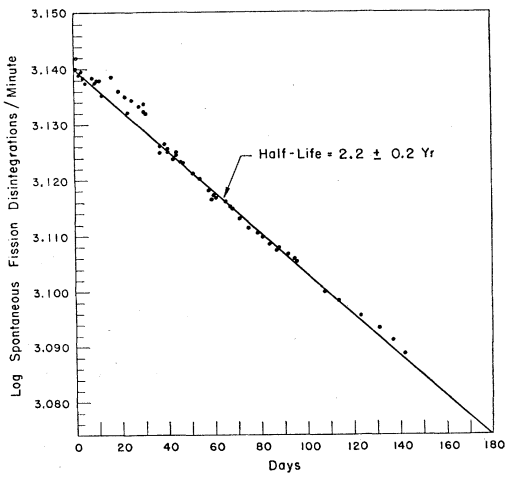

FIG. 4. Decay of Cf^{252}. The log of the spontaneous fission disintegrations/minute of californium sample II is plotted as a function of time.

[17] We are indebted to Kevin Flynn for the absolute beta-disintegration rate of this berkelium sample.

The half-life of Cf^{252} (6.12-Mev alphas) has also been determined relative to Cf^{249} from californium sample IIa with the aid of an alpha-pulse analysis and a mass spectrometric determination. Californium sample IIa was found to have 0.34±0.10 percent 5.81-Mev alpha activity. Employing the mass spectrometric data of Table I, a 470±100-yr half-life for Cf^{249}, and alpha-pulse analysis data showing the sample to be 77 percent by activity 6.12-Mev alpha particles one calculates a half-life for Cf^{252} of 2.2±1.0 years. Although the experimental uncertainty is large, this half-life value agrees with the previous value. The half-life of Cf^{250} is calculated from mass spectrometric and alpha-pulse analyses of californium sample II to be 4.56±1.00 times longer than that of Cf^{252}, or 10.0±2.4 years.

SPONTANEOUS FISSION HALF-LIVES

The Cf^{252} alpha-/spontaneous fission disintegration ratios from californium samples I, II, III, and IIb are

TABLE IV. Nuclear properties of some isotopes of berkelium and californium.

Isotope	Radiation	Half-life	Alpha-particle energies (Mev)	Spontaneous fission half-life (years)
$_{97}Bk^{249}$	β^-, α branching ratio $\beta^-/\alpha \approx 10^5$	290±20 days	5.40±0.05	$\geqslant 2\times10^8$
$_{97}Bk^{250}$	β^-	<12 hours		
$_{98}Cf^{249}$	α, α/s.f. $\geqslant 1.2\times10^4$	470±100 years	5.81±0.03	$\geqslant 5\times10^6$
$_{98}Cf^{250}$	α, spontaneous fission (s.f.) α/s.f. = 1460±350	10.0±2.4 years	6.033±0.010 (90%) 5.99±0.01 (10%)	1.5±0.5×10⁴
$_{98}Cf^{251}$	(α)	...	...	...
$_{98}Cf^{252}$	α, spontaneous fission α/s.f. = 30±1	2.2±0.2 years	6.117±0.010 (90%) 6.08±0.01 (10%)	66±10
$_{98}Cf^{253}$	β^-	18±3 days	...	...

28±4, 30±1, 30±1, and 31±1, respectively. The partial spontaneous fission half-life of Cf^{252} is calculated to be 66±10 years. The Cf^{250} alpha-/spontaneous fission disintegration ratio is subject to a greater error since a very small change in the calculated content of Cf^{252} in the sample changes the ratio considerably. Californium sample IIc contains 1.3±0.4 percent Cf^{252} (Fig. 5), and the gross californium alpha-to spontaneous fission disintegration ratio is 875±26. Making the correction for the contributions from Cf^{252}, one calculates that the alpha-to spontaneous fission disintegration ratio for Cf^{250} is 1460±350. This ratio and the alpha-decay half-life gives a partial spontaneous fission half-life for Cf^{250} of $1.5\pm0.5\times10^4$ years.

A sample of Bk^{249} (1.5×10^5 beta disintegrations/minute) was fission counted for seven days following chemical separation of the berkelium (a small quantity of Cf^{249} grew in during this period), in which time four spontaneous fission events were recorded. Some or all of these fission events may have been background or Cf^{249} spontaneous fission. The partial half-life for spontaneous fission of Bk^{249} is therefore equal to, or greater than, 2×10^8 years.

Five alpha disintegrations/minute of Cf^{249} were fission counted for 10^4 minutes, in which time 4 spontaneous fission events were observed. The alpha-to spontaneous fission disintegration ratio is equal to or greater than 1.2×10^4. Employing an alpha-decay half-life of 470 years for Cf^{249} gives a lower limit to its spontaneous fission half-life of 5×10^6 years.

The measured spontaneous fission half-lives of Cf^{250} and Cf^{252} do not agree with values predicted for even-even nuclides from the plots of Seaborg[18] and Whitehouse and Galbraith.[19] These authors plotted the logarithms of the spontaneous fission half-lives of several even-even nuclides as a function of their Z^2/A values. Cf^{252} has a shorter spontaneous fission half-life than Cf^{250}, which supports the idea that the even-A isotopes of each even-Z element go through a maximum stability for spontaneous fission as A increases.[20,21]

The spontaneous fission decay rates of the heavy odd-nucleon isotopes are several factors of ten slower than corresponding even-even nuclides, in agreement with previously measured spontaneous fission half-lives of other odd-nucleon isotopes.

PILE NEUTRON CROSS SECTIONS

A sample of Bk^{249} with a disintegration rate of approximately 3.6×10^4/minute was irradiated for 3.5 days in the "rabbit" of the MTR. Cf^{250} with 60 alpha disintegrations/minute was produced. Assuming a flux of 2×10^{14} neutrons/cm²−sec, one calculates the pile neutron capture cross section of Bk^{249} to be 350 barns.

From the amount of Cf^{253} present in a sample of californium (containing isotopes of mass numbers 249,

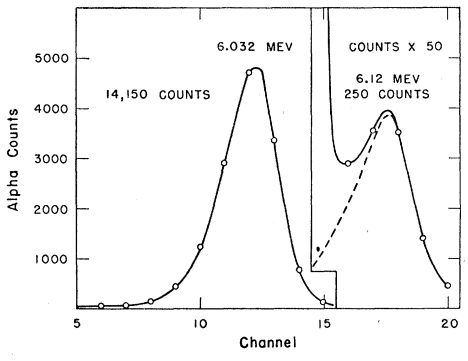

FIG. 5. Alpha-pulse analysis of californium sample IIc. This sample contains 1.3±0.4 percent 6.12-Mev alpha particles (Cf^{252}). The sample was deposited on 2-mil platinum by evaporating a TTA-toluene solution.

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250, 251, and 252) irradiated for six weeks in the MTR, the pile neutron capture cross section of Cf^{252} was calculated to be 25 barns.

Crude calculations of the pile neutron capture cross sections of Cf^{250} and Cf^{251} from mass spectrometric data indicate that these values are about 1500 and 3000 barns, respectively.

A summary of the nuclear properties of the isotopes of berkelium and californium observed in these experiments is given in Table IV.

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Decay of $_{22}Ti^{51}$ (5.8 min)

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The radiations from Ti^{51} have been studied with a scintillation coincidence spectrometer. Gamma rays of 0.32, 0.61, and 0.92 Mev are resolved. Their intensities are estimated to be in the ratio 100:1:5, respectively. The 0.32- and 0.61-Mev gamma rays are in coincidence. Two beta-ray components are resolved; one of approximately 2.3 Mev is in coincidence with the 0.32-Mev gamma ray, and one of 1.8 Mev is in coincidence with the 0.92-Mev radiation. No indication of a beta transition to the ground state of V^{51} is observed.

A SHORT-LIVED radioactivity induced in titanium by irradiation with deuterons and neutrons was first observed by Walke.[1] His tentative assignment to Ti^{51} has been confirmed.[2] A 72-day activity at first believed to be isomeric has been shown[3,4] to be due to contamination of the source. From a careful measurement,[5] the half-life of the short activity is now known to be 5.79±0.03 min. Gamma radiation of 0.32 Mev has been detected by several experimenters.[6–8] Bunker and Starner[8] have reported additional weak gamma rays of 0.60 and 0.915 Mev. They find that the 0.60-Mev radiation is in coincidence with the 0.32-Mev gamma ray and assume the 0.915-Mev gamma ray to be the cross-over transition. A marked disagreement exists between different measurements of the beta transition energies and hence of the total disintegration energy. Der Mateosian[9] found the beta spectrum to be complex, consisting of two components with energies of 1.85 and 2.1 Mev. He observed coincidences between the 1.85-Mev beta ray and the 0.32-Mev gamma ray. Other investigators[2,10] have also reported the maximum beta energy to be about 1.7 Mev. A disintegration energy of 1.7 Mev is much smaller than that expected from a consideration of the energy systematics of odd-mass beta emitters. Koester *et al.*[7] also find the beta spectrum to be complex but report the maximum beta energy to be 2.2 Mev. They conclude that this transition leads to the ground state.

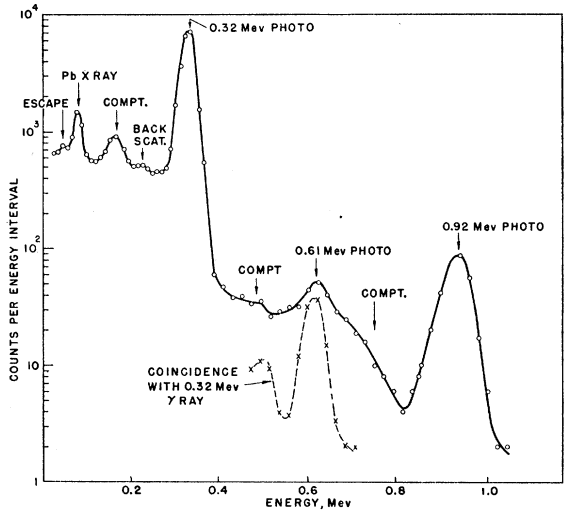

FIG. 1. NaI(Tl) pulse-height distribution of Ti^{51} (5.8 min).

We have undertaken an investigation of the decay of Ti^{51} using our ten-channel scintillation coincidence spectrometer. NaI (Tl) crystals ($2\frac{1}{4}$ in.×$2\frac{1}{4}$ in.×$2\frac{1}{8}$ in.) were used for the detection of gamma radiation, and an anthracene crystal was used for the detection of beta radiations. The anthracene is provided with a thin aluminum window (<0.001 in.). Sources were

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