Nuclear Decay Properties of Heavy Nuclides Produced in Thermonuclear Explosions–Par and Barbel Events*

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The nuclear decay properties of heavy nuclides ($A \leq 257$) produced in two low-yield thermonuclear explosions, the Par and Barbel events, have been studied with the following results: The α-decay branching of Cf^{253} has been observed, $E_\alpha = 5.978 \pm 0.005$ MeV, $\alpha/(\alpha+\beta^-) = (0.31 \pm 0.04)\%$. The α-decay branching of Es^{255} has been observed, $E_\alpha = 6.300 \pm 0.003$ MeV, $\alpha/(\alpha+\beta^-) = (8.5 \pm 0.3)\%$. The spontaneous fission half-life of Cm^{250} was remeasured and found to be $(1.74 \pm 0.24) \times 10^4$ years. Upper limits for the half-lives of Cm^{252} and Bk^{251} have been set at 2 and 3 days, respectively. The existence of 80-day Fm^{257} was confirmed; a sample of Fm^{257} from the Par event decayed with a half-life of 94 ± 10 days. Attempts to produce and detect Fm^{258} by irradiating Par Fm^{257} in a reactor neutron flux were unsuccessful. Upper limits for the half-life of Fm^{258} were determined.

I. INTRODUCTION

THE production of heavy nuclides ($A \leq 257$) in two low-yield thermonuclear explosions, the Par[1] and Barbel[2] events, has been described in earlier reports,[3,4] The experimental yields for each mass number have been reproduced in theoretical calculations assuming time-integrated fluxes in the range $(4.5-7) \times 10^{24} n/cm^2$.[3,5] These fluxes in U^{238} targets were the highest achieved in the current program and showed a marked improvement over the flux produced in the Mike event of 1952. The improvement is evident in Fig. 1, where the yield-versus-mass curves for Par and Barbel are seen to descend more gradually than the curve for Mike. Moreover, the production of mass-254 atoms/kton in the 30-kton Par event exceeded that in the 10,000-kton Mike event by a factor of 10.

This report presents some new nuclear data obtained from an analysis of the debris from the two recent experiments. Additional information has been acquired on the decay of Cf^{253}, Es^{255}, and Cf^{251}; the spontaneous fission half-life of Cm^{250} has been measured; the existence of Fm^{257} has been confirmed; and upper limits

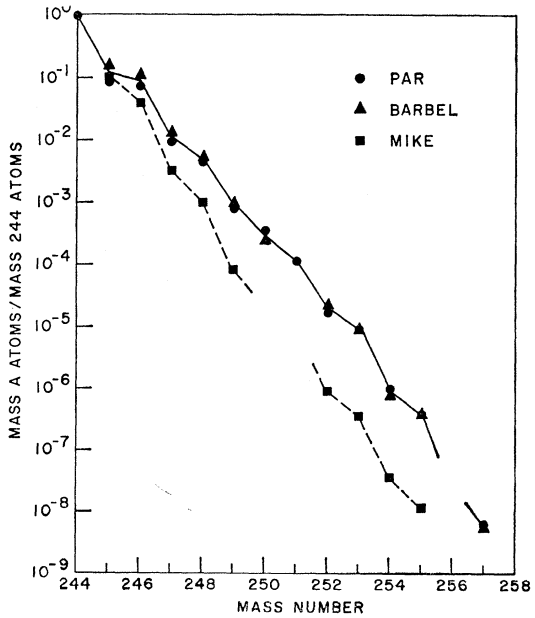

FIG. 1. Heavy nuclide yield for $A = 244-257$, plotted relative to mass 244 yield, for the Par, Barbel, and Mike explosions.

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[1] The Par device was designed by a physics group at Livermore which included D. W. Dorn, L. A. Maizitis, and J. S. Ingley. The Par device test and recovery of debris samples were performed by personnel from the Lawrence Radiation Laboratory, Livermore, California.

[2] The Barbel device was designed by the Los Alamos Theoretical Division under the direction of J. C. Mark. The Barbel device test and recovery of debris samples were performed by personnel from the Los Alamos Scientific Laboratory, Los Alamos, New Mexico.

[3] D. W. Dorn and R. W. Hoff, Phys. Rev. Letters 14, 440 (1965).

[4] Los Alamos Radiochemistry Group, Phys. Rev. Letters 14, 962 (1965).

[5] G. I. Bell, Phys. Rev. 139, B1207 (1965).

have been set on the half-lives of Bk^{251}, Cm^{252}, and Fm^{258}.

II. EXPERIMENTAL METHODS

Since the Par and Barbel tests were fired within a week of each other in October 1964, samples were received and studied concurrently from both events. Samples of the fused rock were recovered by drilling to the appropriate depths, 1550 ft for Par and 880 ft for Barbel. These samples were distributed to groups at the four laboratories for analysis. From the initial recovery effort, samples were received at the laboratories 2 to 4 days after the detonations. Quantities of the order of 100 g of rock which contained fractions in the range $(3 \text{ to } 10)\times10^{-9}$ of the total were analyzed promptly. As larger amounts of sample became available, these were also processed to yield actinide element fractions. Ultimately, 10 kg of rock from the Par event were treated. Although the chemical yield (70%) in this processing was reasonable, the 10 kg of rock was less rich in radioactive debris, on the average, than some of the smaller samples studied earlier. Hence, this processing resulted in the purification of about 10^{-7} of the total actinides produced in the explosion.

The rock samples were treated by a variety of chemical procedures at the various laboratories. An early step in each flow sheet was treatment of the rock with hydrofluoric acid, either aqueous or anhydrous, at elevated temperatures, to remove silica. Further purification was accomplished with a variety of ion-exchange and solvent-extraction procedures. Final separation of the individual actinides was usually accomplished in the elution of a Dowex 50 ion-exchange column with α-hydroxyisobutyric acid at 85°C.

Decay of α-emitting samples was followed on Frisch-grid ionization chambers. Detailed α spectra were measured with gold-silicon surface-barrier detectors. Spontaneous fission rates were measured in ionization chambers with extremely low backgrounds, e.g., ≤ 1 event per week.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Isotopic Content of Californium and Einsteinium

Production of heavy nuclides in thermonuclear explosions differs from that in nuclear reactors in two respects–the energy of the neutrons captured and the time during which capture occurs. These have an important bearing on measurements of nuclear decay properties. In a nuclear explosion, the mean neturon energy in the target is assumed to be 10 to 20 keV while the neutrons are being captured.[5,6] Neutron cross sections at these energies are relatively regular. The entire sequence of neutron captures is accomplished in less than a μsec, i.e., before any significant amount of β decay can occur. Thus, each of the high-mass nuclides observed in the laboratory has been produced by a series of β decays that occurred after the neutron-capture sequence. The regularity of the capture cross sections is is reflected in the regularity of the yields of the observed products, as shown in Fig. 1. The experimental errors on the points plotted in this figure are equal to or less than the actual size of the symbols used; hence, no error bars are shown.

The situation is different in the neutron spectrum of a reactor where the yields of heavy nuclides are a function of neutron-capture and neutron-induced fission cross sections at reactor thermal energies, the values of which may vary by large factors for different nuclides. Neutron capture is accomplished in fluxes of the order of 10^{15} n/cm^2 sec and appreciable conversion of targets such as Pu^{242} to heavy nuclides, e.g., Fm^{257}, requires months to years of irradiation. With this time scale for neutron capture, spontaneous fission can also become an important means of destruction for certain nuclides. Thus, the relative yields of nuclides in reactor-produced materials can be considerably different from those found in debris from nuclear explosions. These differences are illustrated in Table I where the abundances of the Par and Barbel californium and einsteinium isotopes are compared with similar isotopic abundances from a curium target which had received a total exposure (nvt) of 3.1×10^{22} n/cm^2 in a reactor.[7]

B. The α Decay of Cf^{253}

The α-decay branching of Cf^{253}, a previously unobserved mode of decay for this nuclide, was measured in samples of californium recovered in the Par and Barbel events. Cf^{253} was discovered during the initial studies of the heavy element fractions from the "Mike" thermonuclear test in November 1952.[8] The best value of its half-life, 17.6 ± 0.2 days, was measured by observing the growth of 6.63-MeV α particles of the Es^{253} daughter into a sample of purified californium.[9]

The α spectrum of the Par californium is shown in Fig. 2. The α group at 5.98 MeV was resolved from the higher energy Cf^{252} α particles by subtracting the proper peak shape derived from a standard sample of Cf^{252} free of Cf^{253}. This group at 5.978 ± 0.005 MeV decayed with a 17 ± 2-day half-life. The energy of this group was measured by using the most abundant α groups of Cf^{252} at 6.112 MeV and Es^{253} at 6.633 MeV as standards. This α group is assigned to Cf^{253} because its observed half-life is consistent with that measured for Cf^{253} β decay, and because the α-particle energy is consistent with a predicted value, 6.06 MeV, for Cf^{253} derived from

[6] D. W. Dorn, J. S. Ingley (private communication).

[7] R. M. Latimer and J. T. Haley, University of California, Lawrence Radiation Laboratory, Berkeley, Report No. UCRL-16191, 1965 (unpublished).

[8] P. R. Fields *et al.*, Phys. Rev. **102**, 180 (1956).

[9] D. N. Metta *et al.*, J. Inorg. Nucl. Chem. **27**, 33 (1965).

TABLE I. Isotopic abundances of Es and Cf from reactor irradiation of a heavy curium target and from Par and Barbel debris. SF≡spontaneous fission.

Nuclide	$t_{1/2}$	Radiations	Atom ratio at end of irradiation: Cm irradiations, reactor	Par debris	Barbel debris
Cf^{249}	360 yr	5.81-MeV α	0.0023	0[a]	0[a]
Cf^{250}	13.2 yr	6.02-MeV α	0.122	0[b]	0[b]
Cf^{251}	800 yr	5.84-MeV α	0.028	5.9	...
Cf^{252}	2.65 yr	96.9% 6.11-MeV α 3.1% SF	1.000	1.000	1.000
Cf^{253}	17.6 days	99.7% β^- 0.3% 5.98 MeV α	0.016	0.50	0.42
Cf^{254}	60.4 days	SF	0.00043	0.055	0.034
Cf^{253}	17.6 days	β^-	1.000(Es^{253})	1.000[c]	1.000[c]
Es^{253}	20.8 days	6.63 MeV α			
Es^{254}	38.5 hr	β^-	0.043	0[d]	0[d]
Es^{254}	270 days	6.43 MeV α	0.0031	0[d]	0[d]
Es^{255}	40 days	92% β^- 8% 6.30 MeV α	0.00031	0.039	0.041

[a] The 314-day Bk^{249} decays by β emission to Cf^{249}. Samples of californium will show varying amounts of Cf^{249}, depending upon when the berkelium and californium are separated from each other.
[b] β decay of Cm^{250} has not been observed; either Cm^{250} is β-stable, or its β half-life is very long. Hence, no Cf^{250} appears in the mass-250 β-decay chain.
[c] The relative value listed (1.000) corresponds to the total atoms of mass 253 formed. Since the half-lives of Cf^{253} and Es^{253} are comparable, the number of mass-253 atoms existing as Es^{253} at any given time will be less than the value quoted.
[d] Cf^{254} does not decay by β^- emission. Hence, no Es^{254} is formed in the mass-254 β-decay chain.

nuclear mass systematics.[10] The agreement between the measured α-particle energy and systematic predictions can be improved if we assume the most abundant α group of Cf^{253} populates an excited state at approximately 100 keV in Bk^{249} in a manner analogous to the α decay of Fm^{255}, an isotone of Cf^{253}. In this case, the predicted α-particle energy for the predominant α group of Cf^{253} is 5.96 MeV.

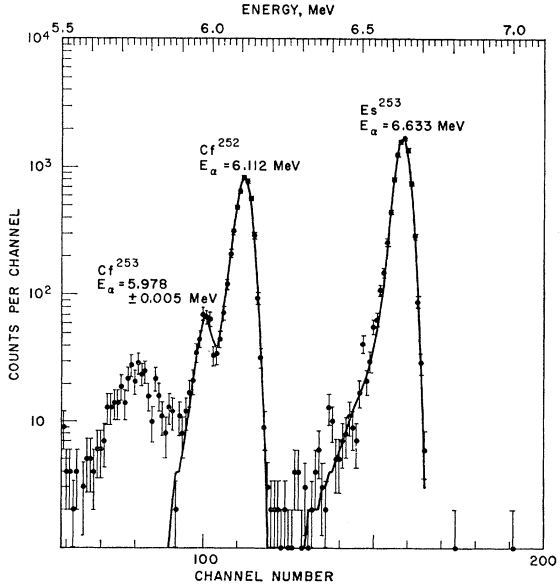

FIG. 2. Alpha spectrum of californium from Par event taken at an early time after the explosion.

[10] V. E. Viola, Jr., and G. T. Seaborg, J. Inorg. Nucl. Chem. 28, 697 (1965); 28, 741 (1966); B. M. Foreman and G. T. Seaborg, *ibid.* 1, 305 (1958).

The α-decay branching of Cf^{253} is $(0.31\pm0.04)\%$, which corresponds to a 15.5-year partial half-life for α-particle emission. A theoretical calculation utilizing the spin-independent equations of Preston[11] would predict an 11-yr half-life for an unhindered group of this energy. The hindrance factor for this α group is 1.4 ± 0.2, which undoubtedly indicates that the state in Cm^{249} populated by this group has the same configuration, i.e., Nilsson quantum numbers,[12] $\Omega\Pi I[Nn_z\Lambda\Sigma]$, as the parent. Cf^{253} has 155 neutrons, as does Fm^{255}, and would be expected to have the same ground-state configuration,[13] $\frac{7}{2}+\frac{7}{2}[613\uparrow]$. This is partially confirmed by the small log ft value, $\sim$6.8, for the β^- decay of Cf^{253} to Es^{253}, which also has a spin of $\frac{7}{2}$. Thus, Cf^{253} would be expected to have about the same α-decay scheme as Fm^{255}.

In the latter case the state receiving the favored α decay decays by emission of $M1$ and $E2$ transitions to a $K=\frac{1}{2}$ rotational band with a 37×10^{-9} sec half-life because of admixtures due to the accidental occurrence of a level of the same spin and parity 0.5 keV away.[13] In α spectra of Fm^{255} taken with 2π grid chambers, the coincident electrons cause the main α group to have a width larger than expected for a single peak. In Es^{254}-α decay, the same states are involved by the lifetime is $\sim200\times10^{-6}$ sec because only $E2$ transitions can occur and because the near degeneracy with a level of the same spin and parity is removed.[14] In Cf^{253}-α decay, the α spectrum was examined in a detailed manner for

[11] M. A. Preston, Phys. Rev. 71, 865 (1947).
[12] S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. 29, No. 16 (1955).
[13] F. Asaro, S. Bjornholm, and I. Perlman, Phys. Rev. 133, B291 (1964).
[14] W. C. McHarris, F. S. Stephens, F. Asaro, and I. Perlman, Phys. Rev. 144, 823 (1966).

evidence of coincident conversion electrons. Utilizing the Es^{253} peak shape and fitting it to the Cf^{252} peak (see Fig. 2), there was no indication in the Cf^{253} peak of excessive width, i.e., coincident conversion electrons. This is reasonable in that, depending upon the excited-state energy, the expected transitions to a $K=\frac{1}{2}$ ground-state rotational band would be $M3$, K-forbidden $E2$, or doubly K-forbidden $M1$ transitions.

As can be seen in Table I, the abundance of Cf^{253} relative to Cf^{252} at the end of irradiation was a factor of 30 higher in Par and Barbel debris than in reactor-produced californium. Thus it can be understood why the α decay of Cf^{253} had not been detected before the Par and Barbel debris were examined. The multiple neutron-capture process in these devices provided the effective isotopic enrichment required to allow detection of the Cf^{253} α-decay branching.

C. The α Decay of Es^{255}

The α-decay branching of Es^{255}, a previously unobserved mode of decay for this nuclide, was measured in samples of einsteinium recovered from the Par and Barbel events. Es^{255} was discovered during the initial studies of the heavy-element fractions for the "Mike" thermonuclear test in November 1952. The β decay of Es^{255} was observed with a half-life reported as 30 days.[8,15,16] In later studies of Es^{255} produced in reactor irradiations, the half-life was reported to be 24 ± 2 days.[17] In a more recent study, the half-life of Es^{255} was found to be 38 ± 3 days.[14]

The spectrum of the Par einstenium is shown in Fig. 3. A purified sample of Es^{255} will show initial growth of 7.03-MeV α particles of 20-hour Fm^{255} and subsequent decay with the half-life of the parent, Es^{255}. In addition to this 7.03-MeV-α group and the 6.63-MeV Es^{253}-α group, an α group appears at 6.300 ± 0.003 MeV. The energy of this new α group was measured with the most abundant α groups of Es^{253} at 6.633 MeV, Cf^{252} at 6.112 MeV, and Cm^{244} at 5.802 MeV used as standards. The half-life of the 6.300-MeV α group is 39 ± 3 days. The half-life of Es^{255} was measured in our samples by following the decay of the Fm^{255} α peak with this result: Es^{255} $t_{1/2}=39.8\pm1.2$ days. A value of the Es^{255} half-life can also be derived from measurements of the Fm^{255}/Es^{253} α-activity ratio as a function of time. The difference in decay constants between Es^{255} and Es^{253} has been measured in this manner in Par and Barbel einsteinium samples and found to be $(0.01584\pm0.0003$ days$)^{-1}$. The half-life of Es^{253} has been reported by Jones *et al.*[17] to be 20.03 ± 0.01 days where the decay was followed over a period of two half-lives. Four samples of Es^{253} from Par and Barbel were followed over periods ranging from 3 to 12 half-lives with the resultant average Es^{253} half-life, 20.7 ± 0.3 days. In spite of the excellent precision quoted by Jones *et al.*,[17] the Es^{253} half-life measured in our work is thought to be at least as accurate as this earlier measurement, especially since our samples were counted over a considerably longer period of time. Thus, combining the accurately measured Es^{255}-Es^{253} decay constant difference with our measured Es^{253} half-life, one derives an Es^{255} half-life of 39.3 ± 1.5 days, while use of the Es^{253} half-life of Jones *et al.*[17] gives an Es^{255} half-life of 37.9 ± 0.8 days. The first of these half-lives confirms the directly measured value, 39.8 ± 1.2 days, which we adopt as the best value.

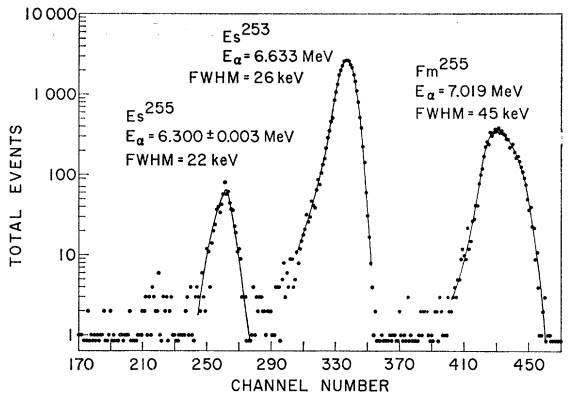

FIG. 3. Alpha spectrum of einsteinium from Par event.

This new α group at 6.300 MeV is assigned to Es^{255} because its observed half-life is consistent with that measured for Es^{255} β decay and because the α particle energy is consistent with a predicted value, 6.23 MeV, for Es^{255} derived from nuclear mass systematics.[10] The most prominent α group of Es^{255} would be expected to populate either the ground state or an excited level close to the ground state of Bk^{251} in a manner similar to the α decay of Es^{253}. Of course, precise agreement is not always obtained between experimentally measured α-decay energies and systematic predictions.

The α-decay branching of Es^{255} is $(8.5\pm0.3)\%$, as derived from the Es^{255}-Fm^{255} α-activity ratio. Thus, the experimental partial half-life for α decay is 470 days, whereas theoretical calculations for an unhindered 6.30-MeV α group in Es^{255} predict a 450-day half-life. The hindrance factor for this group is 1.0 ± 0.1, undoubtedly indicating that the level populated in Bk^{251} by this group has the same configuration as the parent, probably $\frac{7}{2}+$.

The width of the Es^{255}-α peak is comparable to that of Es^{253}, i.e., about 25 keV full width at half maximum in Fig. 3. It should be noted that studies of Es^{253}-α groups made with high-resolution spectrometers have shown the α spectrum to be complex, i.e., 90% of 6.633-MeV α, 6.6% of 6.592-MeV α, and 10 less abundant groups with energies <6.632 MeV.[18] Our experimental

[15] A. Ghiorso *et al.*, Phys. Rev. **99**, 1048 (1955).
[16] G. R. Choppin, S. G. Thompson, A. Ghiorso, and B. G. Harvey, Phys. Rev. **94**, 1080 (1954).
[17] M. Jones *et al.*, Phys. Rev. **102**, 203 (1956).
[18] E. K. Hyde, I. Perlman, and G. T. Seaborg, *The Nuclear Properties of the Heavy Elements* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1964), Vol. II, p. 952.

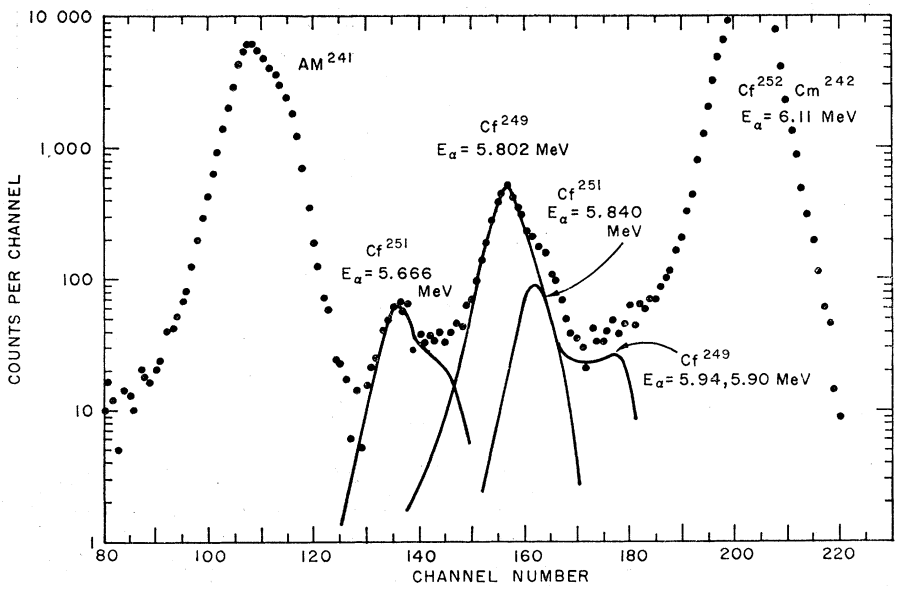

FIG. 4. Alpha spectrum of californium from Par event taken at a later time after the explosion.

measurement of Es^{255} α decay does not allow any conclusions to be reached regarding the complexity of the decay except that the Es^{255} peak does not show any evidence of abundant coincident conversion electrons, as observed in the perturbed shape of the Fm^{255} α peak.

It is reasonable that the Es^{255}-α branching had not been observed before the study of the Par and Barbel debris, because the enrichment of mass 255 relative to other einstenium isotopes in einstenium from long-term reactor irradiations of heavy targets is much too low (see Table I). In the Par einstenium the Es^{255}-Es^{253} ratio, which is a factor of 12 higher than in reactor-produced einstenium, and the absence of 270-day Es^{254} allow the 6.300-MeV Es^{255}-α peak to be detected.

A sample of Par einstenium was counted for spontaneous fission activity. Although Es^{253} is known to have a very long partial spontaneous-fission half-life, 6.3×10^5 y,[9] predictions for the Es^{255} partial spontaneous-fission half-life are as follows: Viola and Wilkins,[19] 92 days; Foreman and Seaborg,[10] 40 yr; Dorn,[20] 230y; and Johansson,[21] 1600 y. From the observed limit on spontaneous-fission events, ≤ 0.0016 fissions/min, in a sample containing 2×10^5 atoms of Es^{255}, a lower limit of 170 y can be set for the partial spontaneous-fission half-life of Es^{255}.

D. The α Decay of Cf^{251}

In a Par sample of californium, two α groups attributable to Cf^{251} were observed at 5.666 ± 0.005 MeV ($\sim$55%) and 5.840 ± 0.005 MeV ($\sim$45%) (see Fig. 4). It was previously known that α groups of this energy belonged to Cf^{251} α decay.[22] In addition, the 5.666-MeV group was known to be in coincidence with a 180-keV γ transition and K x rays. No coincidences had been observed with the 5.840-MeV group. The spectrum obtained in the present work shows a definite high-energy bulge on the 5.666-MeV group, which is consistent with conversion electrons being in coincidence with the α group. No such bulge is observed on the 5.840-MeV group, which again is consistent with the previous information that no coincidences were observed. The state in Cm^{247} populated by the 5.666-MeV α group had been assigned quantum numbers[22] $\frac{1}{2}+\frac{1}{2}[620\uparrow]$. The state populated by the 5.840-MeV α group and presumably by the 180-keV transition from the higher energy level has not been yet assigned.

E. Spontaneous Fission of Cm^{250}

The spontaneous fission of Cm^{250} is of interest because it has been invoked as a possible source of energy for the still perceptible remnants of old supernovae; e.g. Crab nebula, SN1054.[23,24] An earlier determination of the Cm^{250} half-life (approximately 2×10^4 years) employed "Mike" curium whose Cm^{250} content was extrapolated from other curium abundances.[23] The isotopic composition of Par curium, determined with a mass spectrometer, is shown in Table II. In this curium, two-thirds of the spontaneous fission activity was due to Cm^{250}. The number of atoms of Cm^{250} in a sample is easily determined from the α activity of Cm^{242} and the mass ratio of Cm^{242}/Cm^{250}, shown in Table II.

A sample of Par curium was repeatedly separated from californium by α-hydroxyisobutyrate cation

[19] V. E. Viola, Jr., and B. D. Wilkins, Nucl. Phys. (to be published).
[20] D. W. Dorn, Phys. Rev. **121**, 1740 (1961).
[21] S. A. E. Johnsson, Nucl. Phys. **12**, 449 (1959); **22**, 529 (1961).
[22] F. Asaro and I. Perlman (unpublished).
[23] J. R. Huizenga and H. Diamond, Phys. Rev. **107**, 1087 (1957).
[24] P. Morrison and L. Santori, Phys. Rev. Letters **14**, 771 (1965).

columns and saturated HCl-ethanol anion columns. The ratio of spontaneous fission activity to α activity was not changed by the last two separations, thus demonstrating the radiometric purity of the curium.

The final sample gave 0.1563 ± 0.0017 total spontaneous fissions/min and $(4.127\pm0.006)\times10^5$ α disintegrations/min of Cm^{242}. The resultant partial spontaneous-fission half-life for Cm^{250} is $(1.74\pm0.24)\times10^4$ years. Most of the uncertainty comes from the mass-spectrometric data.

F. Upper Limits for Bk^{251} and Cm^{252} Half-Lives

The curium and berkelium fractions isolated from the Par and Barbel debris were examined promptly with a mass spectrometer of high sensitivity for evidence of heavy isotopes. Since all the capture products begin their existence as isotopes of uranium (or perhaps neptunium or protactinium), subsequent β decays may produce a β emitter with a half-life long enough to be detected with a mass spectrometer. Two nuclides which might possibly be expected to have observable half-lives are Cm^{252} and Bk^{251}, whose predicted total β-disintegration energies are 0.78 MeV and 1.44 MeV, respectively.[10] From the absence of these two isotopes in the mass-spectrometric measurements, upper limits of two days for Cm^{252} and three days for Bk^{251} can be set on their half-lives.

G. Fm^{257} and Fm^{258}

The existence of Fm^{257} was confirmed by workers at all four laboratories with the identification of this nuclide in samples from both the Par and Barbel debris. A fermium sample was counted over a period of 270 days in a detector-pulse-height-analyzer system which provided some α-particle energy discrimination but did not allow resolution of Fm^{257} (6.56 MeV) and Es^{253} (6.63-MeV) α groups. The decay data were fitted in a calculation, assuming half-lives of 17.6 days for the Cf^{253} daughter and 20.0 or 20.7 days for the Es^{253} granddaughter. The resultant Fm^{257} half-life was 94 ± 10 days; the choice of half-lives for Es^{253} made a negligible difference (0.7 day) in the Fm^{257} half-live. The best value for the Fm^{257} half-life derived from the reactor-produced sample with which this half-life was first measured is 97 ± 10 days.[25]

The ratio of spontaneous-fission events to α-particle emission in the Par-Barbel Fm^{257} sample was $(2\pm0.7)\times10^{-3}$, which is consistent with the value $(2.1\pm0.4)\times10^{-3}$ originally reported by Hulet *et al.*[26] This fact is significant since, if appreciable amounts of Fm^{258} were present in the sample, the spontaneous-fission rate of Fm^{258} or of Cf^{254} which is the alpha-decay daughter of Fm^{258}, or both, might contribute to give a considerably higher (spontaneous fission)/(α decay) ratio than observed. From Bell's treatment[5] of Par and Barbel mass-abundance data in which he succeeded in fitting the experimental yields with his calculations, one would predict a 258/257 mass ratio of 0.05 at the end of irradiation.[27] From the absence of excess spontaneous-fission events over those attributable to Fm^{257} in the Par fermium fraction, one can assume that Fm^{258} half-life is either too short or too long to allow detection.

TABLE II. Isotopic composition of curium.

Isotope	Relative number of atoms
Cm^{242}	$15.4\ \pm0.8$[a]
Cm^{246}	$27.0\ \pm1.4$
Cm^{248}	$1.95\ \pm0.2$
Cm^{250}	0.155 ± 0.016

[a] At midtime of spontaneous-fission count.

The estimated α-decay half-life of Fm^{258}, based upon well-established systematics,[10] is about 100 days. If the spontaneous-fission half-life of Fm^{258} were very long, the α decay would predominate and determine its measurable half-life. After purification, a sample containing Fm^{258} would show initial growth of the α-decay daughter, Cf^{254}, a 60 day spontaneous-fission activity. If the α half-life of Fm^{258} were 100 to 200 days and the spontaneous-fission half-life considerably longer, we would have observed a fission rate due to Cf^{254} activity seven times that observed in the Par-Fm sample. Wider limits on the estimated Fm^{258} α half-life, 50 to 400 days, would still imply spontaneous fission rates 3 to 5 times higher than that observed. Thus, we conclude that the absence of Fm^{258} in the sample can be attributed to its relatively short half-life for spontaneous fission. The fission rate observed in the Par fermium sample, already attributed to the spontaneous fission of Fm^{257}, corresponds to the expected rate from Fm^{258} in the sample, assuming a Fm^{258} spontaneous-fission half-life of 25 days. Alternative explanations–that Bell's estimated Fm^{258}/Fm^{257} ratio is too high, or that the estimated Fm^{258} α-half-life range of 50 to 400 days is wrong–seem less likely.

In order to lower the limit for the spontaneous-fission half-life for Fm^{258}, the 0.3 α disintegrations/min, or 5×10^4 atoms of Fm^{257}, extracted from 10 kg of Par rock was irradiated in a neutron flux of 1×10^{14} n/cm^2 sec for 4 h. The sample was removed from the reactor and counting was begun within an hour. This search for spontaneous-fission events from the decay of Fm^{258} revealed no evidence of the presence of Fm^{258}. If one assumes a cross section of 100 b for the neutron-capture cross section of Fm^{257}, an upper limit of 2 h can be set on the half-life of Fm^{258}, assuming its predominant mode of decay is spontaneous fission.

An earlier effort to produce and identify Fm^{258} re-

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ported by Sikkeland *et al.*[28] involved the irradiation of 2.5×10^4 atoms of Fm^{257} in a flux of 1.7×10^{14} n/cm^2 sec for 8 days. Within statistical errors, the only spontaneous-fission activity observed in the sample was attributed to Fm^{257}. This result was used to calculate an upper limit of 2 barns for the production of a 10-day spontaneous-fissioning isotope in the neutron irradiation of Fm^{257}, with the implicit assumption that the half-life of Fm^{258} might be 10 days, as reported by Gatti *et al.*[29] More recent efforts[30] with larger amounts of Fm^{257} target and higher neutron fluxes have failed to produce observable amounts of Fm^{258}, with even lower limits being set on the half-life of Fm^{258}.

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Photoneutron Cross Sections of Pr^{141} and I^{127} from Threshold to 33 MeV*

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The photoneutron cross sections for I^{127} and Pr^{141} have been measured with about 170- and 320-keV photon energy resolution, using monoenergetic photons obtained from annihilation in flight of positrons. The (γ,n), $(\gamma,2n)$, and $(\gamma,3n)$ cross sections were determined experimentally by a neutron-counting technique; hence, the nuclear photon-absorption cross sections were obtained without calculated multiplicity corrections. The photon-absorption cross section obtained for Pr^{141} is a single-peaked, giant dipole resonance, well described by a Lorentz-shaped curve with a maximum of 320 mb at 15.2 MeV and a width of 4.5 MeV. The I^{127} giant resonance is broadened slightly by its vibrational deformation. Although a single Lorentz curve is in fair agreement with the data, better agreement is obtained by the sum of two Lorentz curves, when adjusted to fit the data with the restrictions implied by the hydrodynamic model. The parameters of the two curves are: resonance energies, 15.2 and 15.8 MeV; peak cross sections, 180 and 50 mb; and widths, 4.1 and 8 MeV. The integrated nuclear photon-absorption cross sections of I^{127} and Pr^{141}, obtained from the areas under the Lorentz curves which fit the data, are 1.8 ± 0.2 and 2.3 ± 0.2 MeV b, respectively. These integrated cross sections are in good agreement with the dipole sum rule uncorrected for exchange effects, which predicts 1.9 and 2.1 MeV b. The $Pr^{141}(\gamma,3n)$ threshold, which was measured to be 28 ± 1 MeV, determines the mass of Pr^{138}.

INTRODUCTION

EXPERIMENTAL results for the magnitude and energy dependence of photoneutron cross sections of medium-heavy nuclei can be easily compared with theoretical predictions based on nuclear models. The interpretation is not confused by uncertainty in the physical description of the incoming projectile which occurs in particle-induced reactions, or by Coulomb effects present when charged particles are emitted. The energy dependence of the cross section for photon absorption is predicted by the hydrodynamic model of Steinwedel and Jensen[1] and by extended hydrodynamic models[2,3] which include deformation and vibration-rotation effects. The shell model has been applied to photon absorption in medium-heavy nuclei,[4] but exact calculations accounting for particle-hole effects[5] are very difficult. Various moments of the photon absorption cross section have been calculated by using sum rules,[6,7] for which no model is required to describe the excited states reached by photon absorption. The effect of exchange forces between neutrons and protons has been included in these sum rule calculations.[8] Statistical theories of nuclear level densities[9] can be used to predict the ratio of the partial cross sections,[10] (γ,n), $(\gamma,2n)$, $(\gamma,3n)$, (γ,p) etc. These predictions must be modified if "direct" or "fast" interactions occur. There exists a wealth of theoretical predictions about photoneutron reactions; however, comparison of experiments with theories has been hindered by a lack of consistency in

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