TITLE: NEUTRON MULTIPLICITY MEASUREMENTS OF Cf
AND Fm ISOTOPES

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Prompt neutrons in coincidence with the two fission fragments from the SF of californium 250, 252, and 254, and fermium 256 and 257 were measured with a 75-cm-diameter, Gd-loaded liquid scintillation counter having a neutron-detection efficiency of about 80%. The kinetic energies of both fission fragments and the number of neutrons for each fission event were recorded. From these data, the fragment masses and kinetic energies, the average number of neutrons per fission, and the neutron multiplicity distributions and their variances as a function of total fragment kinetic energy and the ratio of fragment masses have been calculated and will be presented.

*Work performed under the auspices of the US DOE

INTRODUCTION

We first became interested in measuring the number of prompt neutrons emitted in spontaneous fission (SF) in connection with our early studies¹ of the mass distributions from SF of ^{257}Fm . A knowledge of prompt neutron emission as a function of fragment mass, charge, and kinetic energy is necessary in order to obtain pre-neutron-emission mass-yield distributions from radiochemical or kinetic-energy measurements of the fission products. Figure 1 illustrates the difference in appearance of the mass-yield curve for ^{257}Fm depending on the neutron-emission function used in the calculation of pre-neutron-emission masses from the kinetic-energy data. Information concerning the excitation or deformation energies of the fragments can also be deduced from studies of prompt neutron emission. In the case of the fermium isotopes, these quantities have turned out to be of particular interest because of the trend with increasing mass toward symmetric mass division, accompanied by very high total kinetic energy releases and reduced neutron emission. The average neutron emission per fission event, $\bar{\nu}_T$, for low energy fission of actinides generally increases with Z and A , except for the fermium isotopes where $\bar{\nu}_T$ decreases with increasing A . We have attributed these phenomena to the approach of the symmetric fragments to the doubly magic ^{132}Sn configuration--thus the fragments might be expected to be nearly spherical which results in the maximum kinetic energy due to Coulomb repulsion, a minimum of excitation energy, and fewer neutrons emitted from the fragments. Since our first measurements of neutron emission for ^{257}Fm , we have tried to measure neutrons emitted from fission fragments of other nuclides which spontaneously fission in order to see if there are indeed the expected differences in neutron emission as a function of total kinetic energy, mass split, and Z and A of the fissioning nucleus.

EXPERIMENTAL

In the current studies we have used a 75-cm diam. cylindrical tank containing Nuclear Enterprise NE-323 liquid scintillator loaded with 0.5 weight percent gadolinium. The tank has a 15.2-cm diam. cylindrical hole in the center in which an evacuated chamber containing a pair of silicon surface-barrier detectors and the SF sources on $40\text{-}\mu\text{g}/\text{cm}^2$ carbon or VVNS films is placed. In this arrangement the neutrons emitted from the SF of ^{250}Cf , ^{252}Cf , ^{254}Cf , and ^{257}Fm have been measured as a function of mass split and kinetic energy of the fragments with a neutron detection efficiency of 75 to 80%. In the case of the measurements for ^{256}Fm , the fission detectors and source were placed just outside the tank and the resultant neutron detection efficiency was 31%. The detection of coincident fission fragments was used to trigger a 40- μsec gate after a 2- μsec delay to allow prompt gamma rays from fission and proton recoils from thermalization of the neutrons to decay. Details of construction and operation of the tank have been given in a previous report².

The sources of ^{250}Cf , ^{252}Cf , and ^{257}Fm were produced by successive neutron captures in targets irradiated in the High Flux Isotope Reactor (HFIR), processed at the Oak Ridge National Laboratory, and made available to us under the Transplutonium Production Program of the Division of Research of the U. S. Department of Energy. The ^{257}Fm was further purified from rare earths and other actinides by elution from ethanol-HCl and hot alpha-isobutyroxyisobutyrate columns. Only relatively small samples of ^{257}Fm are available and the measurements were performed on sources of only a few spontaneous fissions per minute. The ^{254}Cf ($T_{1/2} = 60$ days) and ^{256}Fm ($T_{1/2} = 2.6$ hours) sources were prepared by collection on carbon foils of products recoiling from triton irradiations of ^{254}Cf and ^{254}Es . The ^{254}Cf was self-transferred during irradiation of a ^{254}Cf target and the ^{256}Fm was the result of beta decay of ^{256}Es prepared by the (t,p) reaction on ^{254}Es . These sources were therefore essentially weightless.

RESULTS

The frequency distributions, $P_t(v)$, for ^{250}Cf , ^{252}Cf , ^{254}Cf , and ^{257}Fm are shown in Figs. 3-6. These "unfolded" distributions were obtained by a matrix inversion of the equation shown in Fig. 2. In the case of ^{256}Fm where the efficiency was only 31%, the distribution was not unfolded, but the variance, $\sigma_{v_t}^2 = \epsilon^{-2}\sigma_d^2 + \epsilon^{-1}(1-\epsilon^{-1})(\bar{v}_d)$ and $r_2 = (\sigma_v^2 - \bar{v})^{-1}(\bar{v}^{-2}) + 1$, where the subscripts t and d refer to true and observed, respectively. The data for these nuclides are summarized in Table I. The variance for ^{257}Fm shows a considerable increase over the values for the lighter nuclides. Our value of 1.82 ± 0.08 is considerably smaller than that of 2.30 ± 0.65 reported³ previously although it is within the quoted error for the earlier measurement. The larger variance for ^{257}Fm reflects the increased spread in neutron emission, particularly for symmetric mass division where for the lowest total kinetic energy (TKE) events, $\bar{v}$ is 4.86, but only 1.08 for the highest TKE events.

Figures 7 and 8 show contour plots of TKE as a function of mass fraction for ^{254}Cf , ^{256}Fm , and ^{257}Fm , and illustrate the differences in the TKE release and its variance for symmetric mass division. ^{257}Fm exhibits a very large spread in TKE at symmetry and these differences in TKE seem to correlate nicely with the spread in neutron emission, i.e., the very high kinetic energy events are accompanied by the emission of fewer neutrons. Figures 9-12 show $\bar{v}$ as a function of mass split and TKE for the nuclides studied, and illustrate this effect. In particular, for ^{257}Fm , the highest TKE events, observed for symmetric and near symmetric mass splits, show $\bar{v}$'s of only 1.08 and 0.80, while for ^{254}Cf the lowest value of $\bar{v}$ is 2.35 for symmetric mass division. The marginal distributions in Fig. 14 show the changes in $\bar{v}$ and σ_v^2 for all masses as a function of TKE. The highest TKE events in ^{257}Fm for all mass splits have a $\bar{v}$ of only 1.07 while the lowest value for the Cf isotopes is

.1.69 for ^{250}Cf .

If one looks at the events from SF of ^{257}Fm with $\text{TKE} > 235 \text{ MeV}$, where $\bar{\nu} = 1.1$, the symmetric mass distribution shown in Fig. 15 is obtained. This resembles the mass distributions we have measured for SF of ^{258}Fm and ^{259}Fm where the most probable TKE's are 238 and 243 MeV, and would appear to indicate the same mechanism and that neutron emission is similarly low. The large variances observed for the neutron multiplicity distributions for ^{256}Fm and ^{257}Fm may perhaps be attributed to the fact that they are in a "transition" region, i. e., although symmetric fragments can have the magic proton number of 50, they are still 3 to 4 neutrons from the 82-neutron closed shell. This may make these fragments rather "soft" to deformation and the observation of symmetric fragments with both very high and very low TKE suggests a rather large difference in their shapes, ranging from elongated to nearly spherical. The number of neutrons emitted per fission and the variance of the multiplicity distributions should decrease very sharply for ^{258}Fm and ^{259}Fm and higher mass fermium isotopes as symmetric mass division results in fragments which more closely approach the rigid doubly magic configuration. If this hypothesis is correct, then neutron emission might be expected to increase again for fissioning systems with A greater than 100. It would be interesting to check this with experimental measurements and to determine whether the proton or neutron shell influence is more important.

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TABLE I.

VARIANCE OF NEUTRONS EMITTED IN SF
OF Cf AND Fm ISOTOPES

<u>Nuclide</u>	<u>$\bar{\nu}_T$</u>	<u>$\sigma^2_{\nu_T}$</u>	<u>r_2</u>	<u>Peak-to-Valley Ratio</u>
$^{246}\text{Cf}^a$	3.14 ± 0.09	1.66 ± 0.31	0.850	-----
^{250}Cf	3.50 ± 0.09^b	1.51 ± 0.02^c	0.838	-----
^{252}Cf	3.735 ± 0.014	1.55 ± 0.02^c	0.843	750^d
^{254}Cf	3.89 ± 0.05^b	1.56 ± 0.02^c	0.846	$\geq 145^e$
$^{254}\text{Fm}^a$	3.96 ± 0.14	1.50 ± 0.20	0.843	$\approx 60^f$
^{256}Fm	3.70 ± 0.18	1.82 ± 0.08^c	0.863	12^f
	3.73 ± 0.18^a	2.30 ± 0.65^a	0.897	
$^{257}\text{Fm}^g$	3.77 ± 0.02	2.51 ± 0.02^c	0.911	$\approx 1.5^g$

^a Yu. A. Lazarev, At. Energy Rev. 15, 75 (1977).

^b Values from C. J. Orth, Nucl. Sci. Eng. 43, 54 (1971) adjusted to $\bar{\nu}_T = 3.735$ for ^{252}Cf .

^c This work.

^d W.E. Nervik, Phys. Rev. 119, 1685 (1960).

^e J.E. Gindler et al. Phys. Rev. C 16, 1483 (1977).

^f J. P. Unik et al., Proc. IAEA Symp. Phys. Chem. Fission III, Rochester, NY, 1973, Vol. II, 19 (1974).

^g J. P. Balagna et al., Phys. Rev. Letters 26, 145 (1971).

FIGURE CAPTIONS

1. Pre-neutron-emission mass-yield curve for ^{257}Fm from Ref. 1. The two sets of points correspond to different assumptions concerning ν (M).
2. Formula for calculation of $P_t(\nu)$.
3. Neutron multiplicity distribution for ^{250}Cf .
4. Neutron multiplicity distribution for ^{252}Cf .
5. Neutron multiplicity distribution for ^{254}Cf .
6. Neutron multiplicity distribution for ^{257}Fm .
7. Contour diagram showing pre-neutron-emission TKE distributions for ^{254}Cf and ^{256}Fm .
8. Contour diagram showing pre-neutron-emission TKE distributions for ^{257}Fm from Ref. 1. The contours are lines of relative numbers of events, based on data groupings 5 Mev x 0.01 units of mass fraction.
9. $P_t(\nu)$ for ^{250}Cf as a function of TKE and mass split.
10. $P_t(\nu)$ for ^{252}Cf as a function of TKE and mass split.
11. $P_t(\nu)$ for ^{254}Cf as a function of TKE and mass split.
12. $P_t(\nu)$ for ^{257}Fm as a function of TKE and mass split.
13. Table of $\bar{\nu}$, σ_t^2 , and Γ_2 as a function of TKE AND mass split for ^{256}Fm .
14. Marginal distributions for ^{250}Cf , ^{252}Cf , ^{254}Cf , and ^{257}Fm .
15. Mass distribution for ^{257}Fm for fission events with TKE >235 MeV.

Figure 1

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$$P_d(n) = \sum_{k=0}^n \sum_{v=n-k}^{v_{\max}} \binom{v}{n-k} \epsilon^{n-k} (1-\epsilon)^{v-n-k} P_b(k) P_t(v)$$

ϵ = EFFICIENCY OF NEUTRON DETECTION.

$P_d(n)$ = PROBABILITY THAT n EVENTS ARE DETECTED.

$P_t(v)$ = PROBABILITY THAT v NEUTRONS ARE EMITTED IN A FISSION.

$P_b(k)$ = PROBABILITY OF MEASURING k BACKGROUND COUNTS. FOR

OUR SYSTEM, $P_b(k) = \frac{e^{-a} a^k}{k!}$, THE POISSON DISTRIBUTION.

a = THE AVERAGE NUMBER OF DETECTED BACKGROUND EVENTS.

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 11

Figure 12

^{256}Fm (SF = 46943)

$$\bar{\nu}_T = 3.70 \quad \sigma_{\nu_T} = 1.82 \pm 0.08$$

ALL ENERGIES	16362 3.60 2.65	21899 3.73 1.31	7671 3.71 1.31	965 3.92 2.40	ALL MASSES
240	2260 1.98 1.15	451 1.51 0.96			2711 1.90 1.15
220	8047 3.29 1.67	7088 2.80 0.56	629 2.01 0.51		15764 3.02 1.22
200	5214 4.53 2.18	11944 4.05 0.49	4617 3.43 0.88	337 2.95 0.69	22112 4.01 1.12
180	SF = 760 $\bar{\nu} = 5.69$ $\sigma_{\nu}^2 = 0.92$	2414 5.30 1.26	2424 4.69 0.20	626 4.45 2.59	6224 5.01 1.19
160					
	128.0	138.2	148.5	158.7	169.0
	MASS				

Figure 13

Figure 14

Figure 15