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PROMPT NEUTRONS FROM THE SPONTANEOUS FISSION OF FERMIUM-257[*]

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ABSTRACT

Prompt neutrons emitted following the spontaneous fission of ^{257}Fm were measured using a 75-cm diam. Gd loaded liquid scintillator (neutron detection efficiency = 66.5%). A chamber containing ^{257}Fm and ^{252}Cf sources, each facing a Si-surface-barrier detector, was placed in the center of the tank. The average number of neutrons emitted for ^{257}Fm spontaneous fission was found to be 3.77 ± 0.02 based on a value of $\bar{\nu}$ = 3.735 ± 0.014 for ^{252}Cf. The variance of the neutron multiplicity distribution is 2.49 ± 0.06 for ^{257}Fm compared with 1.56 ± 0.02 for ^{252}Cf. The variation of $\bar{\nu}$ as a function of single fragment kinetic energy was investigated. For ^{257}Fm, $\bar{\nu}$ is ≈ 1 for the highest 1% of the single fragment kinetic energies while for ^{252}Cf the value is 3. The high kinetic energy fragments from the fission of ^{257}Fm are principally from symmetric mass splits while in ^{252}Cf they are from asymmetric divisions. Low neutron emission from symmetric division in ^{257}Fm is consistent with the prediction of spherically stabilized fragments near the scission point due to the proximity of the ^{132}Sn doubly magic core.

Introduction.

Previous studies of the kinetic energies of ^{257}Fm spontaneous fission products indicated for the first time that a low energy fission process can have a high yield for symmetric mass division [1]. Subsequent studies of fragment kinetic energy measurements on ^{255}Fm(n,f) [2] ^{257}Fm(SF) [3] and ^{257}Fm(n,f) [3], as well as radiochemical mass yield determinations for ^{255}Fm(SF) [4] and ^{255}Fm(n,f) [5] have substantiated that the low energy fission of these heavier Fm isotopes results in a higher yield of symmetric fission than has been observed in the low energy fission of other nuclei. These results have stimulated renewed interest in the problems associated with mass distributions in fission [6-8]. The accurate determination of the mass yields for symmetric division, however, requires knowing the number of neutrons evaporated as a function of fragment mass. Figure 1 (from ref. [1]) emphasizes this by showing two mass yield distributions calculated from the observed fragment kinetic energies following the spontaneous fission of ^{257}Fm; the first distribution is calculated with the assumption that the average number of neutrons emitted per fragment has a constant value of 2, while the second assumes the average number of neutrons emitted per fragment has a mass dependence the same as that observed in the spontaneous fission of ^{252}Cf. It is clear that the yield calculated for symmetric fission is dependent on the assumptions concerning the neutron multiplicities.

To elucidate this problem in the calculation of the symmetric fission yields we have measured the neutron multiplicities associated with the fission of ^{257}Fm.

Experimental.

The ^{257}Fm was produced by successive neutron capture in Cm targets irradiated in the high-flux isotope reactor (HFIR) and processed at the transuranium processing facility (TRU) at Oak Ridge National Laboratory. The ^{257}Fm was further purified at Los Alamos by standard cation column procedures involving elution with ethanol-HCl and hot α-hydroxy-isobutyrate to separate the Fm from other actinides. The principle contaminant before separation was ≈ 10^6 dis/min of ^{253}Es. A solution of the purified source was evaporated on a 5-mil Pt disk. This source was positioned ≈ 3 mm from a 1-cm diam. by 30-μm deep Si-surface-barrier detector. A ^{252}Cf source, prepared in a similar manner, was placed in the same position relative to a second surface barrier fission detector. The two sources, back-to-back, and their detectors were placed in a vacuum container in the center of a 75-cm diam. spherical Gd-loaded liquid scintillator tank having a 15-cm diam. cylindrical hole through the center.

The tank was divided optically into quadrants with each quadrant having two RCA-4522 photomultiplier tubes mounted on glass windows. The tank was filled with Nuclear Enterprise NE-323 liquid scintillator loaded with 0.5 wt.% gadolinium. The neutrons emitted in fission thermalized in the liquid and were captured by the gadolinium. The cascade gamma rays following neutron capture were detected by the photomultipliers after interaction with the scintillator liquid. The mean lifetime for the thermalization and capture of neutrons in the tank is about 10 μsec.

Detection in the solid state detectors of a fission fragment from either ^{252}Cf or ^{257}Fm opened a gate for 40 μsec following a 2 μsec delay to avoid counting the prompt fission gamma rays and the proton recoils resulting from the thermalization of the neutrons. During the open gate period the sums of the dynode signals from the four photomultipliers on each half of the tank were fed into a dual discriminator. The discriminator levels determined the efficiency of the tank, and their 150-nsec output pulse lengths determined the dead time. A pulser was used to open the 40-μsec gate every 100 sec so that background counting rates of the system could be measured.

An on-line PDP-8/L computer recorded the outputs of fast scalers, which gave the neutron multiplicities, and of analog-to-digital converters (ADC) which measured the linearly amplified signals of the fission fragment solid state detectors.

The apparatus was placed in a low background room having thick concrete walls. During the experiment the average background rate was 0.1243 ± 0.0010 based on more than 125000 background gates. No more than three pulses were measured in any of the background gates and the frequency distribution of the various multiplicity events was consistent with a Poisson distribution. By having the ^{252}Cf source present in the tank it was possible to monitor continuously the efficiency of the system. The overall neutron detection efficiency was determined to be (66.49 ± 0.28)%, based on an average neutron multiplicity value of 3.735 ± 0.014 for the fission of ^{252}Cf [9].

Results.

Experimental data were collected for more than five months. During that time 10532 ^{257}Fm and 98659 ^{252}Cf fission events were detected. The ^{257}Fm fission detection rate varied from 5.5 SF/h at the start of the experiment to 1.8 SF/h at the conclusion. From a least squares fit to the observed decay rate we obtained a value of 99.3 ± 1.6 d for the half life of ^{257}Fm, in good agreement with the recently published value of 100.5 ± 0.2 d [10]. Figure 2 presents the probability distribution of the observed multiplicity of neutrons from the fission of ^{257}Fm and ^{252}Cf. The probability of observing n events is

$$P_d(n) = \sum_{k=0}^{n} \sum_{\nu=n-k}^{\nu_{max}} \binom{\nu}{n-k} \epsilon^{n-k} (1-\epsilon)^{\nu-n-k} P_b(k) P_t(\nu) \qquad [\text{Eq. 1}]$$

where ϵ = efficiency for neutron detection

$P_t(\nu)$ = probability that ν neutrons are emitted in a fission event, and

$P_b(k)$ = probability of measuring K background counts.

Through a matrix inversion of Eq. 1 it is possible to determine the quantity $P_t(\nu)$, the multiplicity distribution of neutrons emitted in fission. Table I presents the observed and "unfolded" distributions for both ^{257}Fm and ^{252}Cf. The $\bar\nu$ value for ^{257}Fm is found to be 3.769 ± 0.021 which is less than 1% higher than the value of 3.735 ± 0.014 [9] for ^{252}Cf. The 2.49 ± 0.06 variance for ^{257}Fm is much larger than the value of 1.57 ± 0.02 measured for ^{252}Cf. In Table II we compare our current values of these quantities with those reported for ^{257}Fm by Cheifetz et al. [11] and with various reported results on ^{252}Cf [11-14].

A plot of our "unfolded" $P_t(\nu)$ distributions is presented in Fig. 3. The ^{252}Cf distribution is seen to be very nearly Gaussian. Although the ^{257}Fm results show more scatter, they are also represented reasonably well by a Gaussian function.

A notable feature in the fission of ^{257}Fm compared to ^{252}Cf is the higher probability of emission of 0 or 1 neutron. In fact, the whole neutron multiplicity distribution is much flatter for ^{257}Fm than for ^{252}Cf, as evidenced in their relative variances. This implies a broad distribution in excitation energies for ^{257}Fm fission products.

In this experiment only one fission fragment energy was recorded per fission. We have analyzed the variation of $\bar\nu$ and σ_ν^2 as a function of single fragment energy. Figure 4 presents a histogram of the values of $\bar\nu$ and σ_ν^2 for bins containing ≈10% increments of the single fragment kinetic energies. The variances for both ^{257}Fm and ^{252}Cf are seen to remain reasonably constant over the kinetic energy range. The $\bar\nu$ data for ^{257}Fm, however, show a dramatic decrease as the highest kinetic energy bins are selected while no such strong

effect is observed in the ^{252}Cf data. These results are presented in a cumulative manner in Fig. 5. We see that when all of the data are included (the 100% point on the far right of the graph) the ^{257}Fm and ^{252}Cf have, within 1%, the same value of $\bar{\nu}$, but when only the higher kinetic energy intervals are selected the ^{257}Fm $\bar{\nu}$ results are substantially below the ^{252}Cf data. At the highest kinetic energy interval the ^{257}Fm multiplicity has decreased to less than 1 neutron per fission, whereas the ^{252}Cf data have asymtotically approached about 3 neutrons per fission. The low value of $\bar{\nu}$ for the high kinetic energy ^{257}Fm fission implies a very low fragment excitation energy for these events.

With only one fragment energy measured, definitive mass assignments are not possible. For ^{257}Fm fission, however, some selection of the mass distribution can be made by selection of single fragment kinetic energies. Figure 6 shows a mass distribution obtained by restricting one fragment to be within the top 5.3% of the kinetic energy range. These data, which were taken from the ^{257}Fm two-fragment kinetic energy mass determination measurements of ref [1], show a preference for symmetric division when one fragment has a high kinetic energy. Though not conclusive, the combination of the current neutron measurements with the previous mass determinations implies that the symmetric fission of ^{257}Fm results in fragments having very little internal excitation energy.

Discussion and Conclusions.

We have repeated the ^{257}Fm neutron multiplicity measurements done by Cheifetz et al. [11] using a more efficient detection system (66.5% vs 51.5%). Our results, with smaller statistical uncertainties, are consistent with their values ($\pm 2\sigma$). With our higher efficiency we have been able to "unfold" the observed multiplicity distribution and obtain the true neutron distribution. Perhaps the most interesting observation is the strong decrease in $\bar{\nu}$ with increasing single fragment kinetic energy. These events are correlated with symmetric fission of ^{257}Fm. If we relate the neutron multiplicity to the post scission fragment excitation energy we have an estimate for the excitation energy for symmetric division. We present these data in Fig. 7, a graph containing the predictions of Schmitt and Mosel [7] for fragment excitation energy for symmetric [$Z_{f_1} = Z_{f_2} = Z/2$, $A/2$] and asymmetric [$Z_{f_1} = 50$, $A_{f_1} = 132$; $Z_{f_2} = Z-50$, $A_{f_2} = A-132$] division. Since the $\bar{\nu}$ value for any specific division is unknown, we can use only the measured $\bar{\nu}$ values for given mass or charge. For the fission of ^{252}Cf the neutron multiplicities have been determined both as a function of fragment mass [15] and charge [16] and both results are presented in Fig. 7. For asymmetric fission the mass determination method (i.e. $A_{f_1} = 132$, $A_{f_2} = 120$) gives a substantially higher $\bar{\nu}$ than does the charge determination (i.e. $Z_{f_1} = 48$, $Z_{f_2} = 50$) method. For the symmetric fission the two ^{252}Cf methods give essentially the same value. For ^{257}Fm the symmetric mass division (as estimated from Fig. 5 for the highest kinetic energy events) has a $\bar{\nu}$ of ≈ 1. These results are in at least qualitative agreement with the Schmitt and Mosel predictions. The ^{257}Fm symmetric fission is expected to result in two fragments which are spherically stabilized by the proximity of the ^{132}Sn doubly magic core. Such fragments should be stiff toward distortion and have nearly spherical scission configurations. These circumstances should result in high kinetic energy release (due to large coulomb energy from the compact spherical fragments) and low internal fragment excitation energy.

More definitive experiments in which neutron multiplicities are measured in coincidence with both fragment kinetic energies would improve our understanding of the energy division in the spontaneous fission of ^{257}Fm. We are

currently attempting such an experiment with a slightly stronger source (≈ 4 SF/min).

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TABLE I. EXPERIMENTALLY DETERMINED NEUTRON PROBABILITY DISTRIBUTIONS FOR THE FISSION OF ^{257}Fm AND ^{252}Cf.

| n or ν | ^{257}Fm (10532 f) | | ^{252}Cf (98571 f) | |
	Observed $P_d(n)$	"Unfolded" $P_t(\nu)$	Observed $P_d(n)$	"Unfolded" $P_t(\nu)$
0	0.070 ± 0.003	0.022 ± 0.004	0.041 ± 0.001	0.003 ± 0.001
1	0.176 ± 0.004	0.078 ± 0.014	0.175 ± 0.001	0.021 ± 0.005
2	0.260 ± 0.005	0.077 ± 0.032	0.308 ± 0.002	0.140 ± 0.011
3	0.261 ± 0.005	0.259 ± 0.054	0.276 ± 0.002	0.264 ± 0.017
4	0.154 ± 0.004	0.211 ± 0.068	0.146 ± 0.001	0.307 ± 0.020
5	0.061 ± 0.003	0.259 ± 0.068	0.045 ± 0.001	0.191 ± 0.018
6	0.015 ± 0.002	0.039 ± 0.058	0.008 ± 0.0004	0.061 ± 0.013
7	0.003 ± 0.001	0.058 ± 0.032	0.001 ± 0.0002	0.008 ± 0.007
8	0.0001 ± 0.0002	-0.005 ± 0.015	0.0001 ± 0.00006	0.005 ± 0.003
9	0.0001 ± 0.0001	0.002 ± 0.004	0.00001 ± 0.00001	-0.0006 ± 0.0005
$\bar{n}(\bar{\nu})$	2.636 ± 0.014	3.769 ± 0.021	2.607 ± 0.004	(3.735 ± 0.014)
σ^2	2.068 ± 0.027	2.49 ± 0.06	1.651 ± 0.007	1.57 ± 0.02

TABLE II.   SUMMARY OF NEUTRON EVAPORATION DATA
FOR ^{257}Fm AND ^{252}Cf.

^{257}Fm

	No. of fissions	$\bar{\nu}_T$	$\sigma^2_{\bar{\nu}_T}$
Cheifetz et al. [1]	1499	3.97 ± 0.13	2.92 ± 1.40
This experiment	10532	3.77 ± 0.02	2.49 ± 0.06

^{252}Cf

ref.	[11]	[12]	[13]	[14]	this exp.
$\sigma^2_{\nu_T}$	1.87 ± 0.08	1.55 ± 0.04	1.46 ± 0.14	1.46 ± 0.14	1.57 ± 0.02

FIGURE CAPTIONS

Figure 1. Pre-neutron emission mass yield distributions for ^{257}Fm(SF). The two sets of points correspond to different assumptions on the variation of $\bar{\nu}(m)$ (from ref [1]).

Figure 2. The experimentally observed distributions, $P_d(n)$, of events in the fission of ^{252}Cf and ^{257}Fm.

Figure 3. The "unfolded" frequency distribution, $P_t(\nu)$, of neutrons associated with the spontaneous fission of ^{257}Fm and ^{252}Cf. The solid curves represent Gaussian functions having the same first and second moments as the "unfolded" distributions.

Figure 4. The average $(\bar{\nu})$ and variance (σ_ν^2) of the neutron distributions for the fission of ^{257}Fm and ^{252}Cf as a function of single fragment kinetic energy. The data is presented in $\approx 10\%$ bins corresponding to the lowest kinetic energy fraction on the left to the highest on the right.

Figure 5. The variation of the average number of neutrons as a function of the cumulative fraction of single fragment kinetic energy. The value of the ordinate for abscissa value χ is the average number of neutrons for those events with the top χ percent of single fragment kinetic energies.

Figure 6. Mass distribution for ^{257}Fm(SF) obtained when one fragment is restricted to kinetic energy greater than 117 MeV (the top 5.3% of the single fragment kinetic energy distribution).

Figure 7. The solid curves represent calculated excitation energies (E_X) and neutron emission multiplicities (ν) for asymmetric (A) and symmetric (S) fission of nuclei from $Z = 92$ to 114 (from Schmitt and Mosel [7]). The points correspond to experimental values for the fission of ^{252}Cf and ^{257}Fm. See text for additional details.
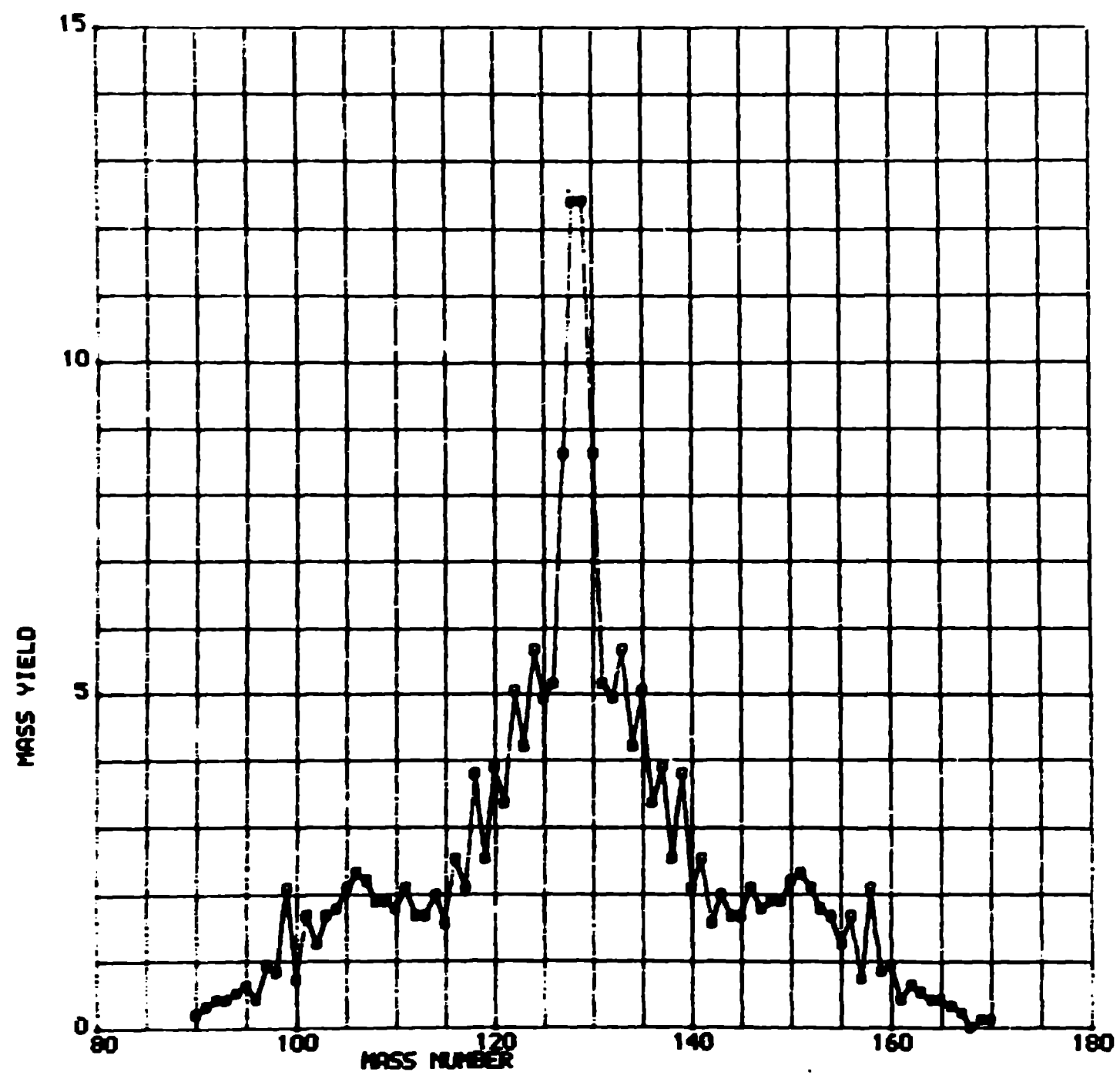
257FM-6 SINGLE FRAGMENT KE GREATER THAN 117 MEV(5.3 PERCENT)

MASS YIELD