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LOW-ENERGY FISSION SYSTEMATICS OF THE FERMIUM ISOTOPES: THE TRANSITION FROM MASS ASYMMETRY AT FERMIUM-254 TO SYMMETRY AT FERMIUM-259*

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Abstract

Recent measurements have shown that ^{259}Fm gives the highest total kinetic energy release and the most symmetric mass division so far observed for spontaneous fission. These results continue the trends observed previously in the fermium isotopes toward higher total kinetic energies and more symmetric mass division with increasing mass of the fermium isotopes. The transition from asymmetric mass division (^{254}Fm) to highly favored symmetric mass division (^{259}Fm) now appears to have been completed. These features are consistent with the simple postulate that the more neutron-rich fermium isotopes show an increase in the yield of symmetric fragments and in the total kinetic energy because symmetric mass division of fermium (Z = 100) nuclei results in two fragments which have the magic proton number of 50 and are close to the magic neutron number of 82. The proximity of the fragments to the doubly magic configuration seems to have a profound effect on the mass division and total kinetic energy release in fission.

1. Introduction

Until about six years ago all the mass distributions measured for low-energy fission, i.e., spontaneous fission (SF) and thermal neutron-induced fission, had been found to be strongly asymmetric[1], and it was commonly believed that such low-energy fission processes always resulted in mass distributions which were highly asymmetric. Even though the SF of ^{254}Fm had also been found[2] to result in asymmetric mass division, some of us continued to speculate that the more neutron-rich fermium isotopes might show an increased yield of symmetric fragments as their configurations more closely approached the stable, doubly-magic ^{132}Sn core. Two doubly magic fragments would, of course, result from symmetric mass division of ^{264}Fm. In 1970, two-parameter kinetic-energy measurements[3] of ^{257}Fm showed substantially increased yields at symmetry and a higher average total kinetic energy (TKE) than would have been expected based on extrapolation from values for lower Z actinides. The TKE as a function of mass fraction increased monotonically with approach to mass symmetry in contrast to the decrease at symmetry observed for lower Z actinides.

Thermal neutron-induced fission of ^{257}Fm showed[4] a still more symmetric but very broad mass distribution, the most probable mass split being symmetric. These observations stimulated considerable interest and since then kinetic-energy and radiochemical measurements have been made for a number of fermium isotopes[5-11]. The results have indicated clear trends toward more symmetric mass distributions, higher TKE's, and decreased neutron emission with increasing mass of the fermium isotopes. The recent measurement[12] of ^{259}Fm showing a narrow peak at symmetry in the mass distribution and a very high TKE are consistent with these trends.

2. Mass Distributions

The mass distributions for the SF of ^{254}Fm, ^{256}Fm, ^{257}Fm and thermal neutron-induced fission of ^{257}Fm (^{258}Fm*) are plotted in Figure 1 and show the increasing yields for symmetric mass splits as a function of increasing mass of the fissioning fermium isotope. Differences in mass distributions between SF and thermal neutron-induced fission caused by the excitation energy of about 6 MeV introduced by the neutron-binding energy can be seen by comparing the mass distribution[10] for SF of ^{256}Fm with that for ^{256}Fm*. The extra excitation energy results in increased yields at symmetry as illustrated in Figure 2. In general, the effect of additional excitation energy on fission is to decrease the influence of shell structure. At sufficiently high energies, ≳ 50 MeV, the most probable mass split becomes symmetric, but the mass distributions are rather broad. Thus the higher yield of symmetric mass division observed for ^{258}Fm* relative to ^{257}Fm (SF) might at least partially be attributed to the effect of the increased excitation energy. However, the recent measurement[12] of the mass distribution for ^{259}Fm (SF) showing a peak at symmetry with a FWHM of only 13 mass units seems to have confirmed the trend toward mass symmetry in the heavier fermium isotopes. In fact, the very narrow symmetric mass distribution for ^{259}Fm appears to have completed the transition to mass symmetry in the fermium isotopes. The mass distributions for ^{254}Fm, ^{257}Fm and ^{259}Fm are plotted in Figure 3 and illustrate this rather dramatic change. The narrow width of the ^{259}Fm symmetric mass distribution can perhaps be explained qualitatively on the basis of the proximity of the fragments from symmetric mass division to the stable, doubly magic $^{132}_{50}Sn_{82}$ configuration which makes symmetric, or nearly symmetric, mass division highly favored. Even if one fragment has the very stable ^{132}Sn configuration, the other would be ^{127}Sn, very close to the most symmetric mass division possible which results in ^{129}Sn and ^{130}Sn. The width of the mass distribution would

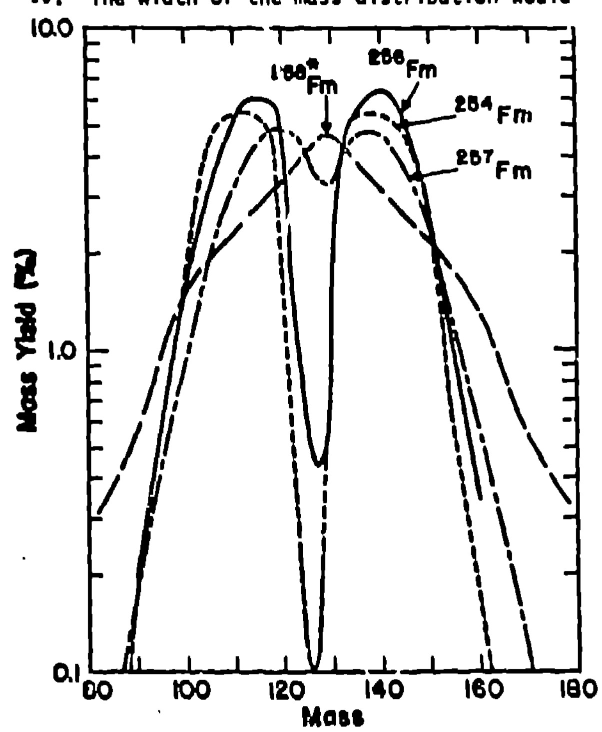

Fig. 1. Normalized mass-yield distributions for SF of ^{254}Fm (RC, Ref. 9), ^{256}Fm (RC, Ref. 7), ^{257}Fm (SS, Ref. 3) and for ^{258}Fm* (SS, Ref. 4).

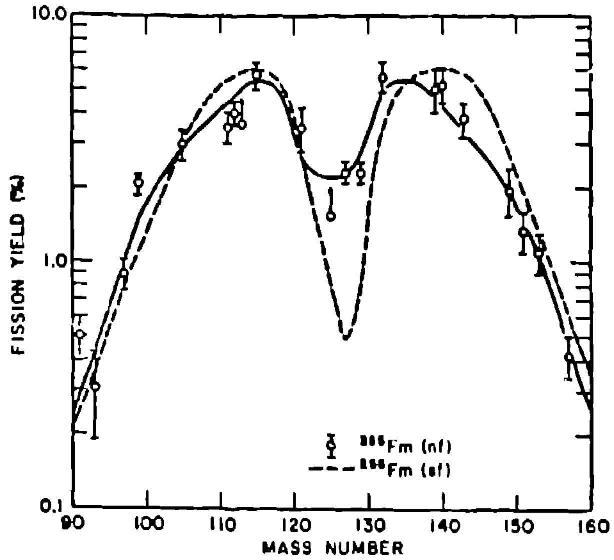

Fig. 2. Mass-yield curves for ^{256}Fm* and SF of ^{256}Fm (from Ref. 10).

be very small even though there might be a spread in total kinetic energy because some of the fragments may still be deformed.

3. Kinetic Energy Distributions

The measured and calculated values for the TKE for low energy fission of the fermium isotopes and peak-to-valley ratios for the mass distributions

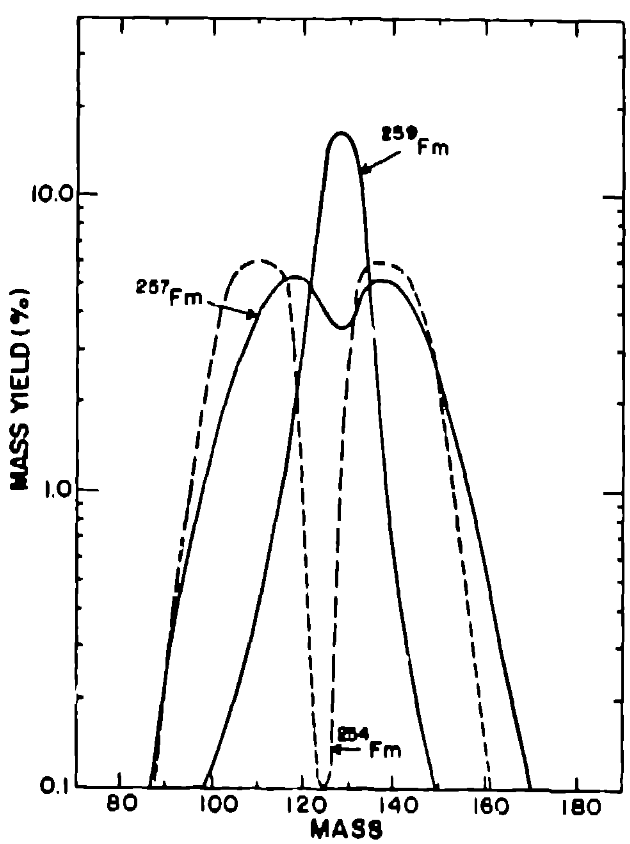

Fig. 3. Normalized mass-yield distributions for SF of ^{254}Fm (RC, Ref. 9), ^{257}Fm (SS, Ref. 3) and ^{259}Fm (SS, Ref. 12).

are summarized in Table I. The calculated values of TKE, based primarily on liquid-drop considerations, i.e., that the kinetic energy is a linear function of $Z^2/A^{1/3}$, show a slight decrease with increasing A while the measured values increase between ^{254}Fm and ^{256,257}Fm and show a precipitous increase at ^{259}Fm, presumably due to the larger proportion of spherical fragments. (Since the kinetic energy of the fragments arises primarily from Coulomb repulsion between the fragments, it is a maximum for spherical shapes.) ^{257}Fm (SF) shows the maximum TKE for symmetric mass splits in contrast to lighter actinides such as ^{254}Cf and ^{239}Pu* which show a sharp decrease in TKE at symmetry. This difference between ^{257}Fm and ^{254}Cf is shown in Figure 4. The high TKE for symmetric mass division of ^{257}Fm is presumably a consequence of the formation of two nearly doubly magic spherical fragments. In the case of symmetric division of the lighter actinides, the resulting fragments are farther from the closed shell configurations and are less spherical and have less TKE. It has been suggested[8]) that the very wide spread in TKE observed at symmetry for ^{254}Fm* and ^{256}Fm* as well as for ^{257}Fm (Figure 4) indicates that some of these fragments arise from symmetric scission of asymetrically deformed nuclei while the high energy fragments come from symmetric fission arising only from symmetric deformations of the fissioning system. The TKE distribution for ^{259}Fm is also very broad (σ = 28 MeV) even though the mass distribution is narrowly symmetric. Again, provided the large spread in TKE is not due to the contribution from another SF component[12]), the large width may indicate that some of the fragments are still rather "soft" toward deformation and that there is a large difference in fragment shapes, ranging from completely spherical to somewhat deformed. Even at ^{259}Fm we may still be observing a "transition" region as far as total kinetic energy is concerned.

The mass distribution for fission events from ^{257}Fm with total kinetic energy greater than 235 MeV has been obtained and it is highly symmetric[13]) with a FWHM of only about 7 mass units. (See Figure 5.) The TKE at symmetry for ^{257}Fm of 220 MeV is comparable to that of 230 MeV obtained for ^{259}Fm (SF) for which the mass division is primarily symmetric.

4. Neutron Emission

The total energy release in fission can be accounted for in the kinetic energies of the fragments prior to neutron emission and the excitation energies of the fragments. Thus as the kinetic energy goes up, the excitation energy, and hence neutron and/or gamma emission from the fragments must go down. From the data given in Table I, it can be seen that $\bar{\nu}_T$, the average number of neutrons emitted per fission, does indeed go down as TKE goes up between ^{254}Fm, and ^{256}Fm and ^{257}Fm. Studies of neutron emission in SF of ^{257}Fm and ^{252}Cf as a function of TKE show[13,14]) that for fission events with TKE > 240 MeV (~5% of the fissions), $\bar{\nu}_T$ drops to only 0.9 while for ^{252}Cf, it is 2.2 for the 3% of the fission events having the highest TKE's. The variances, $\sigma_{\bar{\nu}_T}$, for SF of ^{256}Fm and ^{257}Fm (Table I) are very large compared to that for ^{254}Fm and reflect the large spread in total kinetic energy and hence in excitation energy for those nuclides. The fragments with very high TKE emit very few neutrons and those with low TKE emit a larger number of neutrons.

Neutron emission for ^{259}Fm would be expected to be extremely low since its TKE of 230 MeV is approaching the total energy of about 250 MeV available from fission. However, as discussed in

TABLE I

PROPERTIES OF LOW ENERGY FISSION OF THE FERMIUM ISOTOPES[a]

Nuclide	P/V[b]	$\overline{TKE}$[c] (MeV) exp.	$\overline{TKE}$[c] (MeV) calc.[e]	$\bar{\nu}_T$[d]	$\sigma_{\bar{\nu}_T}$[d]
^{254}Fm	≈ 60 (RC)	192	198.7	3.95 ± 0.19	1.49 ± 0.20
^{256}Fm	12 (RC)	197.9	198.2	3.70 ± 0.18	2.30 ± 0.65
$^{256}Fm^*$	2.6 (RC)	195.6#	198.2	---	---
^{257}Fm	≈ 1.5 (SS)	197.6	197.9	3.77 ± 0.02	2.49 ± 0.06
$^{258}Fm^*$	Broadly (SS,RC) Symmetric	---	197.6	---	---
^{259}Fm	Narrowly (SS) Symmetric	230	197.4	---	---

[a] Data from summaries given in Refs. 5, 9 and 13.

[b] Peak-to-valley ratios for the mass distributions from either radiochemical (RC) or solid-state detector (SS) measurements.

[c] Average values for pre-neutron total kinetic energy except for those designated by # which are most probable values obtained from a Gaussian fit to the peak region. (All corrected to TKE = 186.1 MeV for ^{252}Cf.)

[d] Relative to $\bar{\nu}_T$ = 3.735 for ^{252}Cf.

[e] Calculated from the linear relationship, $\overline{TKE} = (0.13323Z^2/A^{1/3}) - 11.64$ obtained by Unik et al. (Ref. 9) from best fit to data for SF and thermal neutron-induced fission of nuclides from [illegible]Th to [illegible]Fm.

Section 3, the large spread in TKE for ^{259}Fm may indicate that some of the fragments are still deformed.

5. Dicussion

The trends in the fermium isotopes toward higher yields for symmetric mass division, higher total kinetic energies, and reduced neutron emission as the mass of the fermium isotopes is increased, are consistent with the simple postulate that the closer the fragments resulting from symmetric mass division are to the doubly magic Z = 50, N = 132 configuration, the more highly favored symmetric fission becomes. The fragments become more spherical which results in increased kinetic energy, decreased excitation energy and decreased neutron emission. The variances for the mass, kinetic energy and neutron distributions appear to be largest in the "transition" region including fermium-257 and 258 (and perhaps even fermium-259) where symmetric mass division apparently can result in fragment shapes ranging from rather deformed to nearly spherical as evidenced by the observation of symmetric mass division with both very high and very low total kinetic energies. However, at ^{259}Fm, symmetric mass division gives fragments with 79 or 80 neutrons which are now close enough to the 82 neutron shell so that most of them are quite spherical resulting in the observed highly symmetric mass distribution, very high total kinetic energies, low excitation energies and resultant low neutron emission. If fermium isotopes up to mass 264 could be observed, one might expect nearly all symmetric mass division with the total kinetic energy approaching the total fission energy available. Consequently, there would be very little excitation energy and essentially no neutron or gamma emission.

Various theoretical attempts to explain the existence of both asymmetric and symmetric mass distributions in low energy fission have been made[15-20]. Two main approaches can be identified: those in which the mass division is determined primarily by the deformation energy surface at the saddle point, and the "fragment shell" approaches in which the mass division is correlated with the potential energy surfaces in the neighborhood of scission, i.e., with the properties of the fragments. The trends observed in the fermium isotopes seem to indicate that any complete theoretical treatment will have to in some way recognize the influence of the fragment shells on the fission properties. Although disappearance of the second fission barrier[14,20] at [illegible]Fm can explain the sudden decrease in spontaneous fission half life and a change to symmetric mass division, some mechanism for incorporating the fragment shell effects seems to be required in order to account for the gradual transition from asymmetric mass division at [illegible]Fm to highly symmetric mass division at [illegible]Fm. Mustafa[21] has calculated this transition in the fermium isotopes on the basis of the potential energy surfaces and fragment shell effects in the two-center shell model and shows that the fermium isotopes up to mass 256 should fission asymmetrically, while mass 258 and heavier fermium isotopes should fission symmetrically. Recently, Wilkins et al.[22] have calculated total kinetic-energy and fragment mass distributions based on differences in the total potential energy of complementary fragment pairs, the highest yields being predicted for the lowest potential energies. They find the proportions of symmetric and asymmetric fission to be about equal for [illegible]Fm; at higher mass numbers symmetric fission becomes increasingly favored. The yield of symmetric mass splits is found to be a maximum for [illegible]Fm rather than [illegible]Fm because of the effect of an 80 neutron shell in the deformed fragment. They further predict that the TKE will maximize and be maintained at about 230 MeV in the region between [illegible]Fm and [illegible]Fm due to the formation of a somewhat deformed fragment with 80 neutrons in coincidence with a spherical fragment with 82 neutrons. (If both fragments were spherical a maximum TKE approaching 250-260 MeV, the Q-value for fission, would be expected to occur at a fermium mass of 264.) They calculate a return

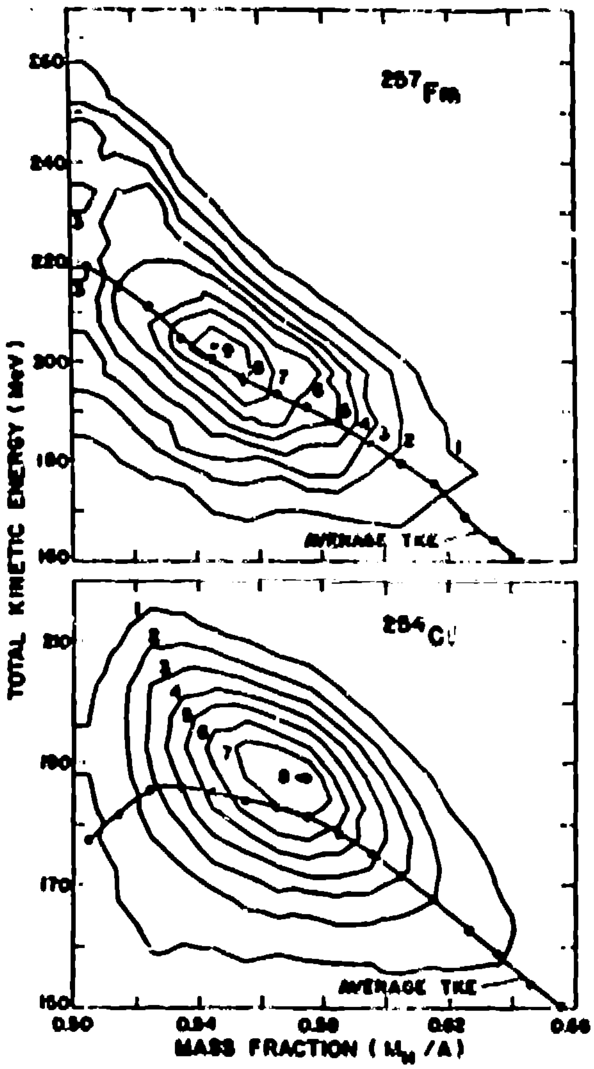

Fig. 4. Contour diagram showing pre-neutron-emission TKE distributions for ^{257}Cf as a function of mass fraction. The contours are lines of relative numbers of events, based on data groupings 5 MeV x 0.01 units of mass fraction (from Ref. 3).

to mass asymmetry for still higher mass fermium isotopes.

Measurements of the mass and kinetic energy distributions for SF of nuclides with Z greater than 100 will be particularly valuable in testing various theoretical predictions. If the second fission barrier is gone and mass division is determined at the first saddle point which is symmetric, then symmetric mass division for low energy fission should continue. If the effect of the fragment shells is the most important influence, then a return to asymmetric fission might be expected. Comparison of isotopes of elements 102 and 104 with those of fermium having the same number of neutrons will also be important in assessing the relative strength of the proton and neutron shells in determining fission properties.

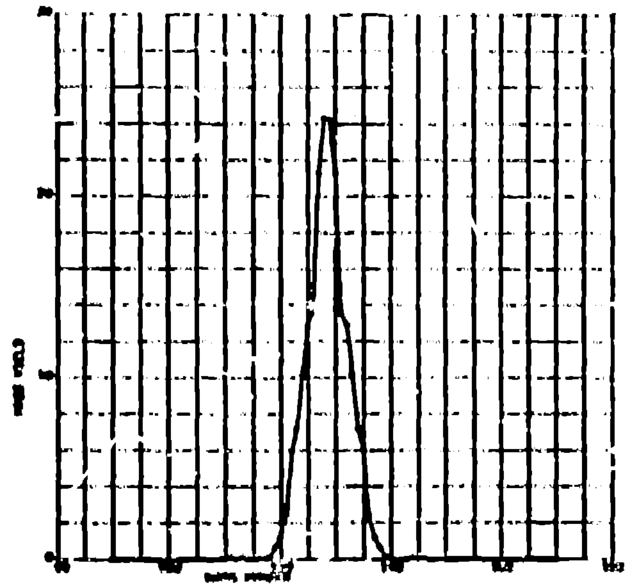

Fig. 5. Mass distribution for ^{257}Fm for fission events with TKE > 235 MeV, approximately 5% of the total fission events (from Ref. 13).

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