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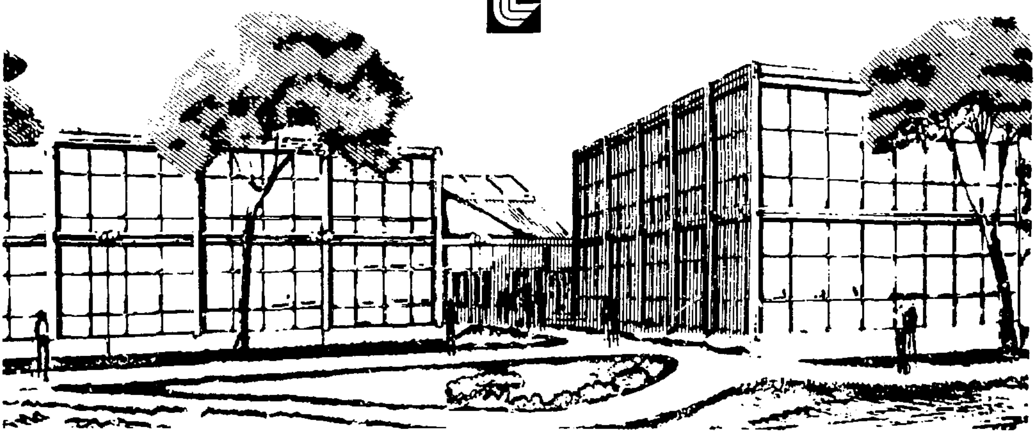

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THE SPONTANEOUS FISSION OF ^{259}Md*

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ABSTRACT

The mass and kinetic energy distributions of fission fragments from the spontaneous fission of the newly discovered nuclide ^{259}Md have been obtained. Mendelevium-259 was identified as the E. C. daughter of ^{259}No and was found to decay entirely (>95%) by spontaneous fission with a 95-min half-life. From the kinetic energies measured for 397 pairs of coincident fragments, we derived a mass distribution that is symmetric with $\sigma = 13$ amu. Mendelevium-259, together with ^{258}Fm and ^{259}Fm, form a select group of three nuclides whose mass division in spontaneous fission is highly symmetric. Unlike the total-kinetic-energy (TKE) distributions of ^{258}Fm and ^{259}Fm, which peak at $\approx$240 MeV, this distribution for ^{259}Md is broad and is 50 MeV lower in energy. Our analysis of the mass and energy distributions show that events near mass symmetry also exhibit a broad TKE distribution, with one-third of the symmetric events having TKE's less than 200 MeV. The association of low TKE's with symmetric mass division in the fission of very heavy actinides is anomalous and inconsistent with theories based upon the emergence of fragment shells near the scission point. We assume either three-body fragmentation or peculiar fragment shapes as the cause for the large consumption of coulomb energy observed for a significant fraction of symmetric fissions in ^{259}Md.

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INTRODUCTION

During the past decade, the mass and kinetic energy distributions have been measured for numerous spontaneously-fissioning (SF) nuclides in the region $92 \le Z \le 100$. With the exception of ^{258}Fm and ^{259}Fm, the mass divisions were found to be decidedly asymmetric with the most probable total kinetic energy (TKE) increasing slowly from ≈ 180 to ≈ 200 MeV with increasing Z of the fissioning nucleus. The mass distributions for the SF of ^{258}Fm and ^{259}Fm, on the other hand, are strongly symmetric and exhibit TKE's that are 40 to 50 MeV higher than predicted by systematics [1,2]. This rather abrupt change in fission properties, beginning with the heavy Fm isotopes, has brought about significant new insights concerning the fission process and has provided a new testing ground for fission theory. A major result has been to stress the importance of the part fragment shells play in the mass and energy division during fission.

The progress of nuclear fission theory in recent years has centered around the double-humped fission barrier caused by the stability of several nuclear shapes in the actinide region. The double-humped feature arises from shell corrections superimposed upon a smoothly varying, rotating liquid-drop potential [3]. Some believe that the inner barrier is responsible for symmetric fission and the outer barrier for asymmetric fission [4]. Calculations by Randrup et al. [5] suggest that the outer barrier disappears in the heavy actinide region, which could account for the observed transition from an asymmetric to symmetric mass division in the heaviest Fm isotopes. On the other hand, if the mass division is governed by the potential energy surface in the vicinity of scission, that is, the decent to the scission point is adiabatic, then the shell structure of the nascent fragments would determine the mass division [6]. According to this argument, the anomalous fission behavior of ^{258}Fm and ^{259}Fm would be due to their ability to fission into two ultra-stable, Sn-like fragments, $Z = 50$, $N \approx 82$.

Currently, fragment shell structure is viewed as the dominant factor in determining the fission mass split and the TKE. The TKE is traced to fragment shells because the TKE appears to be related to the ability of the fragments to remove deformation energy. However, the fission properties of ^{258}Fm and ^{259}Fm represent too limited a test of the validity of this or other possible hypotheses. The fission properties of the lighter Fm isotopes and a wide mass range of Cf isotopes have been examined to supply a baseline of systematic behavior. But to provide more extensive tests of the influence of fragment-shell structure or the effects from the disappearance of the second barrier on fission, we need SF data from selected nuclides in the transfermium region. In this region, it is particularly important that the neutron-rich nuclides be investigated because only here are the effects of the outer barrier fully suppressed. Also, if the fragments are to approach the $N = 82$ shell, then the fissioning species must be very neutron-rich.

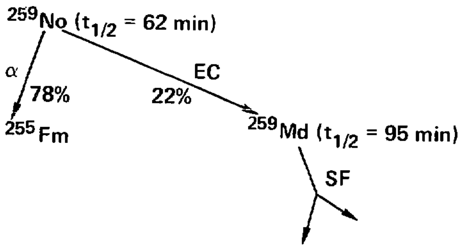
We have started a systematic study of fission in this region with an investigation of ^{259}Md, and we plan to continue with studies of the millisecond isotopes, ^{258}No and ^{260}[104]. The nuclide ^{262}[105] provides the only other case directly relevant to fission theory in the Z>100 region [7]. The mass distribution from the fission of ^{262}[105] was interpreted to be probably asymmetric, although less than 200 events were observed, together with a high background from the fission of ^{256}Fm coproduced in the bombardments. By studying the fission of ^{259}Md, which has the same number of neutrons as ^{258}Fm, we have begun to follow the trend in fission properties from symmetric fission in ^{258}Fm to possibly asymmetric fission in ^{262}[105].

Our first experiments were aimed at identifying the source of spontaneous fission activity which was earlier thought to belong to the decay of ^{259}No [8]. We chemically identified this activity as arising from Md following the E.C. decay of 62-min ^{259}No. After establishing a half-life for this new nuclide, we characterized the decay modes and partial half-lives for both ^{259}Md and ^{259}No. The SF properties of ^{259}Md were next investigated and these results are the main subject we treat in this paper. It should be noted that fission studies of these very heavy nuclei are exceedingly difficult and the results are by no means predictable.

EXPERIMENTAL

In all experiments, we initially prepared a pure sample of the parent, 62-min ^{259}No, from which the ^{259}Md would grow following E.C. decay. The ^{259}No was produced by the bombardment of a target of ^{248}Cm with 96-MeV ^{18}O ions from the 88-in cyclotron at the Lawrence Berkeley Laboratory. Products of the (^{18}O, α3n) reaction recoiling from the target were collected on a thin foil of either Pd or Au positioned directly behind the target. At the end of a bombardment typically 2-h long, the recoil foil was dissolved. The Pd or Au from the dissolved recoil collection foil was removed by adsorption on an anion-exchange column. The eluate, containing mainly No^{2+} and other trivalent actinides, was evaporated to dryness, redissolved in 0.1 M HCl, and eluted from a chromatographic-extraction column consisting of HDEHP dissolved in n-heptane, adsorbed on a fluoroplastic powder. This column adsorbed all of the trivalent actinides, including ^{256}Fm, ^{254}Fm, ^{248}Cm transferred from the target, and any other potential SF contaminant. We thus were assured of producing an isotopically pure source of ^{259}Md SF activity. Finally, we eliminated most of the inactive mass contamination, such as Ca or Mg, by means of a small cation-exchange column.

In the identification experiments, we evaporated samples of the purified ^{259}No parent onto Pt disks which were then pulse-height analyzed using surface-barrier detectors. The output from the counting system was routed through an ADC to a PDP-15 computer which recorded the energy and time of occurrence of each alpha and fission event on magnetic tape for subsequent off-line data analysis. A decay curve of the SF activity coming from samples of isolated Md is shown in Fig. 1 and indicates a single component decaying with a half-life of 95 min.

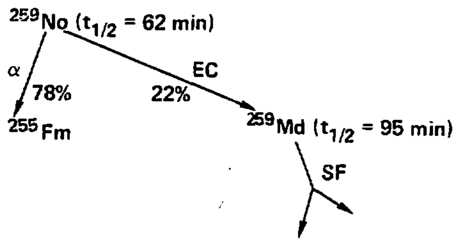

259No (t1/2 = 62 min)
α
78%
EC
22%
255Fm
259Md (t1/2 = 95 min)
SF

To characterize the SF decay properties of ^{259}Md, we arranged two coincidence counting systems, each consisting of two 450-mm^2 surface-barrier detectors mounted facing one another inside a vacuum chamber. Samples of ^{259}No-^{259}Md evaporated on thin VYNS films (typically 25-35 μg/cm^2) were placed between the detectors. Fission fragments from an event were detected in coincidence and the kinetic energy of each fragment was measured. As before, the output from the counting system was processed by the PDP-15 computer, which recorded the fragment energies and event times on magnetic tape. The mass of each fragment was derived from kinematic considerations.

We used a ^{252}Cf SF source to calibrate the fragment-energy response of the detectors, and employed the mass-independent calibration procedure of Schmitt, Kiker, and Williams [9]. The ^{252}Cf source on VYNS film was similar in thickness to the ^{259}Md sources, which reduced our calibration errors due to differing source thickness. Shortly after collecting the SF data for ^{259}Md, we prepared sources of ^{256}Fm (2.6 h, 92% SF) and ^{257}Fm (101 d, 0.2% SF) on VYNS film and analyzed these in our coincidence counting system. The kinetic energy and mass distributions of these two nuclides are already known [10,11] and they, therefore, served to verify our calibrations. The mass vs TKE distributions for ^{256}Fm and ^{257}Fm, as well as that obtained for ^{252}Cf, are shown in the contour plots of Fig. 2.

RESULTS AND DISCUSSION

We became aware that ^{259}No did not possess a spontaneous fission mode of decay when no SF activity was found in samples of ^{259}No immediately after chemical separation. But as the ^{259}No decayed with a 1-h half-life, SF activity grew in and eventually decayed with a new half-life of 95 min. The 95-min SF activity was proven to be associated with ^{259}Md by chemical separation of Md from purified No and observation of the 95-min decay of spontaneous fissions in the Md fraction. Mendelevium was repeatedly separated from a sample of pure ^{259}No and from this we were able to show that the growth period for ^{259}Md corresponded to the 1-h half-life of ^{259}No. An α-decay branch in ^{259}Md was not observed in the α spectra obtained with pure Md samples. These data would allow an upper limit of 5% for α-decay by ^{259}Md; thus, the predominant decay mode is spontaneous fission. The decay sequence is summarized as follows:



Our experiments do not rule out the possibility of ^{259}Md decaying by E.C. to 1.5-s ^{259}Fm, which would then be the source of the observed SF activity. However, this seems unlikely because most (but not all) mass equations [12, 13] and closed decay-cycle calculations [14] indicate that ^{259}Md is stable with respect to decay toward ^{259}Fm and, indeed, that ^{259}Fm is β^- unstable.

Following the identification of the new isotope ^{259}Md, we performed eighteen separate bombardments in which ^{259}No was chemically separated, deposited on thin films, and the energies of coincident fission fragments measured. A total of 397 fragment pairs was obtained from which we calculated mass and kinetic energy distributions. The mass distribution, illustrated in Fig. 3, is seen to be highly symmetric; however, there are indications of a small asymmetric component. This symmetric mass division is most comparable with the symmetric fission of ^{258}Fm and ^{259}Fm, the only other nuclides found so far that yield highly symmetric mass distributions. In the way of further comparisons to ^{259}Md, both the SF and neutron-induced fission of ^{257}Fm [11,15] contain a much larger component of asymmetric mass division.

The distribution of total kinetic energy (TKE) is shown in Fig. 4. The most probable TKE is 188 MeV while the average TKE is 185 MeV. The most probable TKE's measured for the nuclides ^{256}Fm and ^{257}Fm were found to be 195.0 MeV and 187.5 MeV, respectively, which can be compared with the post-neutron TKE's of 194.8 MeV and 195.1 MeV measured for these Fm isotopes by Unik et al. [10] and Balagna and co-workers [11]. Although our TKE for ^{257}Fm is low, the ^{256}Fm value is easily within the error limits of the measurements and we are satisfied with the accuracy of our calibrations.

As a result of comparing the SF properties of ^{259}Md with those of ^{256}Fm through ^{259}Fm, we find that ^{259}Md is unique. Although the mass distribution is closely comparable to the symmetric division in ^{258}Fm and ^{259}Fm, the most probable TKE is $\approx$50 MeV lower than found for these heavy Fm isotopes. The ^{259}Md TKE is most comparable to those of ^{256}Fm and ^{257}Fm. Such a low TKE associated with symmetric mass division is unusual and is inconsistent with current fission theory in which fragment shells appear to govern the fission process. Symmetric division of the heavy Fm isotopes leads to fragments approaching the magic nucleon numbers $Z = 50$, $N = 82$ which, due to their spherical rigidity, possess low deformation and internal excitation energy. Therefore, fissions with near-symmetric mass division exhibit correspondingly higher TKE's than those with asymmetric division, which yields fragments that are soft toward deformation.

To determine the extent of this deficit in TKE for ^{259}Md, we sorted our coincident events with respect to selected bands of TKE or mass. We then estimated the extent of correlation between symmetric mass division and high TKE. In the shaded section of Fig. 4, we see that events very close to mass symmetry have an average TKE about 20 MeV greater than the most probable TKE observed for all events. These high kinetic energy events seem to support the two-spheroid model of Schmitt and Mosel [16], based on fragment shell effects. However, about 35% of these highly symmetric fissions still exhibit TKE's less than 200 MeV. If broad energy cuts are taken for those events above and below 200 MeV, the mass distributions shown in Fig. 5 are obtained. The greater portion of symmetric mass divisions result in higher than average TKE's, but a significant percentage are associated with low kinetic energies. Finally, in Fig. 6, where the ^{259}Md mass distributions are compared with those of ^{256}Fm and ^{257}Fm, we find that only ^{259}Md yields a symmetric mass distribution in fission with TKE's under 200 MeV.

In sum, our analysis indicates we are observing a unique fission mode in the SF of ^{259}Md. To account for the 40-50 MeV loss in TKE for about a third of the symmetric events, it would be necessary to admit about 60 MeV of internal excitation and deformation energy in the fragments or to hypothesize three-body fragmentation whereby a light particle and two heavy fragments are emitted at the scission point. The storage of 60 MeV internal energy in the fragments is much more than can be accounted for by collective motions, angular rotation, or internal heating. Highly deformed symmetric fragments, in principle, can incorporate such a large potential energy. However, there is no evidence of such events (i.e., sizeable symmetric fission with TKE's <200 MeV) in the nearby Fm isotopes. Most fission studies have indicated a maximum excitation energy of ≈10 MeV in each fragment [17]. On the other hand, we estimate that light-particle emission would remove at least 25 to 30 MeV from the fissioning system and would be energetically favored if the particle is hydrogen-like (p,d,t). The emission of a Z = 1 particle obviously provides the opportunity for the remaining mass to divide into two Z = 50 fragments, which would be stabilized by filled proton shells. A test of this hypothesis is underway, but we expect it will require another long series of experiments to detect and identify light particles coming from the SF of ^{259}Md.

A trend toward mass asymmetry in the SF of nuclides with atomic numbers greater than that of Fm is barely discernible. The fission of ^{262}[105] is very probably asymmetric but ^{259}Md is only slightly less symmetric than ^{258}Fm and ^{259}Fm. Therefore, it might be expected that neutron rich nuclides between Md and element 105 would show a sharp reduction in symmetric fission properties, but the present evidence is still too slight to offer this as any more than a prediction. Only after the fission properties of other neutron-rich nuclides such as ^{258}No and ^{260}[104] have been measured can a trend be established.

REFERENCES

[1] HOFFMANN, D. C., WILHELMY, J. B., WEBER, J., DANIELS, W.R., LASL; HULET, E.K., LANDRUM, J. H., LOUGHEED, R. W., WILD, J. F., LLL, Detection of the new isotopes, 12.3-minute ^{256}Cf and 43-minute ^{258}Md, Los Alamos Scientific Laboratory Report LA-UR-77-2901 (1977); International Meeting on Reactions of Heavy Ions with Nuclei and Synthesis of New Elements, 13-16 December, 1977, Dubna.

[2] HOFFMANN, D. C., WEBER, J., WILHELMY, J. B., HULET, E. K., LOUGHEED, R. W., LANDRUM, J. H., and WILD, J. F., "Transplutonium 1975" (Proc. 4th Int. Transplutonium Element Symposium, North Holland, Amsterdam, 1976) 363; Proc. 3rd Int. Conf. on Nuclei Far From Stability, Cargèse, Corsica (CERN, Geneva, 1976) 558.

[3] STRUTINSKY, V. M., Nucl. Phys. A95 (1967) 420; A122 (1968) 1. MEYERS, W. D. and SWIATECKI, W. J., Nucl. Phys. 81 (1966) 1.

[4] MÖLLER, P. and NILSSON, S. G., Phys. Lett. 31B (1970) 283. PAULI, H. C., LEDERGERBER, T. and BRACK, M., Phys. Lett. 34B (1971) 264.

[5] RANDRUP, J., TSANG, C. F., MÖLLER, P., and NILSSON, S.G., Nucl. Phys. A217 (1973) 221.

[6] MOSEL, U. and SCHMITT, H. W., Phys. Rev C4 (1971) 2185.

[7] BEMIS, JR., C.E., FERGUSON, R. L., PLASIL, F., SILVA, R. J., O'KELLEY, G. D., KIEFER, M. L., HAHN, R. L., HENSLEY, D. C., HULET, E. K., and LOUGHEED, R. W., Phys. Rev. Lett 39 (1977) 1246.

[8] SILVA, R. J., DITTNER, P. F., MALLORY, M. L., KELLER, O. L., ESKOLA, P., ESKOLA, K., NURMIA, M., and GHIORSO, A., Nucl. Phys. A216 (1973) 97.

[9] SCHMITT, H.W., KIKER, W. E., and WILLIAMS, C. W., Phys. Rev. B137 (1965) 837.

[10] UNIK, J. P., GINDLER, J. E., GLENDENIN, L. E., FLYNN, K. F., GORSKI, A., and SJOBLOM, R. K., (Proc. 3rd IAEA Symp. Phys. Chem. of Fission, Rochester, New York, 1973), IAEA, Vienna, Vol. II (1974) 19.

[11] BALAGNA, J. P., FORD, G. P., HOFFMANN, D. C., and KNIGHT, J. D., Phys. Rev. Lett. 26 (1971) 145.

[12] WAPSTRA, A. H. and BOS, K., Atomic Data Nucl. Data Tables 17 (1976) 593.

[13] KOLESNIKOV, N. N. and DEMIN, A. G., Joint Institute for Nuclear Research, Dubna, USSR, reprint P6-9421 (1975); Lawrence Livermore Laboratory Translation UCRL-11098.

[14] VIOLA, JR., V. E., SWANT, J. A., and GRABER, J., Atomic Data Nucl. Data Tables 13 (1974) 60.

[15] JOHN, W., HULET, E. K., LOUGHEED, R. W., and WESOLOWSKI, J. J., Phys. Rev. Lett. 27 (1971) 45.

[16] SCHMITT, H. W. and MOSEL, U., Nucl. Phys. A186 (1972) 1.

[17] SCHULTHEIS, H. and SCHULTHEIS, R., Phys. Rev. C18 (1978) 317.

FIGURE CAPTIONS

Figure 1. The decay of spontaneous fission activity in samples of Md chemically separated from ^{259}No.

Figure 2. Contour diagrams of counts versus fragment mass and total kinetic energy for three nuclides used in calibrating the surface-barrier detectors. Numbers on the contours refer to the number of events.

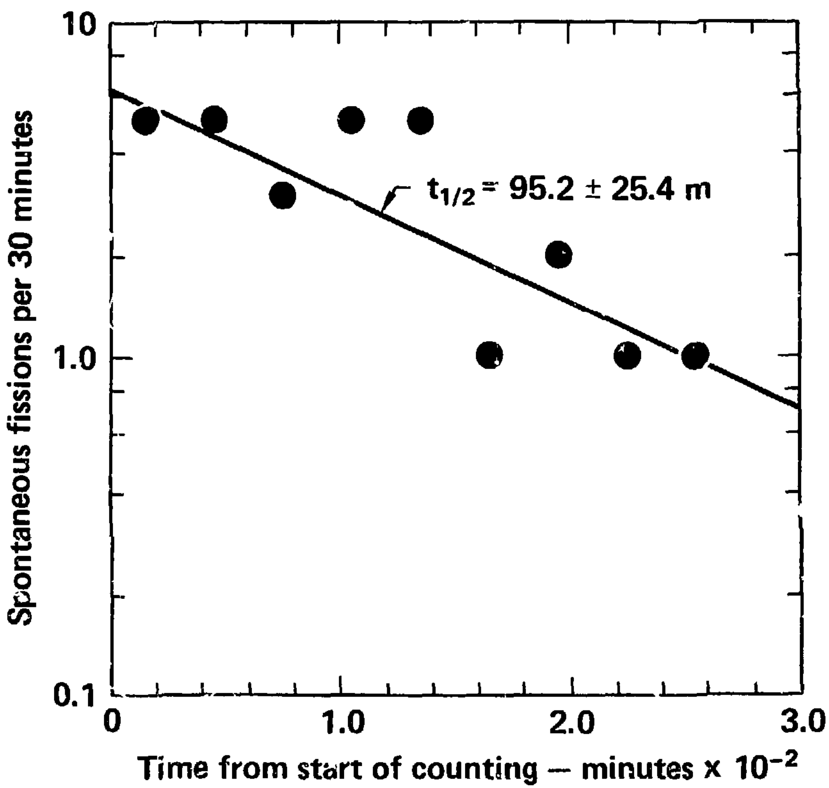
Figure 3. Provisional mass distribution for the spontaneous fission of ^{259}Md.

Figure 4. Postneutron total-kinetic-energy (TKE) distributions for the spontaneous fission of ^{259}Md. The most probable TKE is 187.5 MeV. The shaded region is the TKE distribution for a 5-amu wide band of fission events at mass symmetry.

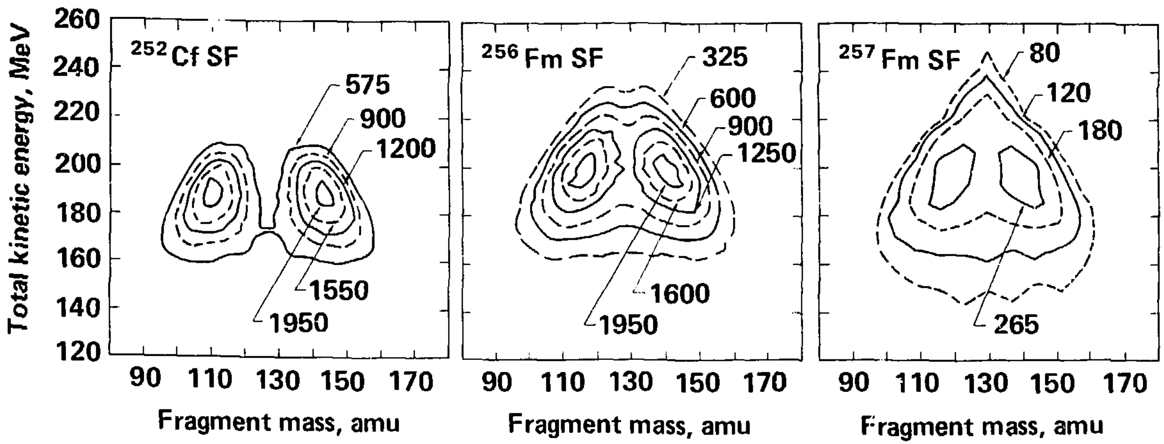
Figure 5. Provisional mass distributions for the spontaneous fission of ^{259}Md separated into two groups according to the total kinetic energy. The lower curve clearly shows a symmetric mass distribution for fission events with low kinetic energies.

Figure 6. Provisional mass distributions for ^{259}Md, ^{257}Fm, and ^{256}Fm sorted into groups as a function of their total kinetic energies. Each curve shows the mass distribution for fission events with total kinetic energies within a 40-MeV band. Only ^{259}Md retains a semblance of a symmetric distribution in low kinetic-energy fission.

Spontaneous-fission decay of ^{259}Md milked from ^{259}No

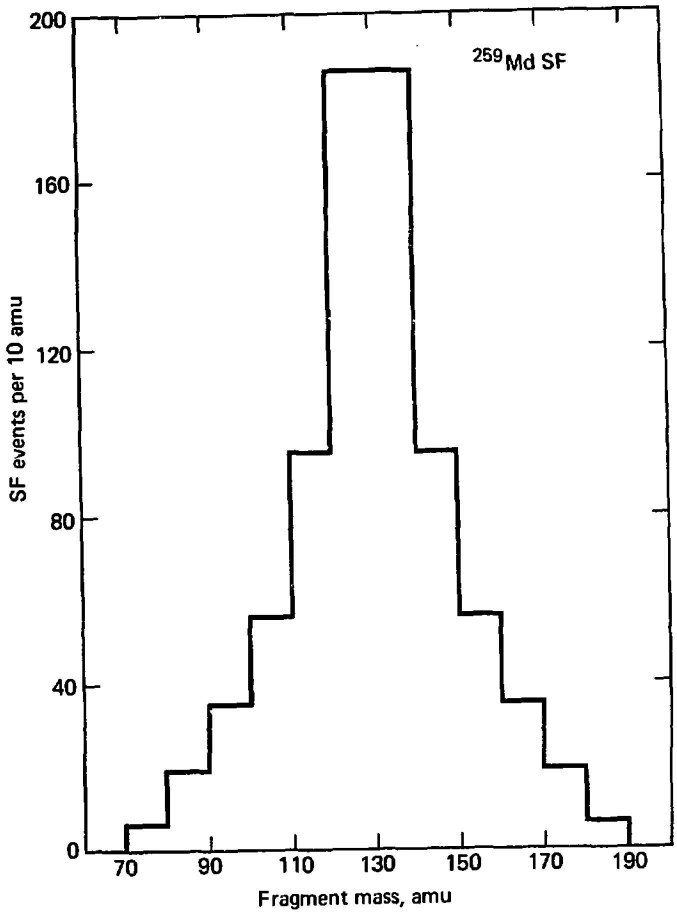


HULET - FIG. 1



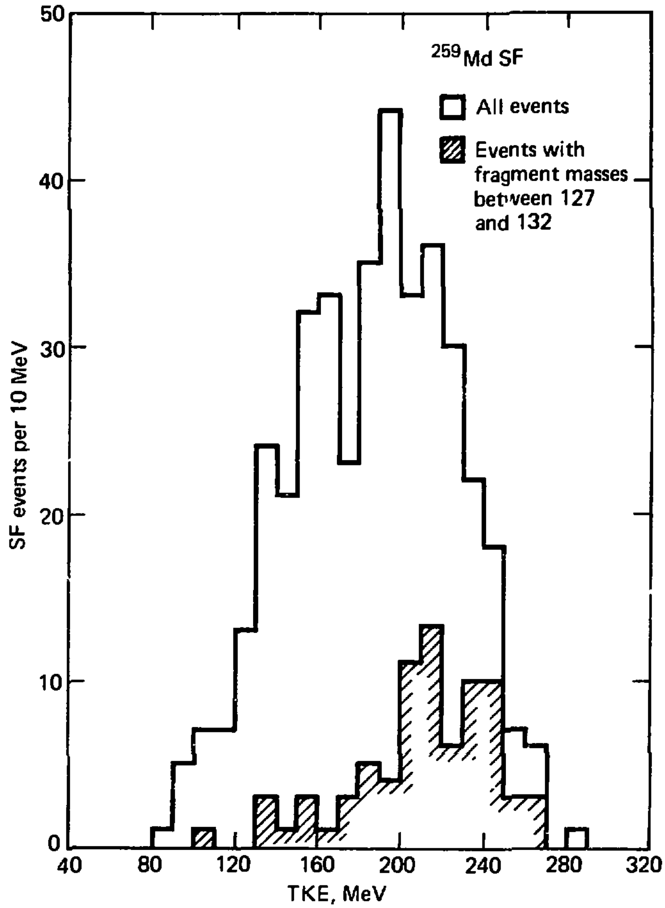
Total kinetic energy, MeV
260
240
220
200
180
160
140
120
252Cf SF
575
900
1200
1550
1950
90 110 130 150 170
Fragment mass, amu
256Fm SF
325
600
900
1250
1600
1950
90 110 130 150 170
Fragment mass, amu
257Fm SF
80
120
180
265
90 110 130 150 170
Fragment mass, amu

HULET - FIG. 2



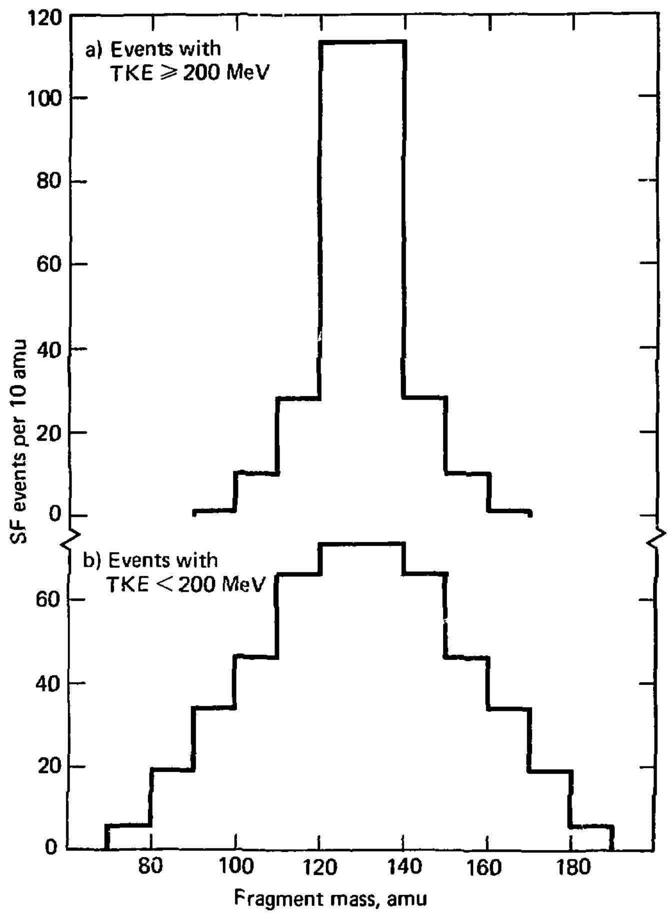
200
160
120
80
40
0
SF events per 10 amu
^{259}Md SF
70
90
110
130
150
170
190
Fragment mass, amu

HULET - FIG. 3

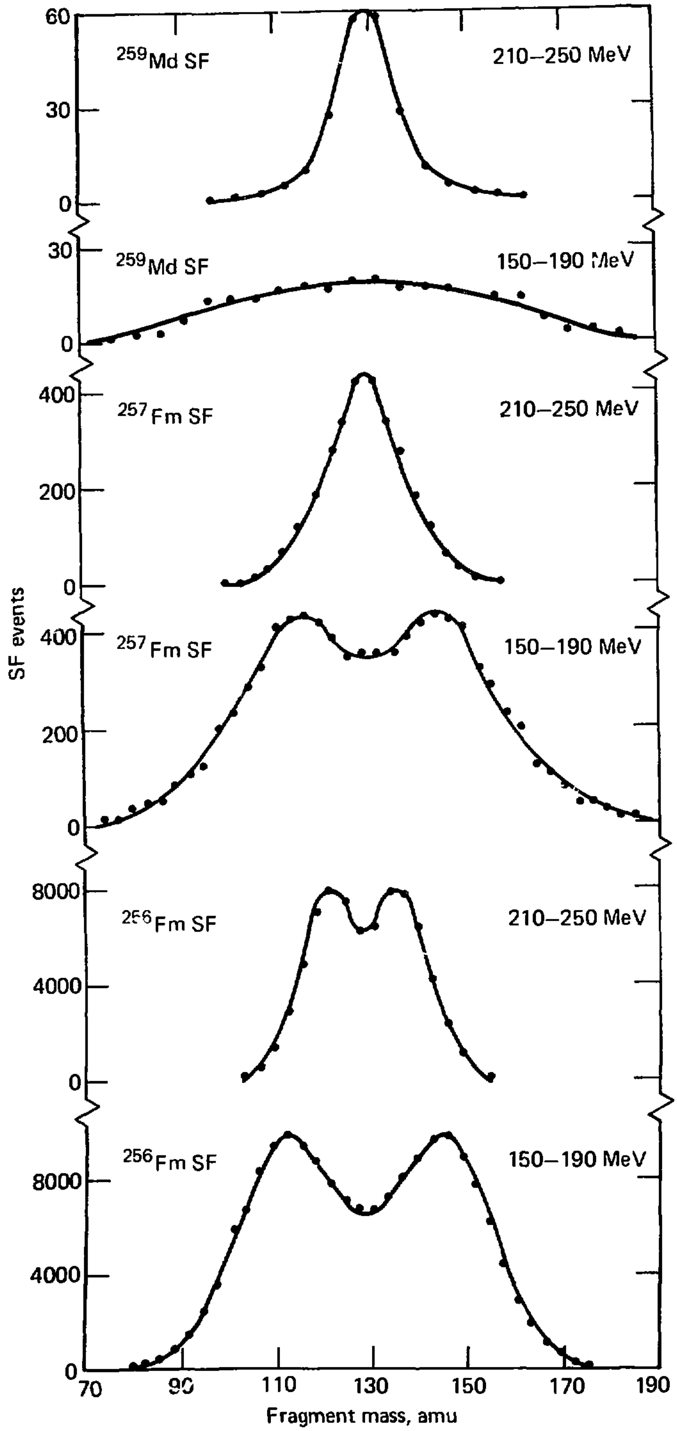


259Md SF
All events
Events with fragment masses between 127 and 132
SF events per 10 MeV
TKE, MeV
0
10
20
30
40
50
40
80
120
160
200
240
280
320

HULET - FIG. 4


120
100
80
60
40
20
0
a) Events with
TKE ≥ 200 MeV
b) Events with
TKE < 200 MeV
60
40
20
0
SF events per 10 amu
80
100
120
140
160
180
Fragment mass, amu

HULET - FIG. 5



60
30
0
^{259}Md SF
210–250 MeV
30
0
^{259}Md SF
150–190 MeV
400
200
0
^{257}Fm SF
210–250 MeV
400
200
0
^{257}Fm SF
150–190 MeV
8000
4000
0
^{256}Fm SF
210–250 MeV
8000
4000
0
^{256}Fm SF
150–190 MeV
SF events
70
90
110
130
150
170
190
Fragment mass, amu

HULET - FIG. 6