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ABSTRACT

Although spontaneous fission was discovered in ^{238}U in 1940, detailed studies of the process were first made possible in the 1960's with the availability of milligram quantities of ^{252}Cf. The advent of solid-state detectors made it possible to perform measurements of coincident fission fragments from even very short-lived spontaneous fission activities or those available in only very small quantities. Until 1971 it was believed that the main features of the mass and kinetic-energy distributions were essentially the same as those for thermal neutron-induced fission and that all low-energy fission proceeded via asymmetric mass division with total kinetic energies which could be derived by linear extrapolation from those of lighter elements. In 1971, measurements of ^{257}Fm showed an increase in symmetric mass division with anomalously high TKE's. Subsequent experiments showed that in ^{258}Fm and ^{259}Fm, the most probable mass split was symmetric with very high total kinetic energy. Measurements for the heavier elements have shown symmetric mass distributions with both high and low total kinetic energies. Recent results for spontaneous fission properties of the heaviest elements are reviewed and compared with theory.

I. INTRODUCTION

During the nearly 50 years since the discovery by Petrzhak and Flerov[1] of the spontaneous fission (SF) mode of decay in ^{238}U, a wealth of information on spontaneous fission half-lives and properties has been obtained and is summarized in several reviews[2-7]. Studies of the SF properties of the heaviest elements have been dependent on the synthesis of nuclides with higher atomic number which have much shorter partial half-lives and consequently much larger branches for SF decay than does ^{238}U. In the 1960's, milligram quantities of 2.6-year ^{252}Cf (3% SF branch) became available to researchers under the transplutonium production program of the U.S.Atomic Energy Commission. It has been widely used for detailed studies of such fission properties as mass and charge division, fragment kinetic energies, neutron emission, and the variation in these properties as a function of total energy release and excitation energy of the fragments. The production[8] of other heavy elements in high flux reactors has also been extremely important. A summary of annual production during operation of the High Flux Isotope Reactor at Oak Ridge National Laboratory is given in Table I. These isotopes have been important both for studies of their SF properties and for use as targets for production of spontaneously fissioning isotopes in accelerator irradiations. Practically speaking, the heaviest isotope which can be produced in reactors is ^{257}Fm, which is produced in only picogram amounts. It was actually used as a target[9] to produce 1.5-second ^{259}Fm via the (t,p) reaction. Studies of the SF of ^{257}Fm were the first[10] to show a high yield of symmetric mass division in spontaneous or thermal-neutron induced fission and sparked a renaissance of interest in the study of

Table 1. Mainline HFIR-TRU production (elements 96–100)

Isotope	Half-life (principal decay mode)	Production (amount/year)
Curium-248	3.397×10^5 years (alpha)	150 mg
Berkelium-249	320 days (beta)	50 mg
Californium-249	350.6 years (alpha)	from ^{249}Bk decay
Californium-252	2.64 years (alpha)[a]	500 mg
Einsteinium-253	20.4 days (alpha)	2 mg
Einsteinium-254	275.7 days (alpha)	4 μg
Fermium-257	100.5 days (alpha)	1 pg

[a] ^{252}Cf has a neutron output from spontaneous fission of 2.3×10^{12} neutrons/s/g.

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spontaneous fission of the heavy nuclides. Subsequent measurements[9,11] showed that the SF of both ^{259}Fm and ^{258}Fm resulted in extremely narrow, symmetric mass distributions and that the total kinetic-energy release was anomalously high. The addition of only a single neutron had completely changed the observed fission properties! Much time had been devoted earlier to trying to explain why the mass distributions for low-energy fission were asymmetric rather than symmetric as predicted by the liquid-drop model. Now suddenly with the addition of one or two neutrons a dramatic change in the observed spontaneous fission properties had occurred! The current state of our knowledge of the half-lives and SF properties of the heaviest nuclides will be summarized and compared with theoretical predictions, and some future experiments to be undertaken will be discussed.

HALF-LIVES

One of the most fundamental properties of spontaneous fission which can be measured is the partial SF half-life (or branching ratio). A complete tabulation[7] as of mid-1986 lists 118 SF half-lives including those for isomeric states and beta-and electron-capture delayed fission, as well as ground-state decay. A few additional very important measurements have been added[12] since that time. The variation in half-life as a function of the atomic number, Z, and neutron number, A, of the fissioning nucleus is of particular interest. Systematic trends have been observed for the even-even (e-e) isotopes. In general, the half-lives decrease by several orders of magnitude with the addition of 2 protons. However, extra stability due to filling of a deformed subshell around 152 neutrons becomes apparent at Cm and is strongly manifested in Fm and No with half-lives decreasing sharply for isotopes with smaller or larger neutron numbers. At Rf there is a marked change in half-life systematics, presumably due to the disappearance of the subshell effect, and the SF half-lives change only slowly as a function of neutron number. A comparison of recent experimental measurements of Münzenberg et al.[13] for the partial SF half-lives of ^{260}106 (7ms) and ^{264}108 (>5 ms) with that of ^{258}Rf (14 ms) shows that the rate of decrease in SF half-lives has slowed dramatically for the higher Z isotopes allowing alpha decay to predominate.

New results of Ćwiok et al.[14] and Möller et al.[15,16], using different calculational methods, both show the appearance of a new, second valley to fission in the potential energy surfaces calculated as a function of deformation energy via the macroscopic-microscopic model using the Strutinsky shell correction method[17]. This new valley leads to near-spherical or compact shapes and shorter SF half-lives, giving better agreement with results for the Fm isotopes and explaining features of the mass and kinetic-energy distributions. Subsequently, Patyk, Skalski, Sobiczweski, and Ćwiok[18] have used both static and dynamic treatments to calculate SF half-lives for a wide region of e-e nuclides with Z=100 to 130. Again, as in Ref. 14, two valleys to fission are obtained, one leading to elongated and the other to compact fragment shapes. A dynamic treatment was used to find the fission trajectory which minimizes the total action integral in the deformation space considered. These calculations fit the experimental data for the Fm SF half-lives rather well although the precipitous drop in half-life at N=158 occurs two neutrons too soon. Surprisingly, their results show that the half-lives for Fm and No isotopes should increase again for neutron numbers larger than about 158 to 160, presumably due to the influence of a deformed shell around N=162 to 164. For element 104 and beyond, the SF half-lives are predicted to increase continually with increasing neutron number. In addition to the "traditional" superheavy element (SHE) nuclei with N close to the spherical 184 shell, they predict well-deformed nuclei with 164 to 168 neutrons for similarly large proton numbers, resulting in an extended peninsula, not just an island of stability.

Hindrance factors (HF) have long been known to be associated with the SF decay of nuclides with an odd number of protons or neutrons relative to the half-lives of their e-e neighbors. A plot of the logarithm of the HF's calculated for odd-neutron and odd-proton nuclides is shown in Fig. 1. The log HF's average about 5 for both odd protons and odd neutrons. Only lower limits are available for Z=105 and 107 and they are somewhat smaller than 5. The log HF for N=157 appears to be exceptionally large, but may just reflect the very short half-life measured for ^{258}Fm. It is still not clear whether the log HF's for nuclides with both an odd neutron and odd proton are the sum of the respective log HF's or not. For example, for ^{260}Ha (16 s) and ^{262}Ha (68 s), the log HF's relative to the neighboring e-e nuclei are only about 3 while for 32-day ^{260}Md relative to ^{258}Fm (0.37 ms) and ^{260}No (106 ms?), the log HF's are 9.9 and 7.5, respectively. Encouragingly, the log HF's for the recently discovered[19,20] 216-min ^{262}Lr (<10% SF) are 6 and 7 relative to ^{260}No and ^{262}No (5 ms), respectively. These observed large hindrances make the existence of longer-lived odd-odd species of the higher Z elements appear quite likely.

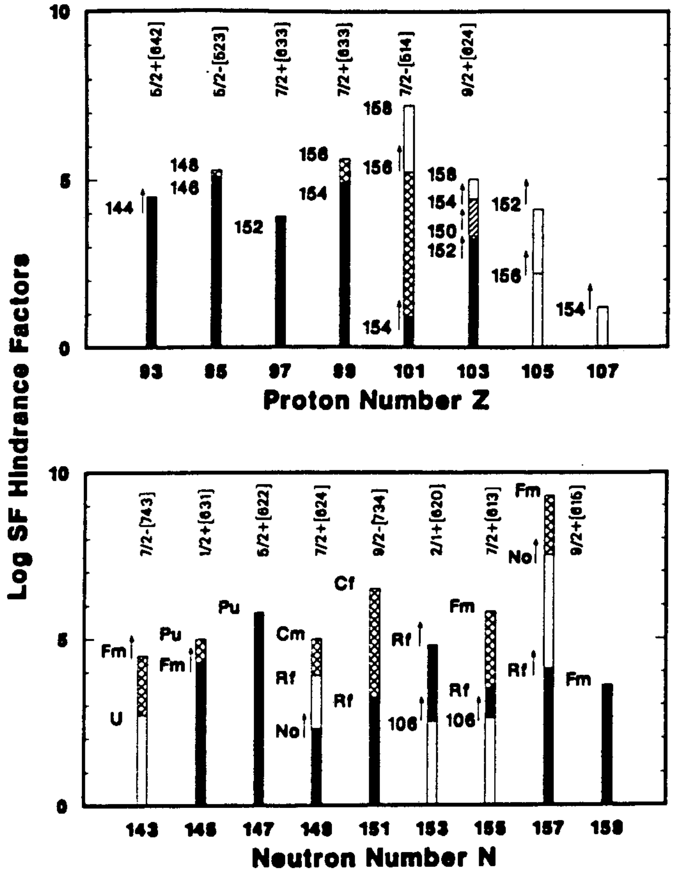

FIGURE 1 LOGARITHMS OF SF HINDRANCE FACTORS FOR ODD-NEUTRON AND ODD-PROTON NUCLIDES. LOWER LIMIT VALUES ARE INDICATED BY ARROWS. AN OPEN BAR INDICATES THAT THE HF WAS CALCULATED RELATIVE TO ONLY ONE E-E NEIGHBOR.

MASS AND KINETIC-ENERGY DISTRIBUTIONS

Historically, fragment mass-yield distributions have been obtained primarily by radiochemical methods[2], but the use of solid-state detectors has made it possible to measure the kinetic energies of coincident fission fragments with rather high efficiency. The masses of the fragments can then be derived assuming conservation of momentum if it is assumed that the isotopic assignment of the fissioning nucleus is known. Most of the information for the heaviest isotopes has been obtained by this method which is readily adaptable to on-line measurements of isotopes with very short half-lives which are often produced only in very small quantities. This technique does not have the "perfect" mass resolution of radiochemical techniques nor does it furnish any information about the atomic number of the fragments, but valuable information about the kinetic energy release is obtained. Both of these methods give fragment masses after prompt neutron emission so it is necessary to make corrections for neutron emission as a function of fragment mass in order to obtain primary (preneutron emission) fragment distributions.

Mass-yield and kinetic-energy distributions have been measured for more than 20 trans-Bk isotopes, the most neutron-rich being the recently discovered[19] ^{262}No. The mass distributions for trans-Bk isotopes are shown schematically in Fig. 2. They illustrate the abrupt change from the asymmetric mass distributions observed for the SF of lighter Fm isotopes (and the lower Z actinides) to the narrowly symmetric mass distributions observed for ^{258}Fm and ^{259}Fm. The average total kinetic energy ($\overline{TKE}$) for these nuclides is also anomalously high as shown in Fig. 3 where $\overline{TKE}$ is plotted as a function of $Z^2/A^{1/3}$. A topographical representation of the mass and kinetic energy surface for ^{259}Fm is shown in Fig. 4.

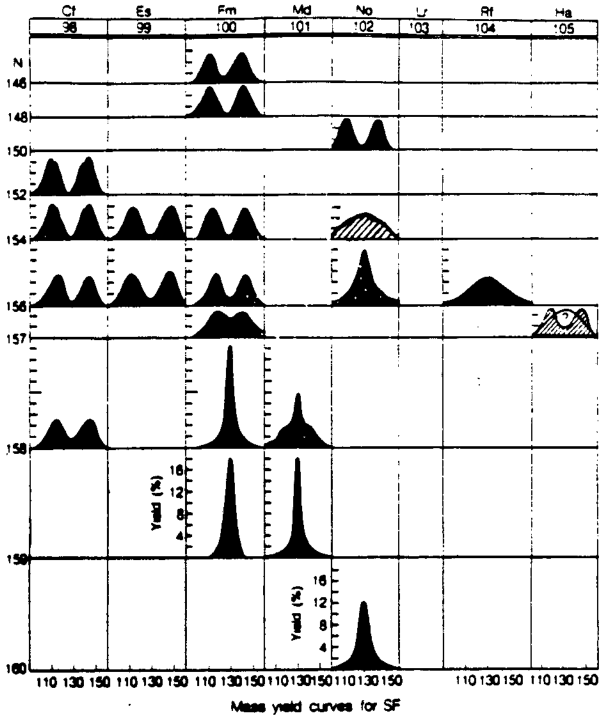

FIGURE 2 SCHEMATIC OF MASS-YIELD DISTRIBUTIONS (NORMALIZED TO 200% FISSION FRAGMENT YIELD) FOR SF OF TRANS-Bk ISOTOPES, 1989A.

The change from asymmetric to symmetric mass division in the Fm isotopes coincides

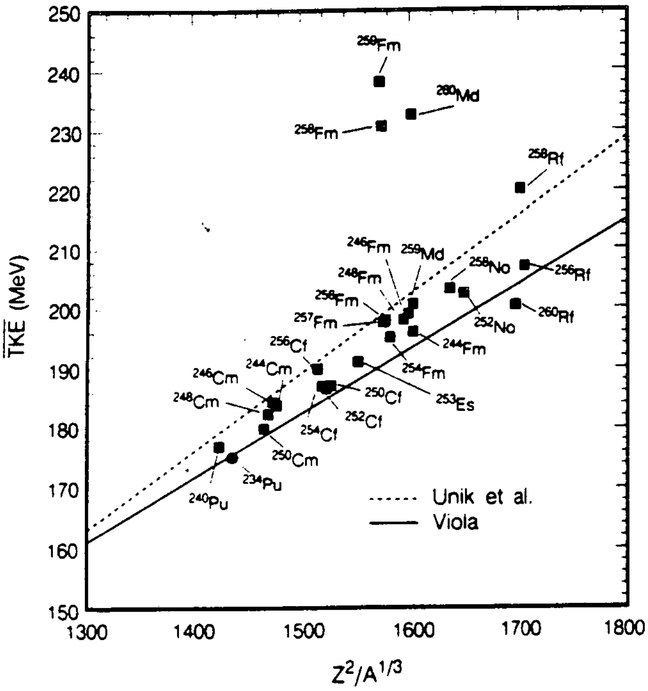

FIGURE 3 $\overline{TKE}$ VS. $Z^2/A^{1/3}$. DASHED LINE IS LINEAR FIT OF UNIK ET AL.[20]; SOLID LINE IS FROM VIOLA[21].

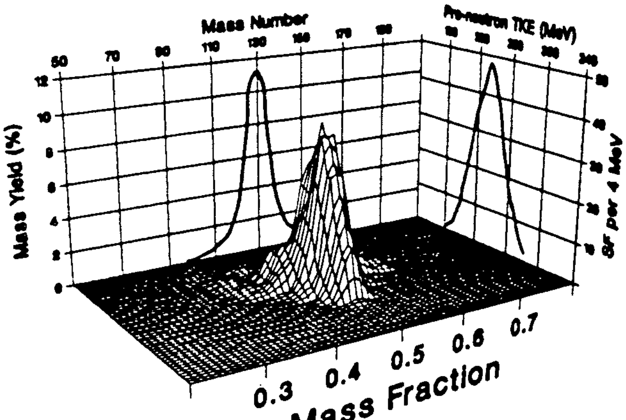

FIGURE 4 TOPOGRAPHICAL REPRESENTATION OF MASS AND KINETIC-ENERGY SURFACES FOR ^{259}Fm.

with the precipitous decrease in half-life of 3 x 10^7 which occurs between the SF half-life of 2.9 hours for ^{256}Fm and that of only 0.37 ms for ^{258}Fm. These effects have been attributed to the disappearance of the second barrier in the potential energy surface of the fissioning nuclide, or more recently[14-16,18] to the appearance of a new symmetric valley leading to near-spherical fragments. ^{259}Md shows a symmetric mass distribution with some evidence of asymmetric division as well, but the $\overline{TKE}$ appears "normal", indicating that a large fraction of the fragments are deformed. ^{260}Md exhibits a narrowly, symmetric mass distribution, similar to that of ^{258}Fm and ^{259}Fm and also shows an "anomalously" high $\overline{TKE}$. These observations can be explained qualitatively on the basis of symmetric mass division into two nearly spherical fragments which approach the doubly magic ^{132}Sn configuration and, therefore, exhibit near maximum TKE's due to the Coulomb repulsion which will be a maximum for two touching spherical nuclei.

Enough information is now available for the No isotopes to see a transition from asymmetric to symmetric mass division which is similar to that observed for the Fm isotopes. The mass distribution for ^{252}No is asymmetric, similar to those observed for Fm isotopes with mass 256 or lighter, and its $\overline{TKE}$ is "normal". Our preliminary measurements for ^{256}No indicate that it is a "transition" nucleus as shown in Fig. 2 by a dashed curve. Its mass distribution appears to be nearly flat across the top, and like the transition nucleus ^{257}Fm which it resembles, whether or not there is a slight decrease in fragment yields at symmetry depends on whether a correction[10] is applied for prompt neutron fission using a "sawtooth" $\bar{\nu}(M)$ function. The most probable mass split for ^{258}No has been shown earlier by Hulet et al.[23] to be symmetric but with a broader distribution than for ^{258,259}Fm and its $\overline{TKE}$ is near "normal" (Fig. 3), suggesting considerable deformation of the fragments. The recent measurements[19,20] for ^{262}No show a narrowly symmetric mass distribution with a most probable TKE of about 237 MeV, but with some evidence for a lower intensity component around 200 MeV. Thus, the transition to predominantly symmetric mass division has occurred by ^{258}No (N=156), two neutrons fewer than for Fm. However, based on the considerably lower TKE, it appears that the fragments from SF of ^{258}No exhibit substantially more deformation. With the addition of 2 more protons, a still broader symmetric mass distribution, reminiscent of liquid-drop type fission, is observed[23] for ^{260}Rf (N=156). Its $\overline{TKE}$ is only about 200 MeV, somewhat lower than the fits shown in Fig. 3. The only information available for still higher Z nuclides is for 35-second ^{262}Ha. Measurement of the mass and TKE distributions for ^{262}Ha (N=157) are very difficult because of the production of other fissioning species which complicate the interpretation of the results. Bemis et al.[24] report the mass distribution to be asymmetric, but they cannot rule out the possibility of a very broad symmetric distribution.

Recently, our group has initiated studies of electron-capture (ec) delayed fission. This decay mode provides a method for measuring fission properties at near ground-state energies of e-e nuclei far from stability which would otherwise be inaccessible. The mass and kinetic-energy distributions have been measured[25] for 1.3-minute ^{232}Am and 2.3-minute ^{234}Am, presumably due to fission of the daughter nuclei ^{232}Pu and ^{234}Pu produced by the ec-decay. A topographical representation of the mass and kinetic-energy distributions for ^{234}Am (^{234}Pu) is shown in Fig. 5. The highly asymmetric mass distribution is typical of the ground-state fission of the lighter actinides and does not show increased yields for symmetric mass division which might be indicative of the three-humped mass yield curves observed[26] in the region of Ra isotopes with similar neutron numbers. The TKE also appears "normal".

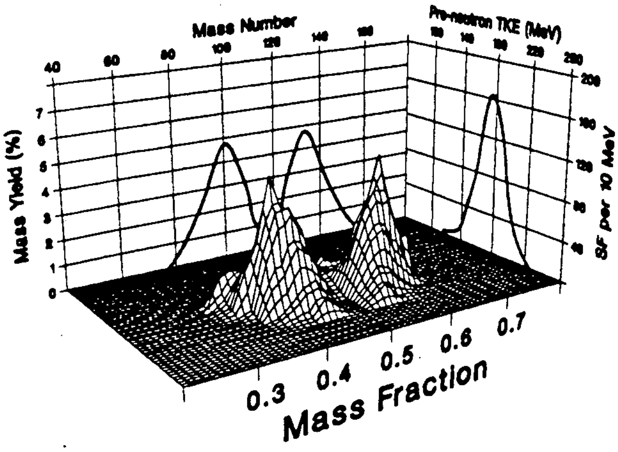

FIGURE 5 TOPOGRAPHICAL REPRESENTATION OF MASS AND KINETIC-ENERGY SURFACES FOR ^{234}Am EC-DELAYED FISSION.

Schematic representations of the TKE distributions for some trans-Es isotopes are shown in Fig. 6. Some of them are asymmetric and Hulet et al.[23] have decomposed the distributions for ^{258}Fm, ^{258}No, ^{259}Md, and ^{260}Md, into two Gaussian distributions of varying intensities, one centered around 200 MeV and the other around 235 MeV. The TKE distribution for ^{262}No has also been similarly resolved by Lougheed et al,[19,20] into a Gaussian distribution centered at 238 MeV and a less intense one at 200 MeV. They call this "bimodal" symmetric fission--postulating that one symmetric mode leads to nearly spherical fragments with anomalously high TKE due to the higher Coulomb repulsion and the other to elongated fragments with lower TKE. These two modes may be associated with the two valleys to fission discussed earlier. Other heavy nuclides appear to show similar features. The "transition" nucleus ^{257}Fm shows a "bulge" on the high energy side which can be seen better in the contour plots shown in Fig. 7. The range of TKE's for symmetric mass division is extremely large, indicating shapes ranging from near-spherical with TKE's approaching the Q value to deformed shapes with low TKE, and combinations of spherical and elongated shapes. This appears to be multimodal rather than just bimodal symmetric fission. Calculations of Paskevich[27] for ^{264}Fm have shown 3 valleys on the potential-energy surface for symmetric fission in the region of the scission point. One corresponds to a compact configuration of two nearly spherical fragments, another to more separated highly elongated fragments, and a third corresponds to a combination of spherical and elongated fragments. Indeed, all three modes seem to exist in ^{257}Fm. The contour plot for our preliminary measurements of ^{256}No shows a nearly constant yield extending from symmetric mass division to a mass fraction of about 0.57 or A=146. The $\overline{\text{TKE}}$ is nearly constant throughout this region at 200 to 210 MeV. This seems to indicate that both symmetric and asymmetric fission events

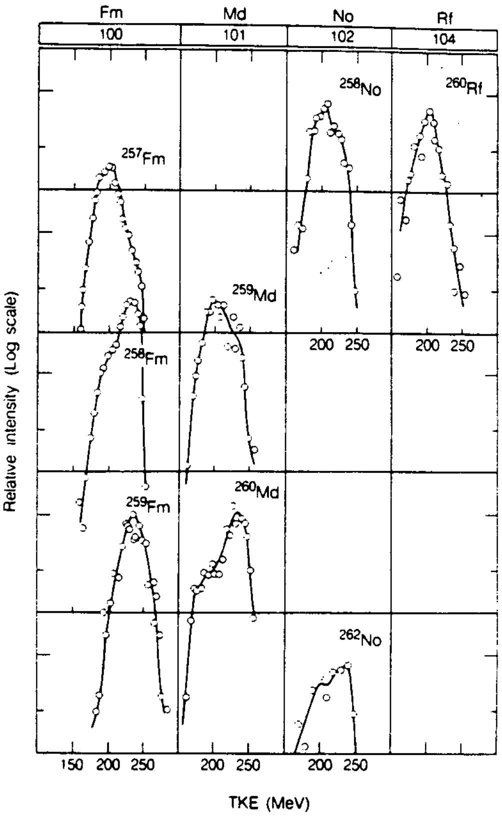

FIGURE 6 SCHEMATIC OF TKE DISTRIBUTIONS OF TRANS-Es ISOTOPES, 1989.

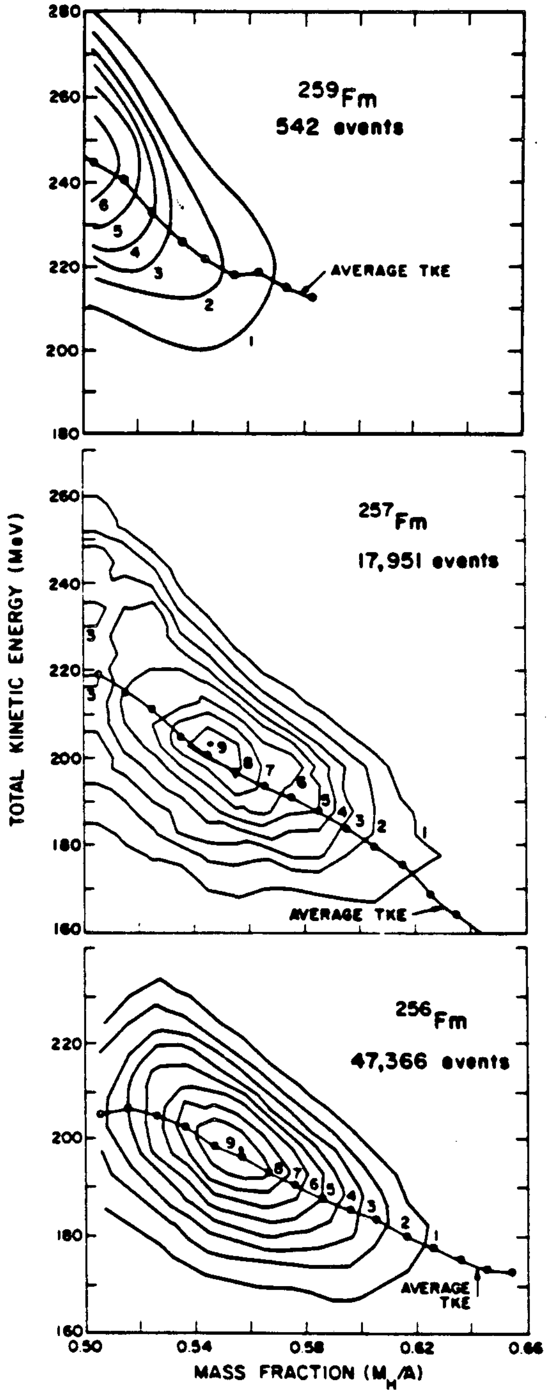

FIGURE 7 CONTOUR PLOTS OF $\overline{TKE}$ AND TKE VS. MASS FRACTION FOR ^{256}Fm, ^{257}Fm, AND ^{259}Fm (FROM REF. 5).

have rather deformed shapes. There is much less high-energy symmetric fission than for the transition nucleus ^{257}Fm, perhaps because two magic Z=50 fragments can no longer be formed.

NEUTRON EMISSION

The total energy (Q-value) for SF appears as kinetic and excitation energy of the fragments. This excitation energy can be dissipated by neutron and gamma emission. In general, the average number of neutrons emitted per spontaneous fission event, $\bar{\nu}_t$, increases with Z and A of the fissioning nucleus as shown in Fig. 8. The exact values depend on the relative amounts of energy appearing as kinetic or excitation energy of the fragments. Neutron emission from "cold", near-spherical fragments with very high TKE due to Coulomb repulsion would be expected to be particularly low. This was verified[28] for ^{257}Fm by measuring neutron multiplicities as a function of TKE and mass split. For fissions with TKE > 240 MeV (the Q-value for symmetric fission is 253 MeV), $\bar{\nu}$ is only 1.1 with a variance of 0.7, compared to $\bar{\nu}_t$=3.8 with a variance of 2.5. The mass-yield distribution for these events is narrowly symmetric with a full-width-at-half-maximum of only about 8 mass units. This resembles the mass-yield distributions for ^{258}Fm and ^{259}Fm for which the most probable TKE's are about 235 MeV. Thus for

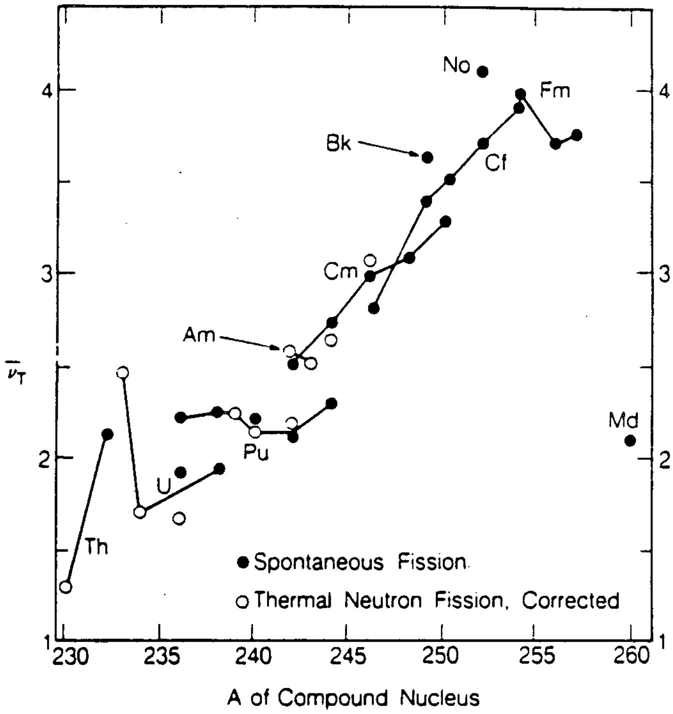

FIGURE 8 $\bar{\nu}_t$ VS. A OF THE COMPOUND NUCLEUS. MEASUREMENTS FOR THERMAL NEUTRON-INDUCED FISSION HAVE BEEN CORRECTED TO ZERO EXCITATION ENERGY USING $d\bar{\nu}_t/dE_x$ = 0.11 MeV^{-1}.

^{258}Fm, ^{259}Fm, and ^{260}Md which have narrowly symmetric mass distributions and anomalously high TKE's which approach the Q-values for fission, neutron emission from the fragments would also be expected to be quite low. Indeed, measurements by Wild et al.[29] for ^{260}Md show that $\bar{\nu}_t$ is only 2.58, (see Fig. 8), a dramatic decrease from ^{257}Fm. The variance is very large and the neutron multiplicity distribution was resolved into two components: fissions in the upper 63% of the TKE distribution gave $\bar{\nu}=1.8$ while the lower 37% gave $\bar{\nu}=3.9$. They associated these with the two Gaussian components of 235 MeV and 195 MeV, respectively, into which they resolved the TKE distribution. (See Fig. 6.) The high TKE mode gives rise to compact shapes with little excitation energy available for neutron emission, and the low TKE mode to more elongated shapes which can emit more neutrons. Again, this may be associated with the two valleys to fission found in recent calculations[14-16,18].

FUTURE

Although a remarkable amount of experimental data on spontaneous fission half-lives and properties of the heaviest elements has been amassed since the discovery of SF nearly 50 years ago, many challenges to experimentalists still remain. It appears likely that longer-lived, neutron-rich, odd-odd isotopes of the heaviest elements will have measurable SF half-lives and can be studied if methods for their production can be devised. Currently, a most promising method is the Large Einsteinium Activation Program (LEAP) proposed[30,31] by a consortium of four U.S. national laboratories. Targets of the rare 275-day ^{254}Es would be bombarded with light to medium ion projectiles to produce new neutron-rich isotopes of still higher Z elements and superheavy elements, as well as larger sources of known isotopes for more detailed study of their fission properties.

Techniques for positive assignment of the mass and atomic number of short-lived nuclides which decay predominantly by SF need to be developed. Physical techniques for performing dE/dx, TKE, and time-of-flight measurements of coincident fragments can now be envisioned. Automated chemical separation methods on the few second time scale are also now within reach and should be invaluable in separating complex mixtures of fissioning nuclides and making positive assignments of atomic number.

Severe challenges to the theorists also still exist. Although general trends in half-lives seem to be reproduced, the extreme sensitivity to details of the fission barriers and paths, and the hindrances due to odd nucleons make predictions uncertain by many orders of magnitude. A comprehensive, dynamic, theoretical model which can reproduce measured SF properties such as fragment mass, charge, and kinetic-energy distributions, neutron and gamma emission, and the dramatic changes in properties which occur with the change of only a nucleon or two, as well as predict the properties of as yet unknown nuclides, still does not exist. A fundamental understanding of spontaneous fission, which is the process that will ultimately limit the number of chemical elements that can exist, is certainly a worthy goal and one which may also provide insights into the optimum synthesis methods for producing still heavier elements.

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