Spontaneous fission

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Abstract

The spontaneous fission (SF) of the heaviest actinides and the transactinides is of particular interest, because of the dramatic changes in properties observed in the region of the heavy fermium isotopes and for still heavier elements. The existing experimental information on SF properties, including half-life systematics, fragment kinetic energy and mass yield distributions, prompt neutron emission and gamma emission, will be reviewed. Possibilities for extending studies of SF properties to other regions are considered, and the potential for obtaining additional information about low-energy fission properties is discussed.

1. Introduction

It seems appropriate, nearly five years after the conferences in Berlin and Washington, DC, where we commemorated the 50th anniversary of the discovery of fission (1938–39), and those commemorating the discovery of spontaneous fission (SF) in 1940, that we should review our progress since that time. Do we still perceive SF as the ultimate limit to nuclear stability at the upper reaches of the chart of the nuclides? Have the predictions of a deformed shell in the region of 162–164 neutrons, which might help to stabilize nuclei towards decay by SF, been confirmed? Have we been able to devise new production methods which will allow us to access the neutron-rich regions of the heaviest elements and the transactinide elements? Have new neutron-rich target materials become available? Have new international collaborations been forged to help us overcome the difficulties in obtaining the resources required to carry on research in the field of the heaviest actinides and transactinides? Have we been able to convince our respective funding agencies that research on the nuclear and chemical properties of the heaviest elements is a truly frontier area, and that such research should be supported at least as well as research in high energy physics at the SSC or the investigations of molecular interactions and surfaces at advanced light sources? After nearly 55 years, do we now understand the fission process and its extreme sensitivity to changes in nuclear structure of only one or two neutrons or protons? Do we understand the sensitivity to shell effects of million electron volts (MeV) or less out of the total energy equivalent of a heavy nucleus of more than 250 000 MeV? Do we understand why a single odd neutron or proton can hinder SF decay (lengthen SF half-lives) by many orders of magnitude compared with even–even neighbors? Is this research which seeks to understand the limits of nuclear stability, the nuances of nuclear structure and the ultimate limit to how many chemical elements there can be, not worthy of our best efforts and the commitment of significant resources?

In an attempt to assess how close we are to finding the answers to the above questions, I have tried to survey our progress and summarize the present state of our knowledge about SF. I find the results both exciting and optimistic. However, with regard to the question concerning funding and the resources available from our respective funding agencies, I am afraid I do not find such optimistic answers.

In this paper, I will summarize the new information about SF obtained since my reviews [1, 2] presented in the spring of 1989 at the conferences commemorating the 50th anniversary of the discovery of fission, and the more comprehensive review on SF published [3] by Somerville and myself in 1989. Surprisingly, in the less than five years since then, a considerable amount of new experimental and theoretical research relevant to SF has been reported. However, because of the constraints of both time and space, I will concentrate primarily on the trans-Bk isotopes.

2. Half-lives

We have recently completed an update [4] to ref. 3 and tabulated the 120 SF half-lives (branches) which had been reported in the literature through mid-1992;

70 of these half-lives were revised or new values. Figure 1 shows the logarithm of the known SF half-lives of even–even (e-e) nuclei plotted *vs.* the neutron number *N*. (Recent results of Lazarev *et al.* [5] for Cf isotopes have been added to the plot.) As has been noted earlier, the strong stabilizing effect of the $N = 152$ deformed shell seen in Cf, Fm and No appears to have disappeared in element 104.

Smolanczuk *et al.* [6] performed a theoretical study of the dependence of the alpha and SF half-lives for element 104 *vs.* even neutron number. They found that the SF lifetimes are smaller than the alpha decay lifetimes for all the even neutrons studied, ranging from one order of magnitude smaller for $N = 154$, to about seven orders of magnitude smaller for $N = 142$ and for $N = 166$, the heaviest isotope studied. Their results are close to those determined experimentally. Only one experimental SF half-life value for element 106 and only a lower limit value for element 108 are available.

We have just heard from Lougheed [7] at this conference about the production of the new isotopes [265]106 and [266]106, but only estimates of the half-lives based on calculations from the experimentally determined alpha decay energies are currently available. However, the resulting estimated half-lives of the order of seconds to half a minute for both isotopes are exciting and certainly rather unexpected for the e-e isotope [266]106. In fact, since these 106 isotopes are believed to decay primarily by alpha emission, these are lower limits for the SF partial half-lives. The results imply that the SF
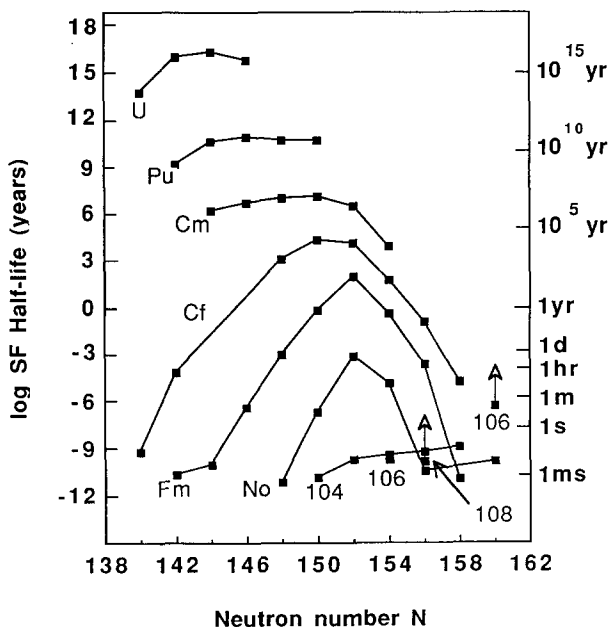
half-lives are longer for the very heavy e-e isotopes than we had dared to hope, even considering the predictions of the stabilizing effect of a deformed shell in the region of $N = 162$–164 [8, 9]. The SF half-life of 20–30 seconds would certainly indicate a very strong effect from this shell, as is shown in Fig. 1. Some hint of this may have been seen for [260]No and [262]No, although the assignment of the 100 ms activity to [260]No is somewhat uncertain.

If we make a similar plot for isotopes with an odd number of neutrons or protons (e-o, o-e) and with both particles odd (o-o), we see the immense variation in SF half-life resulting from the effect of the odd particles (Fig. 2). This illustrates the difficulty in making predictions based on the experimental systematics. This is also a difficult problem theoretically, but Möller and Nix [10] have calculated half-lives for nuclides with $Z = 101$, 103, 105, 107 and 109, and $N = 155$, 157, 159, 161 and 163. These values clearly show the added stability toward SF decay associated with odd nucleons. For example, they calculated that element 109 with 163 neutrons has an SF half-life of 10^{15} years or more!

In an attempt to assess the hindrances associated with the odd nucleons, the logarithm of the experimentally determined SF hindrance factor (HF) as a function of the proton and neutron numbers is plotted in Fig. 3. The HF for the SF decay of an odd proton
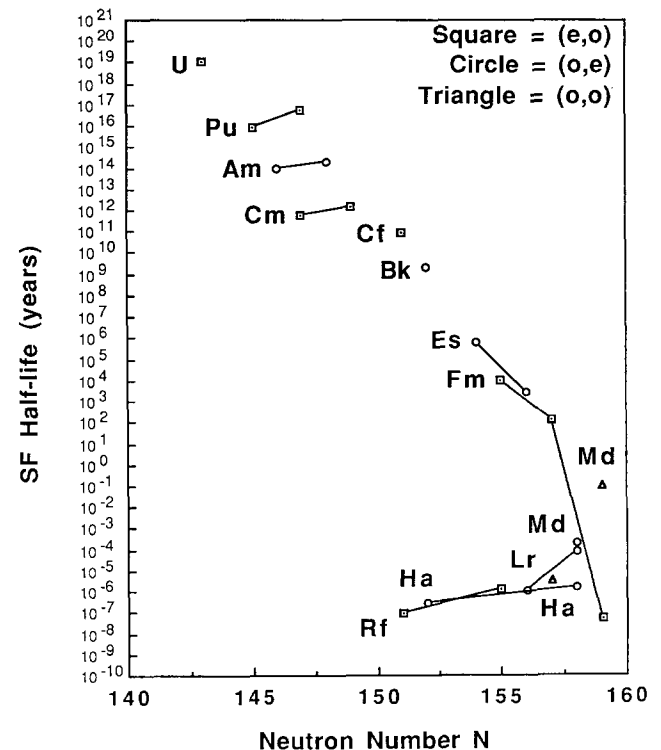

Fig. 1. Logarithms of SF half-lives of e-e nuclei plotted *vs.* neutron number. Arrows are used to indicate lower limits for [264]108 and the recently reported [5, 7] [266]106.

Fig. 2. Logarithms of SF half-lives of e-o (□), o-e (○), and o-o (△) nuclei plotted *vs.* neutron number. (Lower limit values have not been included.)
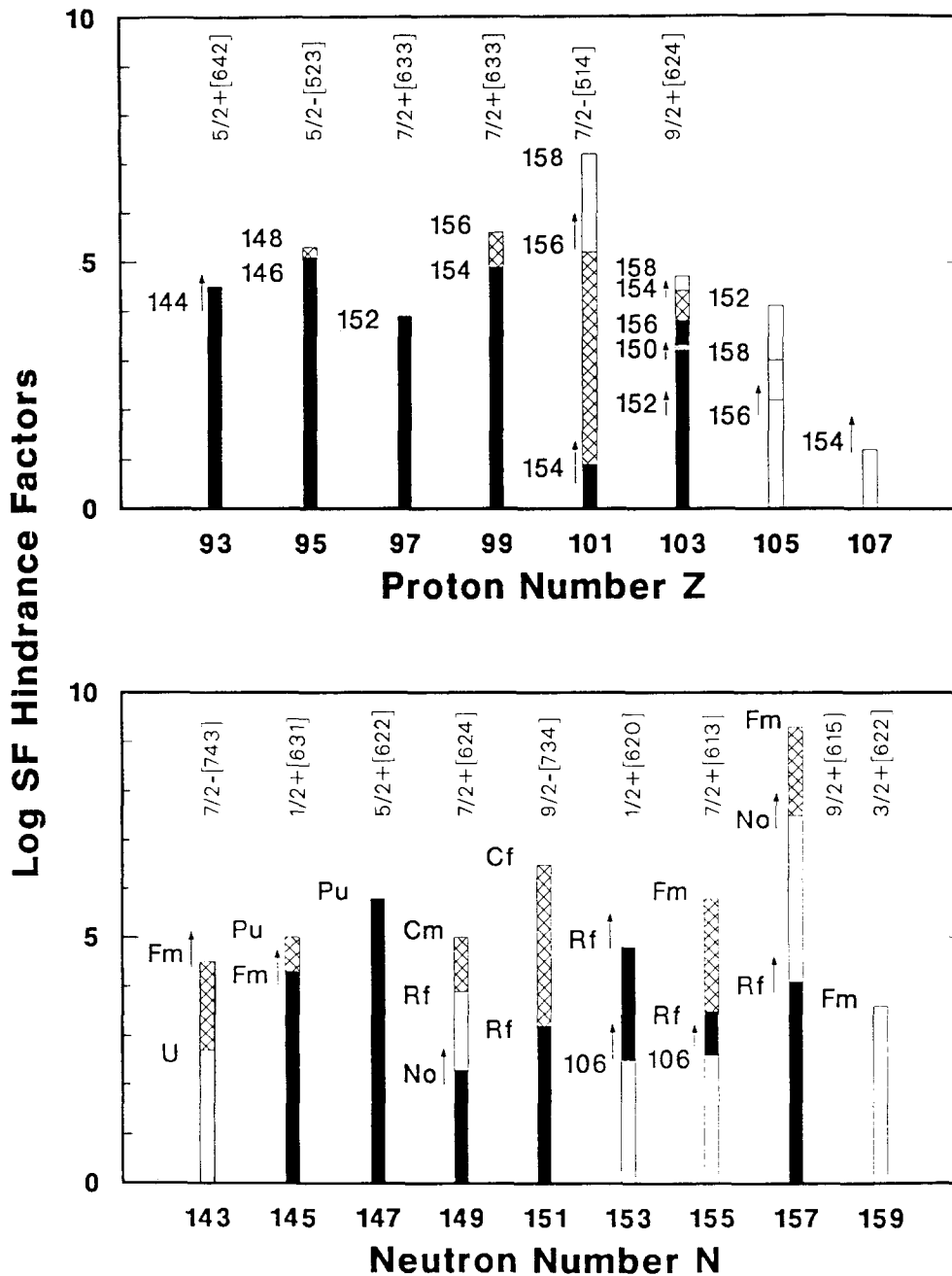
Fig. 3. Logarithms of HF for SF of odd neutron and odd proton nuclides plotted vs. neutron number N and proton number Z respectively. Open bars indicate that the HF was calculated relative to only one e-e neighbor. A filled or hatched bar indicates that the HF was calculated relative to two e-e neighbors.

or an odd neutron nuclide is calculated relative to the geometric mean of the SF half-lives of the two adjacent e-e neighbors, as in ref. 1. In cases where the half-life of only one e-e neighbor is known, that value is used in the calculation. It appears that the HFs are about 10^5 for nuclides with either an odd neutron or an odd proton, where actual measurements and not just limit values exist. The 157th neutron seems to lend special stability to elements 100, 102 and 104, and it would

be interesting to see if this also holds true for element 106.

The HFs for o-o nuclei are expected to be even larger, perhaps comparable with the sum of the log HFs for the two odd particles, or of the order of log $HF = 10$. Using the recently determined values of the SF half-lives for ^{258}Md, ^{260}Md and ^{262}Lr of more than 5×10^3 years, 28–38 days and more than 1.5 days, respectively, I have calculated their log HFs relative

to their corresponding two pairs of e-e neighbors, as follows:

(1) log HFs of over 9.1 and over 10.4 were calculated for ^{258}Md(101) relative to its pairs of e-e neighbors ^{256}Fm(100)–^{258}Fm and ^{258}No(102)–^{260}No;

(2) log HFs of 9.3 and 8.1 were calculated for ^{260}Md relative to ^{258}Fm–^{260}Fm and ^{260}No–^{262}No;

(3) log HFs of over 6.8 and over 6.4 were obtained for ^{262}Lr(103) relative to ^{260}No–^{262}No and ^{262}Rf(104)–^{264}Rf.

These results lend credence to the expectation of very high HFs for o-o nuclides. However, in the case of ^{258}Lr, a lower limit value of only 78 s has been reported, which is smaller than the 550 s SF half-life reported for its e-e neighbor ^{256}No. It is difficult to assess whether this results from the stabilizing effect of the $N = 152$ deformed shell on the No half-lives or from our inability to set a more sensitive limit on the SF half-life for ^{258}Lr.

The log HF calculated relative to the e-e Rf neighbors of ^{258}Lr is more than 3.7 — not a very useful lower limit. In fact, ^{258}Lr is quite likely to have an electron capture (EC) decay branch to the 1.2 ms ^{258}No, and the SF activity observed in studies of the decay of ^{258}Lr could well result from ^{258}No. Similarly, the SF branch observed in ^{262}Ha, which results in a partial half-life for SF of 100 s and a log HF of 3.5, probably also results primarily from SF from its EC daughter, 50 ms ^{262}Rf. Clearly, experimental investigations are needed to determine whether or not this is the case, and to measure the EC branching ratios.

3. Properties of the fission fragments

Since the 1989 reviews, only very few measurements of kinetic energy and mass distributions for additional trans-Bk isotopes have been reported. Among these are ^{256}No [11] and the odd-Z nucleus ^{259}Lr [12]. These are of considerable interest, because they appear to be "transition" nuclides $i.e.$, they show both "symmetric" and "asymmetric" mass division, as shown in Fig. 4.

The most probable total kinetic energy (TKE) for ^{256}No appears to be about "normal", while that of ^{259}Lr appears to be somewhat high compared with the linear fit to $Z^2/A^{1/3}$ shown in Fig. 5. However, both TKE distributions can be fitted with a single Gaussian and show no clear evidence for another component. The ^{256}No contour plot shown in Fig. 4 exhibits a high ridge extending from symmetric mass division out to around a mass of 146, while ^{259}Lr appears to be broadly symmetric, with some events near symmetry having a higher TKE.

In both cases, the appearance of the mass-yield curves is dependent on the exact correction made for neutron
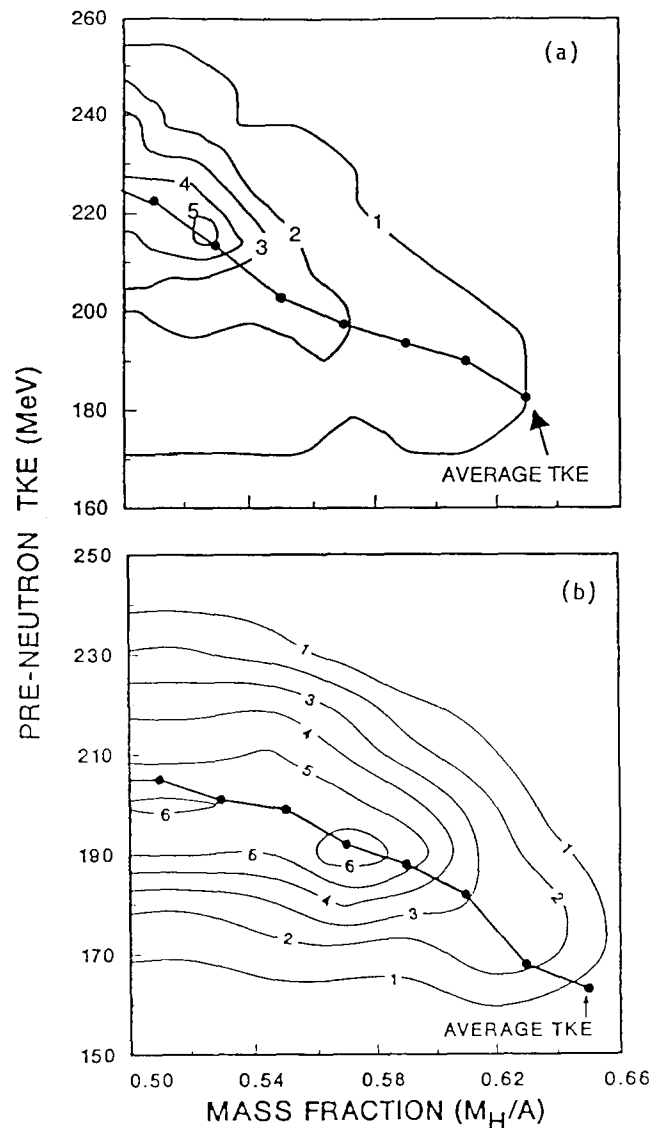

Fig. 4. Contour plots of pre-neutron emission TKE $vs.$ mass fraction for (a) ^{259}Lr and (b) ^{256}No. (a) The contours indicate equal numbers of events based on data groupings of 10 MeV × 0.02 units of mass fraction. Contours labeled 1–5 represent 10–50 events respectively [12]. (b) The contours indicate equal numbers of events based on data groupings of 20 MeV × 0.04 units of mass fraction. Contours labeled 1–6 represent 10–60 events respectively [11].

emission from the fragments, as shown in Fig. 6 for ^{259}Lr. Unfortunately, no information on neutron emission as a function of fragment mass is available for these nuclides, and one is forced either to calculate provisional mass yield curves with no correction for neutron emission or to use a neutron emission function similar to that measured for ^{252}Cf, normalized to an average total neutron emission per fission, consistent with the difference between the average Q values and average TKE. Neither procedure is very satisfactory and we are making an attempt to calculate a more
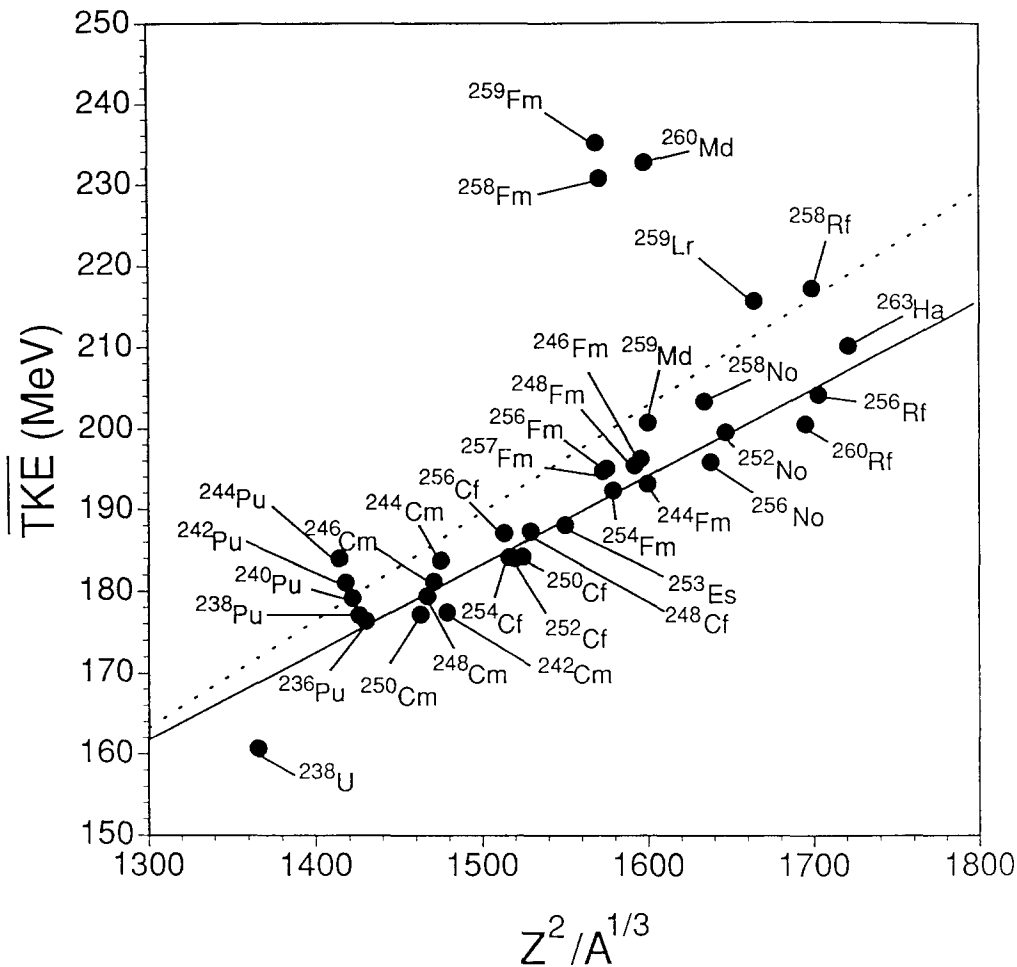
Fig. 5. Average or most probable TKE vs. $Z^2/A^{1/3}$ for SF. The dashed line is the linear fit of Unik et al. [13] and the full line is the fit of Viola [14].

realistic neutron emission function based on the scission point model of Wilkins et al. [15].

Not only are neutron emission data as a function of fragment mass unavailable but average values of neutron emission per fission have been measured for only a very few of the heaviest nuclides, even though the technology for doing so is available. A plot of the average total neutron emission per fission as a function of the mass of the spontaneously fissioning nucleus is given in Fig. 7. The value of 4.2 [16] measured for ^{259}Md is the same as that for ^{252}No, and reflects their "normal" to rather "low" TKEs, as shown in Fig. 5. ^{260}Md is the heaviest nuclide measured and shows a dramatic decrease in average neutron emission, consistent with its very high TKE.

A schematic representation of the measured mass yield curves for the trans-Bk isotopes is given in Fig. 8. As can be seen, relatively few measurements have been made for odd proton nuclei, and it is especially important to try to measure some of these for the elements beyond Fm. I have removed ^{262}Ha from my plot [1], because it is not clear from the existing data

whether its mass distribution is asymmetric or broadly symmetric.

No information on nuclear charge division in SF exists for the trans-Bk isotopes, except for ^{252}Cf, which has been studied in considerable detail. Indeed, ^{252}Cf has been evaluated by Wahl [17] together with data for thermal neutron-induced fission of ^{235}U, ^{233}U and ^{239}Pu, in order to derive elemental yields as a function of Z.

The excitation energy of the fission fragments can be dissipated by the emission of gamma-rays as well as neutrons, but this has been even less well studied than has neutron emission. No studies of gamma-ray emission from fragments from SF of the trans-Cf isotopes have been reported until the recent studies of Sokol et al. [18], who measured energies, intensities, and total gamma energy and average number of gamma-rays per fission for ^{256}Fm and ^{259}Md, as well as for ^{248}Cm and ^{252,254}Cf. Although the characteristics of the prompt gamma radiation from SF of all these nuclei were similar, the number of photons per fission for ^{254}Cf and ^{259}Md (5.3) is somewhat smaller than that of 6.5–7
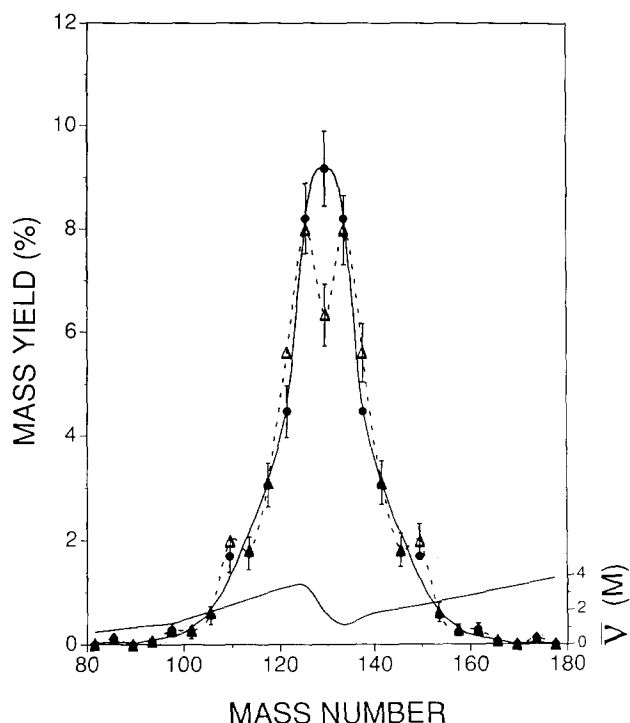
Fig. 6. Post-neutron emission (●) and pre-neutron emission (△) mass yield curves [12] for ^{259}Lr. The pre-neutron emission curve was obtained with a $\bar{\nu}(M)$ function similar to that used for ^{257}Fm, but with $\bar{\nu}_T = 4.5$.
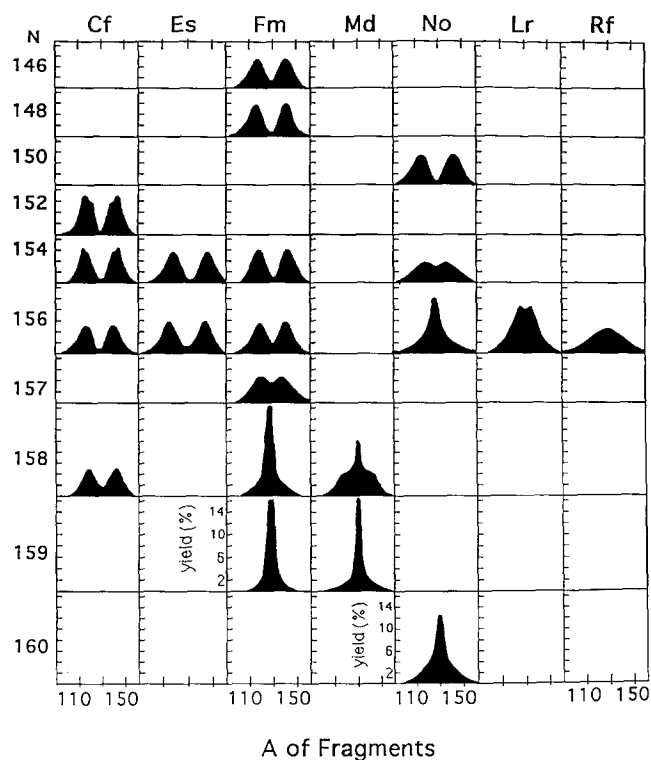

Fig. 7. Experimental values of average $\bar{\nu}_T$ per SF as a function of A of the spontaneously fissioning nucleus.

A of Fragments

Fig. 8. Schematic representation of mass yield distributions (normalized to 200% fission fragment yield) for SF of trans-Bk isotopes [4].
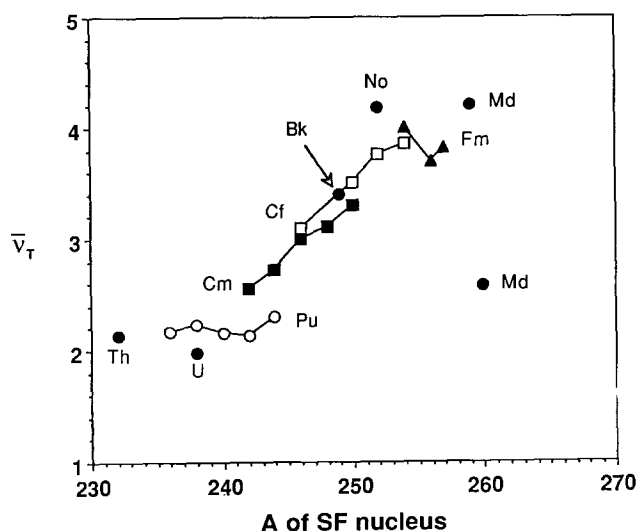
for the other nuclides. The total photon energy per fission was 6.4 MeV for ^{259}Md compared with 9.6 MeV for ^{252}Cf. Data for nuclides such as 1.6 h ^{259}Md are exceedingly difficult to obtain and only 34 SF events were recorded. Such information is extremely helpful in deducing the deformation and excitation energy of the fission fragments at scission. Hence, it is useful in inferring details about the shape of the system at scission

and the subsequent neutron emission from the fragments.

Another type of study which has considerable potential for furnishing information about the fissioning system in regions where the SF half-lives are too long for study or the nuclides are difficult to produce directly involves EC- or beta-delayed fission [19]. In the delayed fission process, the parent nucleus decays via EC or beta emission to excited states in the daughter nucleus, which then rapidly undergo fission. These fissions appear to decay with the parent half-life, which is often long enough, as in the case of ^{228}Np, ^{232}Am and ^{234}Am, to permit chemical separations and/or direct "out-of-beam" studies of the fragment properties.

Time correlations between EC and fissions can be measured in on-line experiments, yielding lifetime information about the fissioning states and the lifetime of populated shape isomers in neutron-deficient nuclei. It can be envisioned that de-excitation of levels in the second well could be observed, which would provide information about the level structure, and, hence, the deformation of the populated shape isomers. Similar studies of beta-delayed fission to furnish information for neutron-rich nuclei also should be undertaken, but these nuclides are considerably more difficult to produce.

4. Future prospects

There appear to be indications that SF half-lives for the isotopes of elements heavier than 104 may not be decreasing as rapidly as previously thought, possibly as a consequence of the stabilizing effect of a deformed shell in the region of $N = 162$–164. The use of multiple-target systems of the most neutron-rich isotopes which can be made available, perhaps ^{244}Pu (8×10^7 years, $N/Z = 1.60$), ^{250}Cm ($\sim 10^4$ years, $N/Z = 1.60$) or even ^{254}Es (275 days, $N/Z = 1.57$), together with high-intensity, neutron-rich heavy ion beams (or exotic radioactive beams) should permit us to produce neutron-rich isotopes of these elements for measurement of half-lives, alpha decay energies, fragment kinetic energies and mass distributions in some cases.

The technology exists currently to make measurements of neutron and gamma emission from the fission fragments, and such studies are extremely important in understanding details of the fission process. However, a considerable commitment of time and resources will be required. Efficient, high-resolution techniques for positive assignment of the mass and atomic numbers of SF nuclides which are too short-lived to permit chemical separation and identification still need more development. Chemical separation and studies of element 106 now appear possible, while heavy, odd neutron isotopes of elements 107–109 may well have half-lives long enough for chemical separation as well as for on-line studies of the SF properties. There are also many challenging problems for the theorists, not the least of which is the development of a single, comprehensive, dynamic model which can describe accurately the fissioning system as it evolves from a single nuclear system into two (or possibly more) separated fragments. To take advantage of all the exciting opportunities and meet all these challenges, it may be an opportune time to pool our resources and organize an international collaborative effort commensurate with the magnitude and importance of these problems.

Acknowledgments.

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