Spontaneous-Fission Properties and Production of Heavy-Element Isotopes†,‡

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Spontaneous fission was discovered[1] in ^{238}U as a natural mode of decay as long ago as 1940. However, because of the long spontaneous-fission half-life of ^{238}U of about 10^{16} years, the decay rate (specific activity) was so low that detailed studies of the properties of spontaneous fission had to await the synthesis of isotopes of higher Z elements with shorter spontaneous-fission half-lives. During the 1960s, milligram quantities of ^{252}Cf became available to researchers through the Transplutonium Production Program of the U.S. Atomic Energy Commission. The availability of ^{252}Cf with a spontaneous-fission half-life of 85 years, and its resultant high specific activity, stimulated many pioneering studies of the spontaneous-fission process. Detailed measurements of the mass, charge, and kinetic-energy distributions of the fission fragments, of prompt neutron and photon emission from the fragments, and the interrelationship of these properties were reported by numerous investigators.[2] Since that time, studies of the spontaneous-fission properties of many other isotopes, some with half-lives of less than a second, have been made.

Spontaneous-Fission Properties

Spontaneous fission (SF) only occurs in nuclides with $Z \geq 90$ where Coulomb forces make the nucleus unstable toward this mode of decay. The half-lives for spontaneous fission generally decrease with increasing Z for a given number of neutrons, although the half-lives tend to be longer for isotopes with an odd proton or neutron. In order to obtain a complete understanding of the spontaneous fission process, a knowledge of the mass, atomic number, kinetic energy, and deformation energy of the fragments at scission, as well as their deexcitation modes, is necessary. In addition, we need to know how these properties change with the mass and atomic number of the fissioning nucleus.

The total energy released in binary fission, E_f, is just the energy equivalent of the difference in mass of the fissioning nucleus, M_f, and the masses of the two resulting fission fragments, M_1 and M_2: $E_f = \Delta M = M_f - (M_1 + M_2)$. The energy is divided between the kinetic energy, E_k, of the fragments prior to prompt neutron emission and the excitation energy, E_X, of the fragments: $E_f = E_k + E_X$. The E_X can be inferred from measurements of neutron and γ-ray emission: $E_X = \nu_t(B_n + E_n) + E_\gamma$, where ν_t = total number of neutrons emitted, B_n = neutron binding energy, E_n = neutron kinetic energy, and E_γ = total γ-ray energy. (Average values are normally used since the actual values for each individual fission event are usually not known.) If the total kinetic energy of the fragments approaches the total energy available from fission, very little energy can reside in the excitation or deformation energy of the fragments and there is correspondingly less energy available for neutron or photon emission.

A schematic diagram of the spontaneous fission process for ^{252}Cf is shown in Figure 1. The most probable mode of mass division for this nuclide is asymmetric; i.e., two fragments of unequal mass are usually produced. Prompt neutron emission from the highly excited fragments occurs in less than 10^{-13} s after scission; emission of γ-rays, conversion electrons, and X-rays follows in 10^{-13}–10^{-7} s. β decay is somewhat slower, with the shortest half-lives being of the order of milliseconds. So-called delayed neutron emission proceeds with half-lives ranging from about 10^{-1} to 10^2 s, depending on the half-life of the β-decaying precursor.

Ideally, we would like to measure everything for each fission event, i.e., the kinetic energy, velocity, atomic number, and mass of each fragment as well as the number and energies of the prompt neutrons and photons emitted from the fragments. Probably the only spontaneously fissioning nuclide that has been studied in anything approaching this detail is ^{252}Cf. Much has been learned about the spontaneous-fission process from the study of ^{252}Cf, but the information cannot be readily extrapolated to the heavier region of the Fm and Md isotopes and beyond.

Mass Distributions

We tend to characterize fission according to the mass distribution of the fragments because this property is among the most obvious and readily measurable results of fission. Fission into two nearly equal-mass fragments is called "symmetric" while fission into two fragments of unequal mass is called "asymmetric" (Figure 1). Actually, a distribution of different mass splits around the most probable split has been found for all the cases studied so far, but the widths of these distributions can vary widely. (For example, more than 450 radioactive

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(1) Petrzhak, K. A.; Flerov, G. N. *C. R. (Dokl.) Akad. Sci. USSR* **1940**, *28*, 500; *J. Phys. USSR (Engl. Transl.)* **1940**, *3*, 275; *Phys. Rev.* **1940**, *58*, 89L.

(2) "A Review of the Accomplishments and Promise of U.S. Transplutonium Research—1940–1981"; Keller, O. L., Wymer, R. G., Eds.; published under auspices of Subcommittee on Nuclear and Radiochemistry, Committee on Chemical Sciences, Assembly of Mathematical and Physical Sciences, National Research Council; report available from Office of Chemistry and Chemical Technology, NRC, 2101 Constitution Avenue, NW, Washington, DC 20418.

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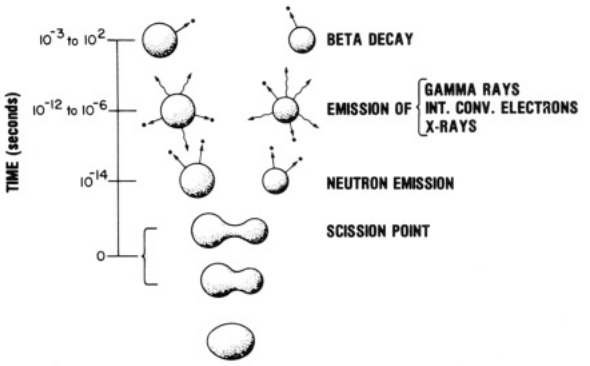

Figure 1. Schematic diagram of the spontaneous fission process for ^{252}Cf, a deformed actinide nuclide. The approximate times for deexcitation of the fission fragments are indicated.

and 100 stable fission products result from the thermal neutron-induced fission of ^{235}U.)

Fragment mass distributions can be obtained directly from radiochemical separation and measurement of as many as possible of the multitude of isotopes produced. Such radiochemical measurements have "perfect" resolution for mass and atomic number because each individual isotope is identified and measured, but the resulting information is for the fragments after they have deexcited by prompt neutron emission, photon emission, and in many cases, after β decay and delayed neutron emission. It is impossible to know exactly the deexcitation processes and therefore to deduce what the primary fragment at scission actually was. Furthermore, the method is limited to isotopes with half-lives long enough for radiochemical assay and to sources intense enough for measurement of the fission products.

In the case of the shortest lived isotopes or those available only in small quantities, the mass distributions are usually derived from the kinetic energy, E_k, of coincident fission fragments measured with solid-state detectors. From such kinetic energy data, the masses can be obtained via the conservation of momentum relationship, $M_1V_1 = M_2V_2$, by assuming the mass of the fissioning nucleus and a prompt neutron emission distribution. Substitution of $E_k = 1/2MV^2$ gives $M_1E_{k_1} = M_2E_{k_2}$, where M is mass, V is velocity, and subscripts 1 and 2 refer to fission fragments 1 and 2, respectively. The kinetic-energy measurements suffer from poorer resolution than the radiochemical methods and corrections still need to be made for neutron emission as a function of fragment mass and energy. If both the velocity and kinetic energy of each fragment can be measured, then a direct measurement of the primary fragment mass can be obtained, but this has only been done for a very few cases, e.g., ^{252}Cf.

In trying to review how our knowledge of mass distributions has progressed, it is instructive to look at the state of our knowledge some 10 years ago.[3] Much larger yields had been measured for symmetric mass division in the spontaneous fission of ^{257}Fm than those previously reported for lighter actinide isotopes. In addition, radiochemical measurements of the mass yields from the spontaneous fission of ^{256}Fm, as well as for thermal neutron-induced fission of ^{255}Fm, (^{256}Fm*), showed significantly increased yields for symmetric mass division. The most probable mass division for the thermal

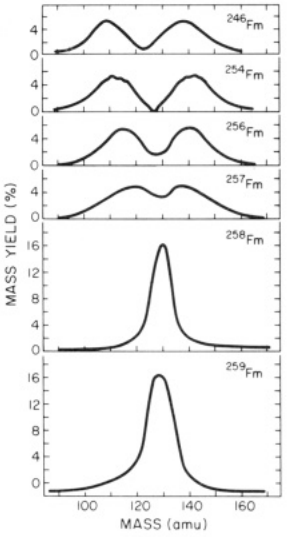

Figure 2. Mass-yield distributions for Fm isotopes.

neutron-induced fission of ^{257}Fm, (^{258}Fm*), had been found to be symmetric although the mass distribution was very broad. Prior to these studies it was generally believed that all spontaneous and thermal neutron-induced (low-energy) fission resulted in highly asymmetric mass division. The new data indicating rapid and unexpected changes in the properties of spontaneous and low-energy fission in the region of the heavy fermium isotopes sparked what might be called a "renaissance" of interest in the fission process and stimulated additional investigations. It was postulated[4] that the observed increase in symmetric mass division with increasing mass of the fermium isotopes is because the heavier Fm isotopes ($Z = 100$) can fission symmetrically into two fragments both with nearly the doubly closed shell ^{132}Sn ($Z = 50$; $N = 82$) configuration, which is expected to be spherical. If so, then departure from this region should result in a return to asymmetric mass division.

The mass-yield distributions for spontaneous fission of seven Fm isotopes have now been measured,[5-9] and some of these are shown schematically in Figure 2. It illustrates the asymmetric mass division observed for the light Fm isotopes, the enhanced yields at symmetry for ^{257}Fm, and the abrupt change to very narrow, symmetric mass distributions for ^{258}Fm and ^{259}Fm. Mass-yield distributions for spontaneous fission of several Cf ($Z = 98$) isotopes have also been measured.[5] Even ^{256}Cf, which has the same number of neutrons as ^{258}Fm, shows only slightly enhanced yields for symmetric mass division. Apparently, the fact that Cf has only 98 protons and cannot divide symmetrically into two closed-shell, $Z = 50$ fragments, as can Fm, has a profound influence on the mass division.

(3) Hoffman, D. C.; Hoffman, M. M. *Annu. Rev. Nucl. Sci.* **1974**, *24*, 151.

(4) Hoffman, D. C. Proceedings of the 3rd International Conference on Nuclei Far from Stability, Cargèse, Corsica, CERN 76-13, Geneva, 1976; p 565.

(5) Hoffman, D. C. "Physics and Chemistry of Fission 1979"; International Atomic Energy Agency; Vienna, 1980; Vol. 2, pp 275–297.

(6) Hoffman, D. C.; Lee, D.; Ghiorso, A.; Nurmia, M.; Aleklett, K. *Phys. Rev. C* **1980**, *22*, 1581.

(7) Hulet, E. K.; Lougheed, R. W.; Landrum, J. H.; Wild, J. F.; Hoffman, D. C.; Weber, J.; Wilhelmy, J. B. *Phys. Rev. C* **1980**, *21*, 966.

(8) Hoffman, D. C.; Wilhelmy, J. B.; Weber, J.; Daniels, W. R.; Hulet, E. K.; Lougheed, R. W.; Landrum, J. H.; Wild, J. F.; Dupzyk, R. J. *Phys. Rev. C* **1980**, *21*, 972.

(9) Hoffman, D. C.; Lee, D.; Ghiorso, A.; Nurmia, M. J.; Aleklett, K.; Leino, M. *Phys. Rev. C* **1981**, *24*, 495.

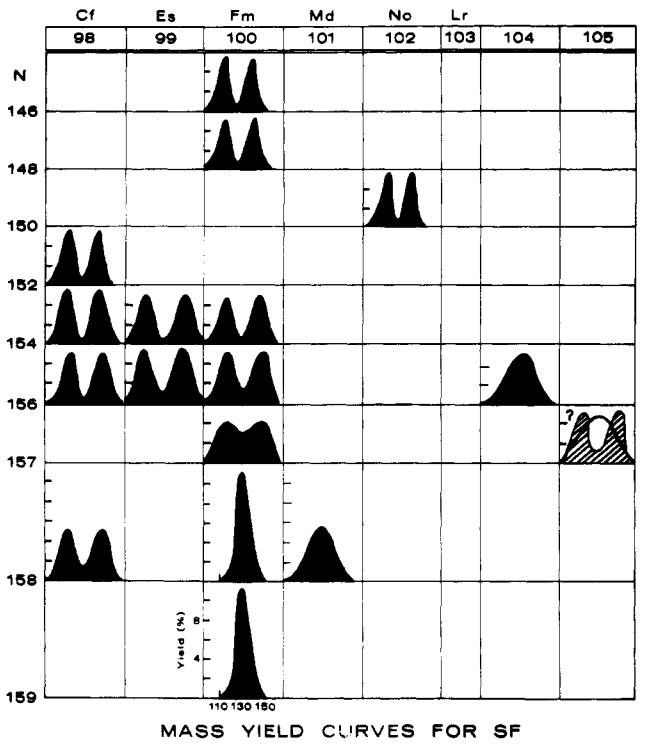

Figure 3. Schematic representation of mass-yield distributions for SF of trans-Bk isotopes.

The mass distribution for ^{259}Md has now been measured,[10] and although it is symmetric, it shows a much broader distribution than does ^{259}Fm. Measurements for 20-ms ^{260}104 recently reported by Hulet et al.[11] show a mass distribution more like that of ^{259}Md than ^{259}Fm. Earlier measurements by Bemis et al.[12] of the mass distribution for ^{262}105 were inconclusive because of interference from spontaneous fission of ^{256}Fm but indicated that if the mass distribution were symmetric it would have to have a full-width-at-half-maximum (FWHM) of more than 47 μm while the FWHM's for ^{258}Fm and ^{259}Fm are only of the order of 10 μm. They concluded that the mass division was most probably asymmetric. However, it now seems possible that a very broad distribution similar to those observed for ^{259}Md and ^{260}104 might also be consistent with their data. Measured[3,5-15] mass distributions from spontaneous fission of the trans-Bk isotopes are shown schematically in Figure 3.

Kinetic-Energy Distributions

In addition to exhibiting predominantly symmetric mass division, the fragments from SF of ^{258}Fm and ^{259}Fm have a markedly higher total kinetic energy than do any of the other isotopes studied. Although ^{259}Md and ^{260}104 also show symmetric mass distributions, they

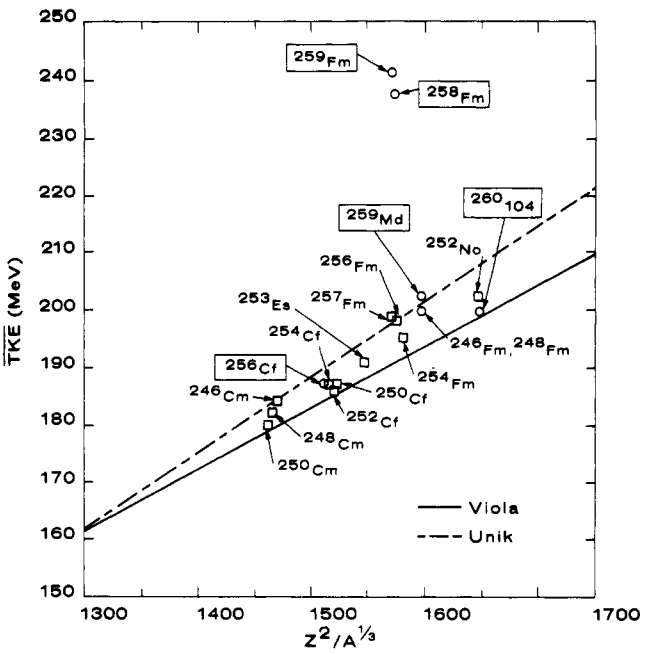

Figure 4. Average total kinetic energy, $\overline{\text{TKE}}$, as function of $Z^2/A^{1/3}$ for heavy actinide isotopes. Solid line is linear fit of Viola.[16] Dashed line is from Unik et al.[14] Data for ^{258}Fm, ^{259}Fm, ^{259}Md, and ^{260}104 are most probable TKE's.

Table I
Summary of Data for Mass and Kinetic-Energy Distributions from SF of Some Trans-Bk Isotopes (Data from Ref 3, 5-15)

nuclide	SF $T_{1/2}$, s	peak-to-valley ratio[a]	pre-neutron TKE,[b] MeV	FWHM of TKE
^{250}Cf	5.4×10^{11}	⩾300 (RC)	187.0	27
^{252}Cf	2.7×10^{9}	⩾750 (RC)	185.7	27
^{254}Cf	5.2×10^{6}	⩾145 (RC)	186.9	28
^{256}Cf	7.4×10^{2}	asymm (SS)	189.8	34
^{253}Es	2.0×10^{13}	326 (RC)	191	31
^{246}Fm	1.2	asymm (SS)	199	35
^{248}Fm	38	asymm (SS)	198	34
^{254}Fm	2.0×10^{7}	60 (RC)	195.1	27
^{256}Fm	1.0×10^{4}	12 (SS)	197.9	34
^{257}Fm	4.1×10^{9}	≈1.5 (SS)	197.6	36
^{258}Fm	3.8×10^{-4}	symm (SS), FWHM = 8	238*	33
^{259}Fm	1.5	symm (SS), FWHM = 12	238*[c]	49
^{259}Md	5.7×10^{3}	symm (SS), FWHM = 28	201*	61
^{252}No	8.6	asym (SS)	202.4	36
^{260}104	2×10^{-2}	symm (SS), FWHM = 35	199*	35
^{262}105	44	asymm (SS) or symm, FWHM ⩾ 47		

[a] Peak-to-valley ratios from radiochemical (RC) or solid-state (SS) kinetic energy measurements of the mass distributions. [b] These are average values of the preneutron emission TKE's except for those designated by an asterisk, which are most probable values. [c] Average of values in ref 7 and 9.

are very broad and the average total kinetic energies are "normal" as shown in Figure 4. A summary of the data for the mass and kinetic-energy distributions for the trans-Bk isotopes is given in Table I.

A plot of average total kinetic energy, $\overline{\text{TKE}}$, as a function of mass fraction (Figure 5) shows the exceedingly high total kinetic energy measured for symmetric

(10) Wild, J. F.; Hulet, E. K.; Lougheed, R. W.; Baisden, P. A.; Landrum, J. H.; Dougan, R. J.; Mustafa, M. G. *Phys. Rev. C* **1982**, *26*, 1531.
(11) Hulet, E. K. "Abstracts of Papers", 185th National Meeting of the American Chemical Society, Seattle, WA, March 1983, American Chemical Society: Washington, DC, 1983; NUCL 4; Lawrence Livermore Laboratory, Preprint UCRL-88414.
(12) Bemis, C. E., Jr.; Ferguson, R. L.; Plasil, F.; Silva, R. J.; Hulet, E. K.; Lougheed, R. W. *Phys. Rev. Lett.* **1977**, *39*, 1246.
(13) Balagna, J. P.; Ford, G. P.; Hoffman, D. C.; Knight, J. D. *Phys. Rev. Lett.* **1971**, *26*, 145.
(14) Unik, J. P.; Gindler, J. E.; Glendenin, L. E.; Flynn, K. F.; Gorski, A.; Sjoblom, R. K. *Phys. Chem. Fission, Proc. IAEA Symp., 3rd, 1973* **1974**, *2*, 19.
(15) Bemis, C. E., Jr.; Ferguson, R. L.; Plasil, F.; Silva, R. J.; Pleasonton, F.; Hahn, R. L. *Phys. Rev. C* **1977**, *15*, 705.

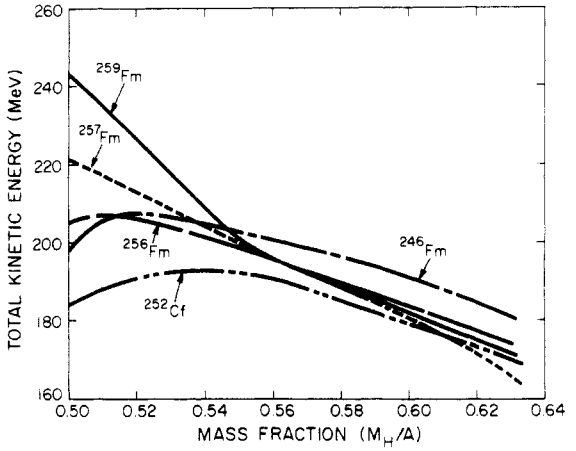

Figure 5. $\overline{\text{TKE}}$ vs. mass fraction (M_H/A) for some Fm isotopes and ^{252}Cf. (Data from ref 6–9, 13, 14.) M_H/A = mass of heavy fragment over mass of fissioning nucleus.

mass division ($M_H/A \approx 0.5$) of ^{259}Fm compared to some other Fm isotopes and ^{252}Cf. It should be noted that the average total kinetic energy for ^{259}Fm (and ^{258}Fm) for symmetric mass division approaches the total energy available from fission. Although the dip in average total kinetic energy near symmetric mass division observed for spontaneous fission of ^{252}Cf and the lighter actinides has disappeared for ^{256}Cf (N = 158), as it has for the heavier Fm isotopes, the value of $\overline{\text{TKE}}$ is only about 200 MeV for symmetric mass division of ^{256}Cf compared to around 240 MeV for ^{258}Fm and ^{259}Fm. In going to ^{259}Md, an addition of only a single proton to ^{258}Fm, the $\overline{\text{TKE}}$ is again only a little over 200 MeV for symmetric mass division.[10]

Contour plots of TKE and $\overline{\text{TKE}}$ as a function of mass fraction are shown in Figure 6 for ^{256}Fm, ^{257}Fm, and ^{259}Fm. It can be seen quite clearly that the regions of highest intensity are at asymmetric mass division ($M_H/A \geq 0.54$) for ^{256}Fm and ^{257}Fm but are near mass symmetry $M_H/A \approx 0.5$) for ^{259}Fm. In contrast to ^{256}Fm, the contour plot for ^{257}Fm shows a rather large number of events near mass symmetry. There is also a very large spread in total kinetic energy at mass symmetry, indicating that the fragments may have both spherical shapes, leading to very high total kinetic energy, and ellipsoidal shapes, leading to much lower total kinetic energy due to Coulomb repulsion. Thus, ^{257}Fm appears to be in a transition region.[13]

Both the very high total kinetic energy and the narrow symmetric mass distributions for ^{258}Fm and ^{259}Fm can be explained on the basis of symmetric mass division into two near-spherical fragments for which the Coulomb repulsion is near maximum, resulting in very high total kinetic energies. On the other hand, the near "normal" total kinetic energies for ^{259}Md and ^{260}104 (Figure 4) indicate that for some reason these fragments no longer have compact, spherical shapes at scission. This may be because in symmetric division of Md (Z = 101) one of the fragments must have 51 protons, no longer a closed proton shell.

Neutron Emission

In general, the average number of neutrons emitted per fission, $\bar{\nu}_t$, from spontaneous and low-energy fission increases with the mass and atomic number of the fissioning nucleus as shown in Figure 7. However, the

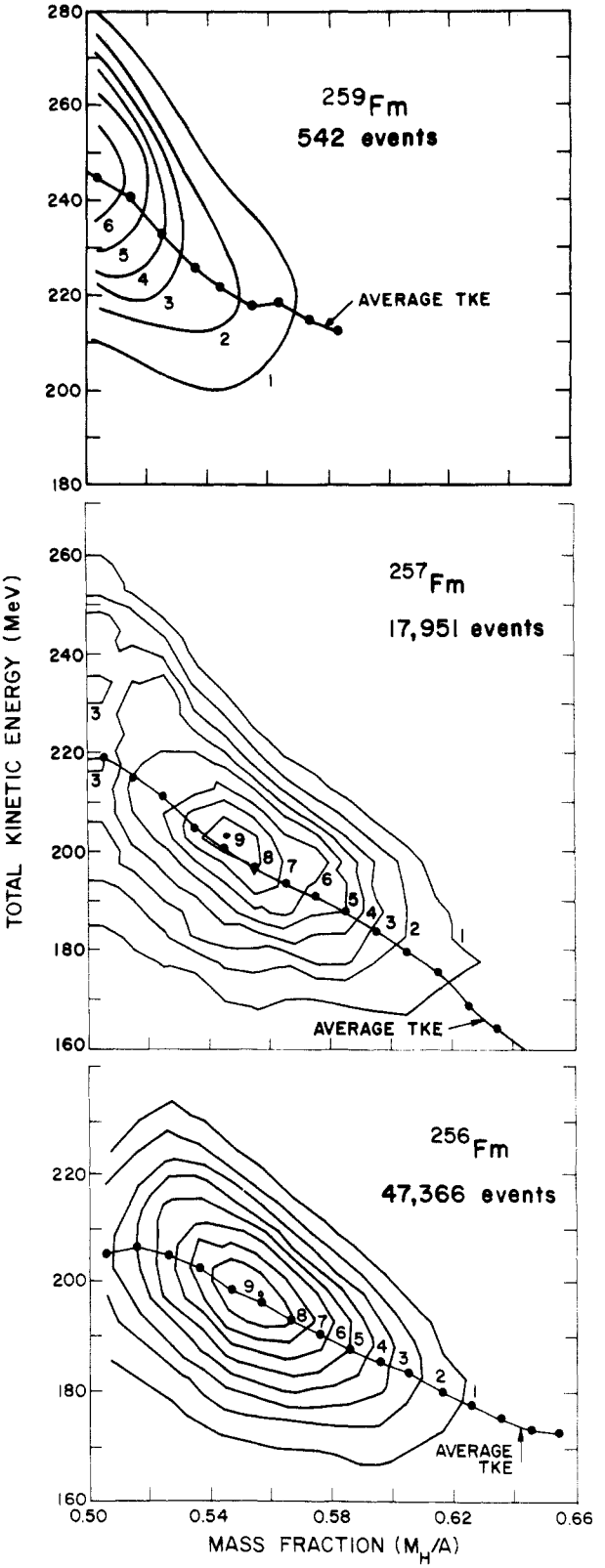

Figure 6. Contour plots of TKE and $\overline{\text{TKE}}$ vs. mass fraction for ^{256}Fm, ^{257}Fm, and ^{259}Fm. (Data from ref 7, 9, 13.)

$\bar{\nu}_t$ of 3.8 for ^{257}Fm does not follow this trend but is less than for lighter mass Fm isotopes and approximately the same as for ^{252}Cf. Measurements[17] of $\bar{\nu}_t$ as a function of total kinetic energy for ^{257}Fm show that it decreases with increasing total kinetic energy and is only 1.1 for

(16) Viola, V. *Nucl. Data, Sect. B* **1966**, *1*, 391.
(17) Hoffman, D. C.; Ford, G. P.; Balagna, J. P.; Veeser, L. R. *Phys. Rev. C* **1980**, *21*, 637.

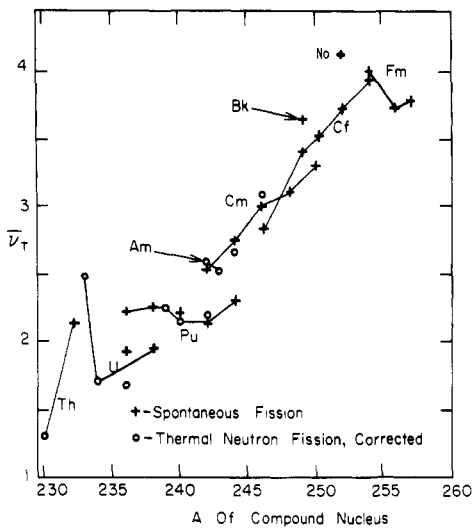

Figure 7. Experimental values of $\bar{\nu}_t$ as a function of A of the compound nucleus. Data for spontaneous fission are shown by plus signs. Measurements for $\bar{\nu}_t$ for thermal neutron-induced fission have been corrected to zero excitation energy, using $d\bar{\nu}_t/dE_x = 0.11$ MeV^{-1}, and are shown by open circles. (Reference 5.)

the ≈4% of the fissions with TKE > 240 MeV. In the case of ^{252}Cf and ^{256}Fm, there are no fissions with TKE > 240 MeV and $\bar{\nu}_t$ is 1.9 for ^{252}Cf for the ≈2% of the events with TKE > 210 MeV and 1.9 for ^{256}Fm for the ≈5% of the events with TKE > 220 MeV. These observations[17] confirm that as the total kinetic energy increases toward the total energy available from fission there is correspondingly less energy available for fragment excitation energy and for subsequent neutron (and photon) emission from the fragments. The $\bar{\nu}_t$'s for ^{258}Fm and ^{259}Fm, whose most probable TKE's are 238 MeV (Table I), should be relatively low. A value of about 1 can be predicted based on the $\bar{\nu}_t$ measured for the events from ^{257}Fm with TKE > 240 MeV. This would indicate a dramatic change from the trend shown in Figure 7. However, no data on neutron emission are yet available for these heavy nuclides.

Comparison With Theory

The high total kinetic energy events observed in ^{257}Fm, ^{258}Fm, and ^{259}Fm seem to be associated with approach of the fragments from symmetric mass division to the spherical, doubly magic ^{132}Sn configuration with $Z = 50$ and $N = 82$ closed shells. Such spherical fragments will have the maximum kinetic energy from Coulomb repulsion and little internal excitation energy, resulting in low neutron and photon emission. All these effects can be attributed to the fragment shells. But what is the explanation for ^{259}Md and ^{260}104 that exhibit broadly symmetric mass distributions with "normal" average total kinetic energies but with large half-widths? Why are there sudden changes in properties for the same neutron number in going from Cf ($Z = 98$) to Fm ($Z = 100$) to Md ($Z = 101$)? Is fission an "adiabatic process" in which fragment shell effects dominate and determine the course of fission? If so, how are they suddenly "turned off" in going from ^{258}Fm and ^{259}Fm to ^{259}Md? These are a few of the questions remaining to be answered.

Classical liquid-drop model calculations describing the potential energy of the fissioning nucleus as a function of its symmetric deformation en route to fission give a reasonable description of average properties and trends, but only symmetric fission is considered and only one barrier to fission results. This model does not explain the sudden, dramatic changes in properties in the region of the Fm and Md isotopes. More recently, theoretical calculations have centered around two main approaches to the problem: (1) correlation of fission properties with the potential energy of the fissioning nucleus after the addition of shell corrections[18,19] to the liquid-drop energy, (2) correlation of fission properties with calculations of the potential energy surface in the neighborhood of scission or with the potential energies of the fragments. The first approach results in a second barrier to fission for the lighter actinides ($Z < 100$) because of the increased stability due to shell effects. The potential energy at this barrier is found to be lower[19-22] for asymmetric than for symmetric mass division. In the region of the heavier Fm isotopes this second barrier is calculated to disappear and a return to symmetric, liquid-drop type fission is postulated. However, this approach does not explain the very high total kinetic energies associated with symmetric mass division observed for heavy Fm isotopes unless spherical fragments are postulated. The experimental observations seem to be best described by the second approach exemplified by the asymmetric two-center shell model calculations of Mustafa et al.[23-25] and the scission point model of Wilkins et al.,[26] who have calculated mass, charge, and kinetic-energy distributions on the basis of the configurations and relative potential energies of two nearly touching coaxial spheroids. (For a more complete comparison of theory with experimental results, see ref 4, 5, 8, 11, and 26.) However, none of these models is able to adequately explain all of the observed phenomena, and development of a complete understanding awaits additional experimental information.

Production of New Heavy-Element Isotopes

Now let us consider how we can produce additional neutron-rich isotopes in this region for study of their SF properties. Although the phenomenon of multinucleon transfer from heavy-ion projectiles to target nuclei has been known for some time, the potential for using such reactions to produce very neutron-rich heavy-element isotopes has not been generally recognized.[27,28] However, among the products of the bombardment of ^{248}Cm with ^{18}O we recently identified[9] 1.5-s ^{259}Fm, the most neutron-rich isotope known. It was apparently produced by an effective transfer of ^{11}Be to the target. In order to help substantiate this interpretation, we radiochemically separated the individual actinides and measured[29] the yields of as many isotopes as possible.

(18) Strutinsky, V. M. *Nucl. Phys. A* **1967**, *95*, 420; **1968**, *122*, 1.
(19) Möller, P.; Nilsson, S. G. *Phys. Lett. B* **1970**, *31*, 283.
(20) Nix, J. R. Los Alamos Preprint LA-DC-72-335, 1972; *Annu. Rev. Nucl. Sci.* **1972**, *22*, 65.
(21) Nix, J. R. *Nucl. Phys. A* **1969**, *130*, 241.
(22) Tsang, C. F.; Wilhelmy, J. B. *Nucl. Phys. A* **1972**, *184*, 417.
(23) Mustafa, M. G.; Mosel, U.; Schmitt, H. W. *Phys. Rev. C* **1973**, *7*, 1519.
(24) Mustafa, M. G. *Phys. Rev. C* **1975**, *11*, 1059.
(25) Mustafa, M. G.; Ferguson, R. L. *Phys. Rev. C* **1978**, *18*, 301.
(26) Wilkins, B. D.; Steinberg, E. P.; Chasman, R. R. *Phys. Rev. C* **1976**, *14*, 1832.
(27) Hahn, R. L.; Dittner, P. F.; Toth, K. S.; Keller, O. L. *Phys. Rev. C* **1974**, *10*, 1889.
(28) Eskola, P. "Studies of Production and Decay of Some Alpha-Active Isotopes of Einsteinium, Mendelevium, Nobelium and Lawrencium"; Ph.D. Dissertation, Dept. of Physics, University of Helsinki, Report Series in Physics 1975, No. D5.

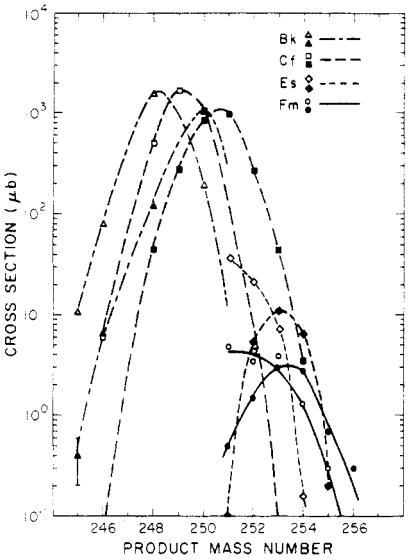

Figure 8. Measured cross sections for production of actinide isotopes from bombardment of ^{248}Cm with 97-MeV ^{18}O and 98-MeV ^{16}O, just above the Coulomb barriers of ≈94 and ≈95 MeV in the laboratory system. ^{18}O data are closed symbols; ^{16}O data are open symbols. (Reference 29.)

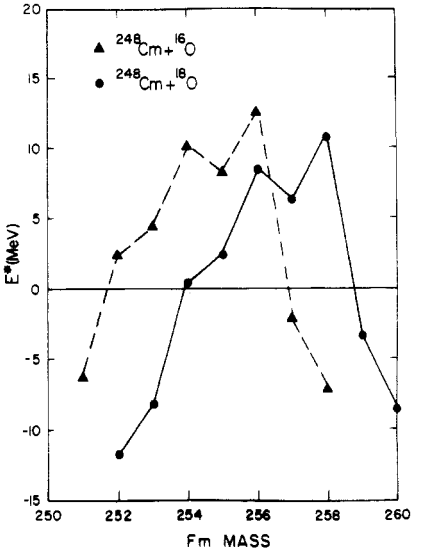

Figure 9. Calculated excitation energy, E^*, for Fm isotopes produced in reactions of ^{16}O and ^{18}O at energies near the Coulomb barrier. (Reference 30.)

We found significant yields of neutron-rich isotopes of all elements between the target and compound nucleus, ^{266}104, except for element 103 whose isotopes are too short to be detected by the procedure we used. A plot of the actinide yields for ^{16}O and ^{18}O reactions with ^{248}Cm is shown in Figure 8.

The excitation energy, E^*, of these actinide products can be calculated by assuming a binary transfer reaction to the heavy target from a projectile whose kinetic energy is approximately equivalent to the Coulomb barrier. The E^* is then essentially the difference between the energies of the Coulomb barriers of the initial target and projectile system and the final actinide product and remaining projectile-like fragment, plus the ground-state Q value. (The ground-state Q value is the energy equivalent of the difference in the masses of the products of the reaction and the masses of the initial projectile and target.) Tables of excitation energies have been calculated[30] for actinide products from a variety of actinide targets and light heavy-ion projectiles. The calculated[30] E^*'s for Fm isotopes formed in reactions of ^{248}Cm with ^{16}O and ^{18}O ions at energies just above the Coulomb barriers are plotted in Figure 9. Comparison of these with the experimental data shown in Figure 8 for Fm isotopes shows that the two mass unit difference in the projectiles is reflected in the E^*'s and the experimental yields, and the peak cross section occurs approximately at the lowest mass for which the E^* becomes 0 or slightly positive, i.e., about at masses 252 and 254 for ^{16}O and ^{18}O projectiles, respectively. Similar comparisons can be made for the Bk, Cf, and Es isotopes.

We have also measured[31] the production of actinides in reactions of ^{18}O with ^{248}Cm and ^{249}Cf targets as a function of projectile energy. The energy dependence is consistent in most cases with the calculations[30] of E^*.

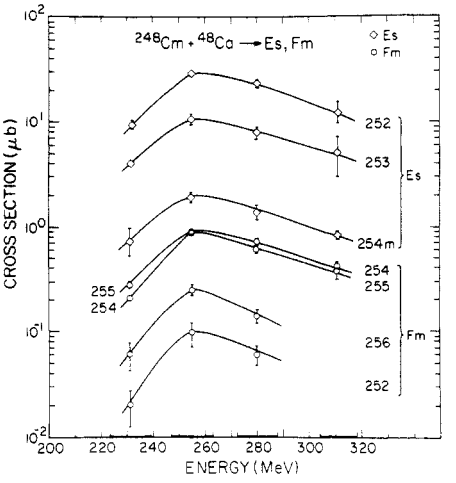

Figure 10. Cross sections as a function of average projectile energy in the laboratory system for production of Es and Fm isotopes in bombardments of ^{248}Cm with ^{48}Ca. (Reference 32.) The spread in projectile energy due to energy loss in the target is indicated on the abscissa. (The Coulomb barrier is ≈236 MeV in the laboratory system.)

Our preliminary data[32] for reactions of ^{48}Ca ions with ^{248}Cm show that the production cross sections for Cf, Es, and Fm isotopes reach a maximum some 20 MeV above the Coulomb barrier, again consistent with the calculations that indicate rather negative E^* values. The excitation functions for production of some Es and Fm isotopes are shown in Figure 10.

Comparison of actinide production data for several different systems seems to indicate that transfers of the same number of nucleons from different neutron-rich heavy-ion projectiles, e.g., ^{18}O, ^{22}Ne, ^{48}Ca, proceed with similar cross sections provided the product excitation energy is not negative. In general, the yields drop off rather rapidly with the total number of nucleons transferred. Maximum cross sections for various mass Be transfers for several different systems are shown in Figure 11. These may actually be transfers of four protons from projectile to target with neutrons being transferred either way. (Similar curves can be con-

(29) Lee, D.; von Gunten, H.; Jacak, B.; Nurmia, M.; Liu, Y-f.; Luo, C.; Seaborg, G. T.; Hoffman, D. C. *Phys. Rev. C* **1982**, *25*, 286.
(30) Hoffman, D. C.; Hoffman, M. M. Los Alamos National Laboratory Report LA-UR-82-824, March 1982.
(31) Lee, D.; Moody, K. J.; Nurmia, M. J.; Seaborg, G. T.; von Gunten, H. R.; Hoffman, D. C. *Phys. Rev. C* **1983**, *27*, 2656.
(32) Hoffman, D. C. et al., private communication.

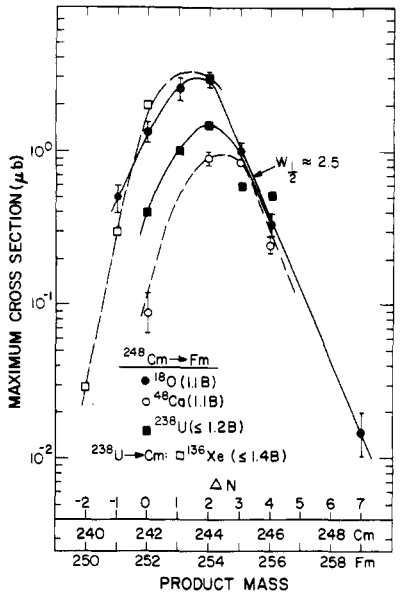

Figure 11. Cross sections for various mass Be transfers from ^{18}O, ^{48}Ca, and ^{238}U projectiles to ^{248}Cm to produce Fm isotopes and from ^{136}Xe projectiles to ^{238}U to produce Cm isotopes. Values in parentheses give the ratio of projectile energy to Coulomb barrier. (Data from ref 31–34.)

Table II
Peak Cross Sections for H through Be Transfers[a]

effective transfer	no. of neutrons transferred	cross section, μb
$^{1,2}H$	0,1	$(2\text{-}6) \times 10^3$
$^{2,3}He$	0,1	$(1\text{-}2) \times 10^3$
$^{4,5}Li$	1,2	$(1\text{-}4) \times 10^1$
^{5-7}Be	1-3	1-3

[a] Representative yields for effective transfers from various neutron-rich projectiles to actinide targets are given for the transferred masses for each Z transferred for which the cross sections are largest.

structed for H, He and Li transfers.) It is somewhat surprising to note that Be transfers from ^{136}Xe projectiles[33] to ^{238}U targets have about the same cross sections as from ^{18}O or even ^{238}U projectiles[31,34] to ^{248}Cm targets. These similarities tend to support the general idea of a binary transfer mechanism that gives a resulting nucleus with low E^*, i.e., a "cold" nucleus that is not immediately destroyed by prompt fission or particle emission. Production of these actinide products via compound nucleus formation followed by multiple particle emission would be expected to result in very low cross sections due to fission competition at each step, and it seems unlikely that comparable actinide yields would result from such dissimilar compound systems as $^{248}Cm + {}^{18}O$ ($^{266}104$) and $^{238}U + {}^{136}Xe$ ($^{274}146$).

Table II shows measured peak cross sections derived from data for effective transfers of H through Be from various neutron-rich projectiles to ^{248}Cm and ^{249}Cf targets. The transferred masses at which the cross sections are largest are shown for each transferred Z. Assuming similar cross sections and widths of the yield curves (Figure 11) for H through Be transfers to ^{254}Es, some of the new neutron-rich isotopes that might be produced with cross sections of a nanobarn or larger are shown in Figure 12. (It might also be possible to produce isotopes of elements 104 and 105 by transfer of B and C, but no data for such transfers are available for this region as yet.) Even at the lower limit of 1 nb, use of a 100 $\mu g/cm^2$ ^{254}Es target and a current of 1 particle μA of heavy ions would produce about 0.1 atom/min. This should permit on-line studies of SF and α-decay properties of many new, neutron-rich heavy-element isotopes; some may even have half-lives long enough for chemical separation. Studies of chemical properties such as volatilities, oxidation states, heats of sublimation, complexation behavior, and ionic radii might be possible. In addition to being of interest in their own right, knowledge of these properties would help assess the expected strong influence of relativistic effects in this region and distinguish among various proposed electronic configurations. Extending our knowledge of the architecture of the periodic table in the region of the heavy actinides should also improve our ability to predict the chemical properties of superheavy elements.

Concluding Remarks

If we are to fully exploit the use of transfer reactions to gain access into this region of new neutron-excess actinide and transactinide nuclides, we must develop high efficiency, on-line methods for positively identifying the Z and A and measuring the properties of these short-lived nuclides. This is of particular importance for studies of spontaneously fissioning isotopes because they cannot be identified by genetically linking them to known decay products as can be done for α-decaying nuclides. The use of newly developed large gas-ionization detectors in combination with time-of-flight techniques may make it possible to measure kinetic energy, velocity, energy loss, and other properties of the fission fragments. From such information, the Z and A of the fission products can be determined and the Z and A of the fissioning nucleus obtained to within a few units of Z and A, depending on the resolution of the system.

Exploration of the fission properties and half-lives for a whole new region of the most neutron-rich heavy-element isotopes will be critical to the development of a complete model of fission. Currently, there is no single fission model that can predict the abrupt changes in spontaneous-fission properties observed with the changes of only a few nucleons discussed earlier. Nor can spontaneous fission half-lives, especially for nuclides with odd numbers of neutrons or protons,[35] be predicted with any degree of accuracy. Hindrance factors due to single particle effects might lengthen the SF half-lives markedly over those for even–even isotopes and studies of isotopes with odd neutrons or protons will be particularly interesting. Furthermore, it is predicted[36] that the half-life systematics for spontaneous fission will change at element 104. Confirmation of such effects will, of course, require positive identification of the fissioning nucleus.

Knowledge of spontaneous-fission properties for nuclides at the edge of stability will be especially significant in determining the relative importance[18-26] of shell

(33) Schädel, M.; et al. *Phys. Rev. Lett.* **1978**, *41*, 469.
(34) Schädel, M.; et al. *Phys. Rev. Lett.* **1982**, *48*, 852.
(35) Randrup, J.; et al. *Phys. Rev. C* **1976**, *13*, 229; *Nucl. Phys. A* **1973**, *217*, 221.
(36) Baran, A.; Pomorski, K.; Lukasiak, A.; Sobiczewski, A. *Nucl. Phys. A* **1981**, *361*, 83.

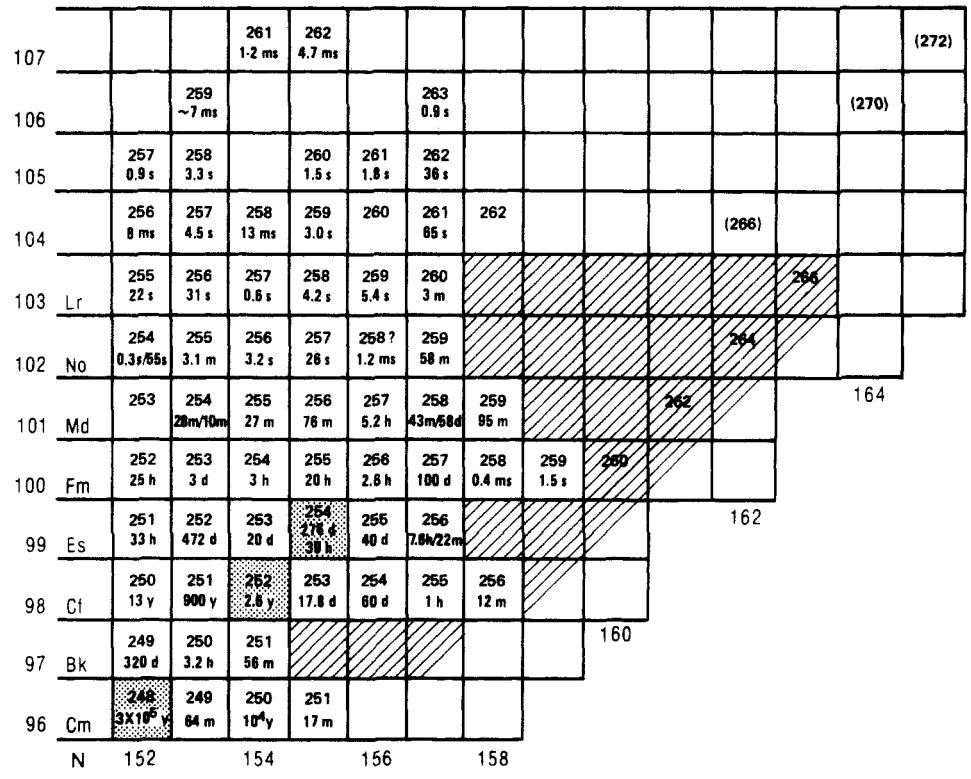

Figure 12. Portion of table of isotopes showing neutron-rich trans-Am isotopes. Hatched regions indicate new actinide isotopes postulated to be produced with >1-nb cross sections from transfer reactions to a 100 $\mu g/cm^2$ ^{254}Es target with a projectile beam of 1 μA. Compound nuclei for reactions of ^{18}O with ^{248}Cm, ^{252}Cf, and ^{254}Es are shown in parentheses.

effects in the fragments vs. those in the fissioning nucleus. Systematic information on the spontaneous fission properties of a range of these nuclides should give a better understanding of the fission process and help in the development of a dynamic, comprehensive model of fission that will enable us to predict the properties of still heavier elements and understand the ultimate limits to nuclear stability.