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### FISSION PROPERTIES OF VERY HEAVY ACTINIDES

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#### ABSTRACT

The existing data on neutron-emission, kinetic-energy and mass distributions, and half-lives for spontaneous fission of the heavy actinides are reviewed. A comparison of the data for the Fm isotopes with heavier and lighter nuclides suggests that the properties of the heavy Fm isotopes may be unique and can qualitatively be explained on the basis of fragment shell effects, i.e., symmetric fission results in two fragments with configurations close to the doubly magic <sup>132</sup>Sn nucleus. The effect of excitation energy and the use of systematics and theoretical predictions of fission properties and half-lives in the identification of new heavy element isotopes is discussed.

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#### I. INTRODUCTION

Much progress has been made in studying the properties of the lowenergy fission of the heavy actinides since the last Symposium on the Physics and Chemistry of Fission [1] in 1973. At that time, a trend toward increased yields of symmetric mass division for both spontaneous fission (SF) and thermal-neutron induced fission (n,f) had been observed [2-5] as the mass of the fissioning nucleus was increased. For 257 (Fm(n,f), the heaviest nuclide studied at that time, symmetric mass division was found [4] to be most probable, although the distribution was very broad. The average total kinetic energy (TKE) for SF and (n,f) of nuclides from <sup>230</sup>Th to <sup>256</sup>Fm was fit rather well by Unik et al. [2] with a linear function of the symmetric-fission coulomb repulsion parameter,  $z^2/A^{1/3}$ . The effect of excitation energy,  $E_x$ , on TKE had also been investigated and in several cases the extra excitation energy of around 6 MeV for (n,f) over SF had been found to increase the yields of symmetric mass division and the TKE for the same fissioning nuclide. However, in some cases the increased  $E_x$  resulted in increased neutron emission and a correspondingly smaller increase in TKE [2]. A review [6] of the data to early 1974 indicated that whether or not the  $\overline{\text{TKE}}$  increases with E<sub>x</sub> for a specific fissioning system depends on the result of averaging a large number of different energy dependences for individual fragment pairs, taking into account that as the fragment yields change with  $E_x$ , the statistical weights of the individual fragments change. Different fragment pairs were found to react differently to changes in  $E_x$  although the maximum TKE at low  $E_x$  was found for fragment masses around 132. At higher excitation energies, a decrease in TKE was noted for fragments around mass 132 which suggests a decreasing influence of fragment shells at higher Ex. Neutron emission from the fragments is also affected by the same considerations and varies with the excitation energy and deformation of the fragments. For symmetric mass division of 257 Fm (SF), TKE's which approached the Q value for fission had been reported [3], indicating that the fragments must have lower excitation energies which might be expected to result in the emission of fewer neutrons from the fragments. Indeed, it was reported [7] at the 1973 conference that the average neutron emission per fission,  $\overline{\nu}$ , for these symmetric mass splits with high TKE is only about 1 for  $^{257}$ Fm while it is 3 for  $^{252}$ Cf. It was postulated that the high TKE and low  $\overline{\nu}$  observed for  $^{257}$ Fm (SF) were because the fragments were becoming more spherical as they more closely approached the doubly magic <sup>132</sup>Sn configuration. This suggested that the effects should be still more pronounced for the heavier Fm isotopes. Whether or not these effects continue for elements of atomic number greater than 100 and for neutron numbers of 157 or more for other elements is important in assessing the relative importance of fragment shell effects near scission, the height of the second barrier relative to the ground state, and the question of adiabaticity [8] in nuclear fission.

Although measurements of SF properties for these heavy actinide isotopes are extremely difficult because of the short half-lives and small production cross sections, much new information has been reported since 1973. I will discuss some of the new data for low-energy fission of the heavy actinides, defined here as those with  $Z \ge 98$ , in terms of the half-lives, fragment mass and kinetic energy distributions, neutron emission, and the use of SF properties and systematics in the identification of new heavy element isotopes.

#### II. HALF-LIVES

Systematic trends in the SF half-lives of actinide and transactinide isotopes have been observed for the even-even actinide isotopes [9]. In general, the half-lives are shorter for the higher Z nuclides. However, beginning with curium, a pronounced stabilizing effect for 152 neutrons has been observed experimentally, and consequently, the half-life values overlap from one element to the next and even for a given Z. The calculations of Randrup et al. [10] and Baran et al. [11, 12] reproduce the general trends very well, but for a given nuclide can show deviations of several orders of magnitude. This is particularly apparent at <sup>258</sup>Fm where a "SF disaster" appears to have occurred--the SF half-life [13] being only 380  $\mu$ s, a factor of some 10<sup>6</sup> lower than the calculated value. This might be explained on the basis of the disappearance of the second fission barrier for <sup>258</sup>Fm. Recently a half-life of 12.3 minutes has been measured [14] for <sup>256</sup>Cf which has the same number of neutrons as <sup>258</sup>Fm. This indicates a reduction in half-life for the addition of 2 neutrons to  $^{254}$ Cf (60 days) of  $1.4 \times 10^{-4}$  compared to  $1.9 \times 10^{-3}$  from <sup>252</sup>Cf to <sup>254</sup>Cf. Although the half-life reduction for 2 neutrons between <sup>254</sup>Cf and <sup>256</sup>Cf is a factor of 10 larger than between <sup>252</sup>Cf and <sup>254</sup>Cf, no real "disaster" at 158 neutrons is indicated.

The theoretical calculations do not treat the odd nucleon cases in general, but the extra hindrance associated with the SF of nuclides having an odd number of protons or neutrons has been recognized for some time [15, 16]. The hindrance is typically of the order of 10<sup>5</sup>, but has been found to be as small as 10 and as large as 10<sup>10</sup>. The hindrance associated with N = 157 nuclei due to the hindrance of the 9/2+[615]neutron orbital has been calculated [16] and appears to be consistent with the data for  $^{257}$ Fm,  $^{259}$ No, and  $^{261}$ 104. The measurement of a 10% SF branch [17] for 1.5-s  $^{260}$ 105 indicates a hindrance of about 10<sup>3</sup> for the odd proton over predictions [16] for even 104 isotopes. The SF halflife of 1.5 seconds [18] measured for <sup>259</sup>Fm also indicates a hindrance factor of about 4  $\times$  10<sup>3</sup> for the odd neutron orbital, assuming that <sup>258</sup>Fm and <sup>259</sup>Fm would otherwise have about the same SF half-lives. The halflife of 95 min for <sup>259</sup>Md [19] indicates a hindrance for the odd proton of more than 10<sup>7</sup> relative to <sup>258</sup>Fm. If the second fission barriers no longer exist for isotopes of the even-even trans-nobelium elements, then the SF half-lives may be expected to be very short, i.e., milliseconds or less, and relatively constant with N. However, if the stabilizing effect of the N = 152 shell experimentally observed in Cf through No persists, half-lives in the region of N = 152 might be longer than calculated [10]. The recent dynamical calculations of Baran et al. [12] which additionally include the effect of  $\varepsilon_6$  deformations, show that the effect on the potential barrier is considerable and increases the half-lives calculated for the Fm isotopes as much as two orders of magnitude for <sup>252</sup>Fm. The calculated change in halflife systematics for trans-nobelium isotopes is less abrupt than proposed by Oganessian et al. [20], and gives much longer half-lives

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than those calculated by Randrup et al. [10]. It is clearly very important to obtain measurements of the SF half-lives of these isotopes in order to check the validity of various theoretical approaches and for extrapolation to still heavier regions. Because of the very short halflives of these nuclides, their production via complex heavy ion reactions with only nanobarn cross sections, and decay by SF which effectively destroys information concerning the Z of the fissioning parent nucleus, unequivocal assignment to a given Z and A is extremely difficult. However, much progress is being made in "on-line" observations of SF properties, including measurement of fragment kinetic energies and mass distributions, and coincidence measurements between characteristic x-rays, following electron-capture or alpha decay, and short-lived SF activities in order to determine the Z of the fissioning nuclide.

#### III. FRAGMENT MASS AND KINETIC-ENERGY DISTRIBUTION

Kinetic-energy distributions have recently been obtained for SF of  $^{256}Cf$ ,  $^{254}Fm$ ,  $^{258}Fm$ ,  $^{259}Fm$ ,  $^{259}Md$ , and  $^{252}No$  [14, 18, 19, 21, 22] from measurements of the kinetic energies of coincident fragments. Total kinetic energies and fragment mass distributions were also derived from these data. Mass distributions for <sup>250</sup>Cf, <sup>253</sup>Es, <sup>254</sup>Fm, and <sup>254</sup>Cf have been obtained from radiochemical measurements [21, 23-26]. The results of these measurements are summarized in Table I together with properties reported earlier for SF and (n,f) of some other trans-berkelium actinide isotopes. The peak-to-valley (P/V) ratios for the mass distributions can be seen to decrease rapidly with the addition of 2 protons between Cf and Fm isotopes having the same number of neutrons: e.g.,  $\geq$ 750 for  $^{252}$ Cf to  $\approx$ 42 for  $^{254}$ Fm;  $\geq$ 145 for  $^{254}$ Cf to 2.5 for  $^{256}$ Fm; asymmetric for  $^{256}$ Cf to narrowly symmetric for  $^{258}$ Fm. The P/V ratios also decrease with the addition of neutrons for both Cf and Fm as illustrated in Figs. 1 and 2. Only a small change is shown in going from <sup>250</sup>Cf to <sup>256</sup>Cf while an abrupt change to symmetric fission occurs for the Fm isotopes at N =The valley disappears completely between <sup>257</sup>Fm and <sup>258</sup>Fm, and 158. narrow, symmetric mass distributions [14, 18] have been measured for <sup>258</sup>Fm and <sup>259</sup>Fm. The mass distribution for <sup>259</sup>Md has also been found [17] to be symmetric and narrow.

A plot of  $\overline{\text{TKE}}$  vs  $z^2/A^{1/3}$  is shown in Fig. 3 with some data for the heavy actinides. The dashed line represents the fit of Viola [27] to the function  $\overline{\text{TKE}} = B(z^2/A^{1/3}) + C$  with B = 0.1071 and C = 22.2. The solid line is that of Unik et al. [2] with B = 0.13323 and C = -11.64. In general, most of the data fall between these two lines except for  $^{258}$ Fm and  $^{259}$ Fm which are 40 MeV or so higher. Schmitt and Mosel [28] have predicted that the  $\overline{\text{TKE}}$  for nuclides having masses of 260 to 275 would also be substantially higher than these linear extrapolations, but the measurement [29] of  $^{262}105(104)$  does not show this. However, the  $\overline{\text{TKE}}$  for  $^{259}$ Md, which also exhibits symmetric mass division, is below the lines although the TKE distribution for  $^{259}$ Md was found [19] to be extremely broad ( $\sigma = 44$  MeV or FWHM = 103 MeV). The value for  $^{252}$ No appears to be perfectly "normal." The  $\overline{\text{TKE}}$ 's of 238 to 242 MeV for  $^{258}$ Fm and  $^{259}$ Fm approach the total energy of about 250 MeV estimated to be available from fission. This is also the case for symmetric mass division of  $^{257}$ Fm where some events with TKE as high as 240 to 260 MeV, as

well as some very low TKE events, were observed. Similarly, the very large  $\sigma$  for the TKE distribution from <sup>259</sup>Md indicates some events with very high TKE.

The dip in TKE near symmetry which is  $\approx 20$  MeV for  $^{236}U^*$  decreases for the heavier fissioning systems and for  $^{255}Es^*$  has essentially disappeared. Similarly, for SF the dip at symmetry, which is around 15 MeV for <sup>246</sup>Cm, decreases to only a few MeV for <sup>250</sup>Cf. The TKE as a function of mass fraction,  $M_{\rm H}/A$ , for the Cf and Fm isotopes is shown in Figs. 4 and 5 respectively. "A gradual increase in the TKE at symmetry with mass of the fissioning nuclide is observed for the Cf isotopes until for <sup>256</sup>Cf the TKE at symmetry is highest by a few MeV. For the Fm isotopes, the TKE at symmetry increases by 15 MeV between <sup>256</sup>Fm and <sup>257</sup>Fm and by 20 MeV between <sup>257</sup>Fm and <sup>259</sup>Fm. A comparison of these trends in the form of contour plots of TKE as a function of mass fraction is shown in Fig. 6 for <sup>254</sup>Cf, <sup>256</sup>Fm, and <sup>257</sup>Fm. The greatly increased yield of symmetric mass division of <sup>257</sup>Fm compared to lighter actinides has been explained on the basis of the approach of the fragments from symmetric mass division to the doubly magic <sup>132</sup>Sn configuration. The high TKE at symmetry then results because of coulomb repulsion which is a maximum for spherical shapes. The large spread in TKE at symmetry indicates a large difference in fragment shapes and that some of the fragments must still be highly deformed. Thus 257Fm would appear to be in a "transition" region. The very high TKE's and narrow, symmetric mass distributions for <sup>258</sup>Fm and <sup>259</sup>Fm can also be explained on the basis of near spherical fragments which give the maximum TKE's. Since these approach the Q values for fission, the excitation energy of the fragments and subsequent neutron or gamma emission should be small. Although the measured [19] mass distribution for SF of <sup>259</sup>Md is symmetric and rather narrow, the very broad TKE distribution indicates a range of fragment shapes as for <sup>257</sup>Fm, again with some of the events having TKE's which approach the Q-value. However, it should be noted that for 259 Md, the most probable mass split is symmetric even for events with TKE <200 MeV while for <sup>257</sup>Fm the mass distribution becomes asymmetric for the lower TKE's. The effect of the odd proton is apparently very strong, and it has been suggested [30] that three-body fragmentation may be occurring, and accounts for the observed low TKE. Measurements of charge distribution for these highly symmetric systems would be extremely interesting, but are probably not feasible because of the short half-lives and low production cross sections.

#### IV. EFFECTS OF EXCITATION ENERGY

In general, increased excitation energy is expected to "wash out" shell effects. If the mass asymmetry and decrease in  $\overline{\text{TKE}}$  at symmetry for the SF of  $^{250}$ Cf,  $^{252}$ Cf, and  $^{256}$ Fm are attributed to shell effects, then an increase in yields and  $\overline{\text{TKE}}$  at symmetry for (n,f) relative to SF would be expected. Some of the pertinent data are summarized in Table I. Indeed, a decrease in the P/V ratios, indicating an increase in the yield of symmetric mass division, has been observed [2, 31] for  $^{250}$ Cf<sup>\*</sup>,  $^{252}$ Cf<sup>\*</sup>, and  $^{256}$ Fm<sup>\*</sup> (Fig. 7) relative to SF of the same nuclides. A small increase [2] in  $\overline{\text{TKE}}$  for  $^{250}$ Cf<sup>\*</sup> and small decreases [5] in  $\overline{\text{TKE}}$ 

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for  ${}^{252}$ Cf<sup>\*</sup> and  ${}^{256}$ Fm<sup>\*</sup> have been found relative to SF of  ${}^{250}$ Cf,  ${}^{252}$ Cf, and  ${}^{256}$ Fm. The changes do not appear to be large, and for symmetric mass division, the TKE's for both  ${}^{250}$ Cf<sup>\*</sup> and  ${}^{256}$ Fm<sup>\*</sup> are higher than for SF of these nuclides. Although these results may appear contradictory, whether or not the overall TKE increases or decreases with increasing E<sub>x</sub> depends on the details of each fissioning system, and is the result of averaging many different energy dependences for individual fragment pairs as their yields change with E<sub>x</sub>.

In the case of  ${}^{258}$ Fm<sup>\*</sup>, the effect of extra E<sub>x</sub> was quite clearly to decrease [4] the TKE and to broaden the mass distribution [4, 32] significantly relative to  ${}^{258}$ Fm (SF) as shown in Fig. 8. Perhaps the apparently conflicting results for  ${}^{258}$ Fm<sup>\*</sup> and  ${}^{256}$ Fm<sup>\*</sup> shown in Figs. 7 and 8 can be explained by postulating that for  ${}^{256}$ Fm and lighter actinides, fragment shells may tend to stabilize asymmetric mass division while in  ${}^{258}$ Fm the fragment shell effects stabilize symmetric division into two near doubly magic fragments. Thus the effect of the extra excitation energy in outweighing the shell effects increases the yield of symmetric mass splits for  ${}^{256}$ Fm<sup>\*</sup>, while for  ${}^{258}$ Fm<sup>\*</sup> the mass distribution is broadened and the yields at symmetry are decreased relative to  ${}^{258}$ Fm (SF). The large reduction in the yield of symmetric, near spherical mass splits for  ${}^{258}$ Fm<sup>\*</sup> also significantly reduces the TKE.

Recently, direct reaction studies [33] of the prompt fission of  $^{255}$ Es<sup>\*</sup>,  $^{256}$ Es<sup>\*</sup>, and  $^{255}$ Fm<sup>\*</sup> at E<sub>x</sub> from threshold to  $\approx$ 15 MeV and  $^{256}$ Fm<sup>\*</sup> from 10 to 24 MeV via the (d,pf), (t,pf), ( $^{3}$ He,df), and ( $^{3}$ He,pf) reactions on  $^{254}$ Es have given information about the effect of E<sub>x</sub> on fragment energies and mass yields. The mass distribution for  $^{255}$ Es<sup>\*</sup> at E<sub>x</sub> = 4 to 6 MeV was nearly the same as for  $^{254}$ Es (n,f) but showed significant increases in the yields near symmetry. In general, the yield of symmetric mass division increased monotonically with increasing TKE. The yield of symmetric fission for events with TKE > 210 MeV decreased for all these nuclides with increasing E<sub>x</sub>. For a given E<sub>x</sub>, the yield of symmetric, high TKE fission was highest for  $^{256}$ Fm and decreased in the order  $^{255}$ Fm<sup>\*</sup>,  $^{256}$ Es<sup>\*</sup>,  $^{255}$ Es<sup>\*</sup>. The TKE generally decreased and these asymmetric mass distributions broadened with increasing E<sub>x</sub> over the range studied. These results seem to be consistent with the weakening of shell effects with increasing E<sub>x</sub>.

The mass distribution of  ${}^{256}$ No<sup>\*</sup> at a  $E_x$  of  $\approx$ 25 MeV has been measured radiochemically [34] to be asymmetric, but at  $\approx$ 53 MeV, it becomes nearly symmetric. Apparently, shell effects are still stabilizing asymmetric fission to some extent up to  $\approx$ 25 MeV, while for the heavy Fm isotopes rather dramatic changes are seen in the mass distributions for thermal neutron fission compared to spontaneous fission even though the excitation energy is only about 6 MeV. This may indicate a rather small difference in potential energy for the asymmetric and symmetric paths to fission with a resulting high degree of sensitivity to  $E_x$ .

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#### V. NEUTRON EMISSION

The average number of neutrons emitted per fission event,  $\overline{\nu}_{T}$ , for low energy fission generally increases with Z as shown in Fig. 9. (Values for  $\overline{\nu}_{T}$  for thermal neutron fission have been corrected to zero  $E_x$ .) For the heavier actinides,  $\overline{\nu}_{T}$  also tends to increase with mass for a given Z. However, this trend is not shown by the Fm isotopes where  $\overline{\nu}_{T}$  is lower for masses 256 and 257 than for 254, although within the quoted errors it might be regarded as nearly constant.

The average values for the number of prompt neutrons emitted per fission event are, of course, not integral, but the probability for emitting a given number of neutrons has been measured for the low-energy fission of a large number of nuclides. It was early shown [35] that these "multiplicity" distributions could be approximated by a Gaussian distribution. Originally, most of the data could be fit with  $\sigma_{\rm V}$  = 1.08 ( $\sigma_{\rm V}^2$  = 1.17), except for  $^{252}$ Cf which required the use of  $\sigma$  = 1.21.

The variances for many heavier nuclides have now been measured and are plotted in Fig. 10. The variance for <sup>252</sup>Cf no longer appears to be anomalous. The variances are relatively constant for the isotopes of a given element, except for Fm, where much larger values are observed for masses 256 and 257. The variance of 4.0  $\pm$  1.3 reported [36] for  $^{252}102$ is still larger, even considering the quoted error. It was proposed by Dakowski et al. [37] that there was a correlation between  $\sigma_V^2$  and the fragment mass distribution, the highest  $\sigma_V^2$  being observed for the most symmetric distribution, i.e., the lowest P/V ratio. Some variances for neutron emission and TKE and P/V ratios are given in Table II and do, indeed, show such a trend. However, this may be attributed to the fact that <sup>256</sup>Fm, <sup>257</sup>Fm, and <sup>252</sup>102 are in transition regions, i.e., symmetric mass division results in fragments which although close to the spherical, doubly magic <sup>132</sup>Sn configuration, are still rather soft to deformation and thus exhibit a large difference in fragment shapes ranging from rather deformed to nearly spherical. This could account for the large variances, for both v and TKE. However, the trend is reversed for the highest TKE events (TKE > 240 MeV) from SF of <sup>257</sup>Fm. These have been found [7] to exhibit a very narrow, symmetric mass distribution  $(P/V \approx 0)$ , but  $\sigma_{\nu}^2$  is only 0.9. (The  $\overline{\nu}$  for these events is also low, 0.9  $\pm$  0.1, as might be expected because the TKE is approaching the estimated Q value for fission.)

Lazarev [38] has recently reviewed the data for  $\sigma_V^2$  and  $\sigma_{TKE}^2$  for low energy fission and summarized their dependences on A, Z,  $E_X$ , and the fissility parameter. He found that the ratio  $\sigma_V^2/\sigma_{TKE}^2$  is nearly constant for all the heavy nuclides studied to date. If this relationship continues for <sup>259</sup>Fm and <sup>259</sup>Md, which have very high  $\sigma_{TKE}^2$  values, then  $\sigma_V^2$  should also be very high and would continue the trend with increasing  $\sigma_V^2$  values toward low P/V ratios, i.e., more symmetric fission. However, it might be argued that for higher mass Fm isotopes,  $\sigma_V^2$  and  $\sigma_{TKE}^2$  should both become very small as the fragments all become nearly spherical which results in maximum TKE's which approach Q, and results in low  $E_X$ , and hence less neutron and gamma emission. It might be postulated that fissioning systems with Z > 100 would again

be in a transition region, e.g.,  $^{259}$ Md, and the variances and  $\overline{v}$  would increase as the fragments move away from the <sup>132</sup>Sn configuration. Relatively little detailed information for neutron emission as a function of fragment mass and kinetic energy has been obtained for heavy actinide nuclei except for  $^{252}$ Cf where a minimum in  $\overline{\nu}$  of  $\approx 0.5$  was found [39] in the region of A = 130. Such information is necessary for each fissioning system in order to obtain accurate pre-neutron emission kinetic-energy and mass distributions from kinetic energy measurements or to obtain pre-neutron masses from radiochemical measurements. Direct measurements of both the kinetic energies and velocities of the fragments require very intense sources which are not available for the heavier actinides, but  $\overline{V}(M)$  functions for  $^{255}$ Es\*,  $^{254}$ Fm, and <sup>256</sup>Fm have been deduced [2, 23] by an iterative method involving comparison of radiochemical and kinetic-energy measurements of the fragment yields. As in the case of <sup>252</sup>Cf, minimum neutron emission is found around mass 130 and can be correlated with the low deformation, closed-shell structure of these fragments.

Measurements of  $\overline{\nu}$  and  $\sigma_{\nu}^2$  as a function of fragment TKE and mass ratio have been made [40, 41] for the SF of <sup>250</sup>Cf, <sup>252</sup>Cf, <sup>254</sup>Cf, <sup>256</sup>Fm, <sup>257</sup>Fm, and the "unfolded" multiplicity distributions were obtained for all except <sup>256</sup>Fm. The  $\overline{\nu}$ 's for <sup>250,252,254</sup>Cf and <sup>256,257</sup>Fm were found to decrease monotonically with increasing TKE for a given mass split. This might be expected since the total energy is constant and is manifested primarily either in  $E_x$  or TKE of the fragments. Thus as the TKE increases,  $E_x$  and hence the energy available for the emission of neutrons (and photons), must necessarily decrease. This effect is most pronounced for <sup>257</sup>Fm.

A comparison of the  $P_t(v)$  distributions for the highest TKE events from <sup>250</sup>Cf, <sup>252</sup>Cf, and <sup>257</sup>Fm is shown in Fig. 11 and illustrates the large probability for the emission of 0 neutrons for <sup>257</sup>Fm for events with TKE > 240 MeV. This is consistent with a large fraction of these fragments being nearly spherical with resultant high TKE's which are nearly equal to the Q value. The measured TKE's for <sup>258</sup>Fm and <sup>259</sup>Fm of  $\approx$ 240 MeV are also close to the estimated Q value of around 250 MeV and indicate that the fragments must emit very few neutrons and are probably nearly spherical.

#### VI. DISCUSSION

The existing data on mass, kinetic-energy, and neutron-emission distributions for low-energy fission of the heaviest actinides and the effect of modest excitation energy on these properties have been reviewed. Most of the data are consistent with the systematics established for the lighter actinides, but the properties of the heavy fermium isotopes appear to be unique. The rather abrupt change in properties observed for SF of  $^{258}$ Fm and  $^{259}$ Fm which fission symmetrically with a very high TKE seems to be associated with the approach of the fragments from symmetric mass division to the spherical, doubly magic  $^{132}$ Sn configuration. The maximum yield of symmetric fragments

with associated very high TKE and low neutron emission, and the resulting decrease in these effects with increasing excitation energy seem to be best described by the asymmetric two-center shell model (ATCSM) calculations of Mustafa et al. [8, 42, 43] and the scission-point model of Wilkins et al. [44]. The cluster model of Gonnenwein et al. [45] also appears to qualitatively describe these results and the differences in mass distributions for Cf and Fm isotopes. Due to the formation of Z = 50 clusters in Fm (which cannot occur in lighter elements), symmetric mass division is energetically favored, the fragment deformation is small, and the TKE high compared to lighter actinides. However, the competition of both symmetric and asymmetric substructure effects should result in large variances, particularly for lower mass Fm isotopes. Almost equal barriers for symmetric and asymmetric fission should result and account for the observed sensitivity of fission properties to small increases in  $E_x$ . Maruhn and Greiner [46] have used the concept of mass symmetry, treated as a dynamical collective coordinate based on the ATCSM, to calculate mass distributions for <sup>226</sup>Ra, <sup>236</sup>U, and <sup>258</sup>Fm which are in qualitative agreement with the data. Ultimately, dynamical calculations which relate the potential energy surface of the fissioning nucleus with those of the fragments will probably be required for a complete understanding of the fission process.

The calculations of Mustafa and Ferguson [8] for  $98 \ge Z \le 106$  show that the transition from asymmetric to symmetric mass division occurs at N = 158 for Fm, a lower value than for the other elements. The calculated potential energy surfaces are rather shallow for both  $^{256}$ Fm and  $^{258}$ Fm, consistent with their sensitivity to the addition of small  $E_x$ . They predict that the transition to mass symmetry should occur at N = 160 for No, and at N = 162 for Cf and elements 104 and 106. This is in agreement with the observed asymmetric mass distribution for  $^{256}$ Cf. Their calculation for  $^{252}$ No also indicates a preference for asymmetric mass division, in agreement with experiment.

They performed calculations for two odd nuclides,  ${}^{257}$ Fm and  ${}^{262}$ 105, and found a preference for symmetric fission for  ${}^{257}$ Fm (similar to  ${}^{258}$ Fm) and asymmetric fission for  ${}^{262}$ 105. However, for  ${}^{262}$ 105, they found symmetric shapes to be preferred before the second saddle with asymmetric shapes becoming increasingly favored as the neck radius decreases en route to scission. They find similar results for  ${}^{256}$ Cf and  ${}^{256}$ Fm for which a preference for asymmetric mass division is shown even though there is no second barrier.

Mustafa and Ferguson find that the second barrier to fission has disappeared in Fm for N > 154, while for elements 102 and 104 the barriers persist for N > 154 but are below the ground state, and disappear entirely for large N. Element 106 isotopes show essentially no barrier. They argue that the observation of asymmetric fission for  $^{262}$ 105, even though it does not show a second barrier, may indicate that the process is adiabatic. Recent calculations of Mustafa [47] indicate that a rapid descent from saddle to scission does not lead to symmetric fission for the heavy Fm isotopes and is thus inconsistent with the measured mass distribution data. He therefore suggests that the potential energy surfaces are moving slowly near scission where fragment shell effects are strong. It is important to measure the properties of more trans-fermium isotopes to check this hypothesis and determine whether or not they all fission asymetrically and whether the TKE's are high as for  $^{258}$ Fm and  $^{259}$ Fm or whether they can again be represented by a linear extrapolation from the lower Z actinides.

Wilkins, Steinberg, and Chasman [44] have used a static model based on the assumption of statistical equilibrium at the scission point to calculate the relative probabilities of formation of complementary fission fragment pairs as determined from the relative potential energies of two nearly touching, coaxial spheroids. They are able to reproduce the general trends in kinetic-energy, charge, and mass distributions for Po to Fm isotopes. They are also able to interpret variations in neutron emission and TKE based on the fragment configurations at scission. The observed dip in TKE for symmetric mass division for the lighter actinides is explained on the basis that the deformation for symmetric mass splits of fissioning nuclides such as <sup>235</sup>U is considerably larger than for asymmetric splits, and hence the coulomb repulsion and TKE will be less at symmetry. They predict the transition to mass symmetry at A = 258 for Fm, but with one spherical and one highly deformed fragment due to stabilization of a deformed neutron shell at  $\beta$  = 0.85. This results in a maximum TKE of 220 MeV, somewhat lower than the observed TKE of about 240 MeV for 258Fm and 259Fm. They expect the maximum TKE for Fm isotopes to be  $\approx$ 225 MeV for <sup>262</sup>Fm rather than for <sup>264</sup>Fm because of this deformed shell at 80 neutrons. However, the total deformation at symmetry for heavy Fm isotopes is small with resulting higher TKE for symmetric relative to asymmetric mass division thus explaining the experimentally observed disappearance of the "dip" in TKE at symmetry for the Fm isotopes. They predict large variances for TKE and neutron emission in the <sup>258</sup>Fm to <sup>264</sup>Fm region because of the large differences in the deformation of the fragments. This is experimentally observed for the TKE and neutron emission of the "transi-tion" nuclide <sup>257</sup>Fm, but the TKE data for <sup>258</sup>Fm and <sup>259</sup>Fm seem to be better explained by two near spherical fragments [48].

Perhaps the low  $\overline{\text{TKE}}$  and extremely large variance in TKE and the narrow, symmetric mass distribution observed for  $^{259}$ Md can be explained on the basis of the stabilization of more deformed fragments for symmetric mass division. At A = 284, Wilkins et al. [44] predict a configuration which is symmetric in deformation as well as mass with a total deformation,  $\beta_1 + \beta_2 = 1.30$ , similar to that for lighter actinides. This suggests a return to the TKE systematics as a linear function of  $Z^2/A^{1/3}$  which have been found for lighter actinides [2, 27] rather than the high TKE predicted by Schmitt and Mosel [48] using a static scission model.

The mass distribution for <sup>258</sup>Fm calculated by Wilkins et al. [44] is triple-peaked while the data for <sup>258</sup>Fm (SF) show a very narrow symmetric mass distribution with essentially no asymmetric mass division. Their calculated mass distribution is more consistent with the data

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for  $^{258}$ Fm<sup>\*</sup> where the effect of the extra  $E_x$  is to broaden the mass distribution and increase the yield of the asymmetric component. Apparently, the details of the mass distribution are very sensitive to small changes in the potential energy surfaces and the overall agreement of their calculations with experimental data is relatively good over a broad range of fissioning systems using only a single set of parameters for the collective temperature, intrinsic temperature, and the spheroid separation distance.

Mosel [49] and Nix [50] have recently reviewed the various theoretical approaches to nuclear fission including the use of the Strutinsky "shell-correction" method [51], recent self-consistent calculations, and attempts to understand the descent from the second saddle to scission, nuclear viscosity, and the time scales involved.

Further measurements of the fission properties of the transfermium isotopes are needed in order to check the various theoretical approaches by determining whether there is a return to asymmetric mass distributions and lower TKE's. The measurements for <sup>252</sup>No and <sup>262</sup>105(104) indicate that this is the case but more data are needed. As the SF half-lives become still shorter, these measurements become increasingly difficult. Methods for measuring the SF properties for millisecond activities [52] will have to be developed. Another problem involves the unequivocal assignment of the Z and A of these nuclides. Assignment on the basis of half-life or fission properties alone is difficult because of the large overlap in properties, although the TKE and mass distributions of the heavy Fm isotopes appear to be unique. The TKE of the higher Z elements may well revert to a linear extrapolation based on asymmetric fission of lower Z actinides. The assumption of compound nucleus formation with the subsequent emission of neutrons becomes increasingly risky for heavy ions on heavy element targets. It is known [53] that SF activity from <sup>256</sup>Md-<sup>256</sup>Fm is formed with a relatively large cross section from bombardments of <sup>248</sup>Cm and <sup>249</sup>Bk with  $^{18}$ O and recently evidence [54] for the production of  $^{259}$ Fm in bombardments of <sup>248</sup>Cm with <sup>18</sup>O has been obtained. Thus a variety of nuclides can be formed in these reactions making identification and measurement of properties exceedingly difficult. Methods such as those of Bemis et al. [17] for measuring coincidences between characteristic x-rays following electron-capture or alpha decay, and short-lived SF activities can perhaps be used if sufficient activity can be produced. Hopefully, with the development of these and other ingenious techniques, it will be possible to elucidate the fission properties and unequivocally identify the many short SF activities of the actinide and trans-actinide elements which are produced in heavy ion bombardments. Knowledge of their properties and production modes should be invaluable in our understanding of nuclear fission and in extending the periodic table still further.

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TABLE I

LOW ENERGY FISSION PROPERTIES OF SOME HEAVY ELEMENT ISOTOPES

Fissioning Nuclide <sup>a</sup>	g SF TL (seconds)	Peak-to Valley Ratio <sup>D</sup>	TKE <sup>C</sup> (MeV)	o <del>tke</del>	$\overline{v}_{\mathrm{T}}^{d}$
<sup>250</sup> Cf	$5.4 \times 10^{11}$	>300 (RC)	187.0	11.3	3.49
<sup>250</sup> cf*	-	> 50 (RC)	189.1	13.0	-
<sup>252</sup> cf	2.7 x $10^9$	<u>&gt;</u> 750 (RC)	185.7	11.6	3.735
<sup>252</sup> Cf*	<b>-</b> • •	≈20 (RC)	185	15.5	-
254 <sub>Cf</sub>	$5.2 \times 10^{6}$	<u>&gt;145 (RC)</u>	186.9	11.8	3.89
256 <sub>Cf</sub>	7.4 x $10^2$	Asymm. (SS)	189.8	14.6	<b>_</b>
· ·					
253 Es	$2.0 \times 10^{13}$	326 (RC)	191	13.4	-
255 * Es	<b>-</b> ,	≈8(SS)	194.3	15.9	. –
254 <sub>Fm</sub>	$2.0 \times 10^{7}$	≈42(RC)	195.1	11.7	3.96
256 <sub>Fm</sub>	$1.0 \times 10^4$	12(SS)	197.9	14.4	3.70
256_* Fm	-	2.5(RC)	195.5	18	-
257 Fm	4.1 x $10^9$	≈1.5(SS)	197.6	15.3	3.77
258 <sub>Fm</sub>	$3.8 \times 10^{-4}$	Symm., $\sigma = 8(SS)$	238 <sup>‡</sup>	14	-
258_* Fm	-	Symm., broad(SS)	197	-	-
259 <sub>Fm</sub>	$1.5 \times 10^{0}$	Symm., $\sigma = 11(SS)$	242 <sup>‡</sup>	21	-
259 <sub>Md</sub>	5.7 x $10^3$	Symm., $\sigma = 13$ (SS)	189 <sup>‡</sup>	44	-
252 <sub>No</sub>	8.6 x 10 <sup>0</sup>	Asymm. (SS)	202.4	15.4	4.15

<sup>a</sup>This is either the spontaneously fissioning nuclide or the excited compound nucleus formed by (n,f) and designated by \*. <sup>b</sup>Peak-to-valley ratios from radiochemical (RC) or solid-state (SS)

<sup>D</sup>Peak-to-valley ratios from radiochemical (RC) or solid-state (SS) measurements from compilation in Ref. 6, p. 159 and Refs. 2, 14, 18, 19, 21-26, 31, 32.

<sup>C</sup>These average values of the pre-neutron emission TKE's except for those designated by <sup>‡</sup> which are most probable pre-neutron emission values from a provisional mass analysis without corrections for neutron emission. Data from compilation in Ref. 6, p. 159 and Refs. 2, 14, 18, 19, 21-26.

<sup>d</sup>Data from Refs. 6, 7, 38, 40, 41.

## TABLE II

Peak-to-Valley Ratios, P/V, and Variances,  $\sigma_V^2$ , of the Unfolded Neutron Multiplicity Distributions for Spontaneous and Thermal-Neutron Fission (\*) of Some Actinide Isotopes [Refs. 2, 6, 7, 12, 17, 18, 21, 22, 23, 36-41]

Nuclide	· · · · · · ·	σ <mark>2</mark> ν	P/V	σ <mark>2</mark> TKE
234 <sub>U</sub> *	1.208	± 0.008	440	98.0
236 <sub>U</sub> *	1.236	± 0.008	620	106.1
240_* Pu	1.40	± 0.01	150	132.3
242 <sub>Cm</sub>	1.21	± 0.03	>700	-
244 <sub>Cm</sub>	1.23	± 0.05	>5700	122.6
<sup>250</sup> Cf	1.49	± 0.03	>300	127.7
<sup>252</sup> Cf	1.57	± 0.01	<u>≥</u> 750	]34.6
<sup>254</sup> Cf	1.56	± 0.01	<u>&gt;</u> 145	139.2
253 Es		-	326	179.6
255 * Es		-	≈8	252.8
254 <sub>Fm</sub>	1.50	± 0.20	≈42	162.6
256 Fm	1.82	± 0.08	12	207.4
257 <sub>Fm</sub>	2.51	± 0.02	≈1.5	197.5
257 Fm(TKE>235 MeV)	0.09	± 0.02	Sym. (0)	-
258 Fm		?	Sym.(0)	≈200
259 Fm		?	Sym.(0)	≈400
259 <sub>Md</sub>		?	Sym.(0)	≈1900
<sup>252</sup> 102	4.0	± 1.3	Asym.	222

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#### FIGURE CAPTIONS

- 1. Pre-neutron emission mass-yield distributions for <sup>250</sup>Cf [2], <sup>252</sup>Cf [2], <sup>254</sup>Cf (solid curve from ref. 2; dashed curve from ref. 14), and <sup>256</sup>Cf [14]. The data for <sup>254</sup>Cf and <sup>256</sup>Cf from ref. 14 were analyzed in 5 AMU mass bins using an empirical neutron correction similar to that for <sup>252</sup>Cf.
- 2. Pre-neutron emission mass yield curves for <sup>254</sup>Fm [21], <sup>256</sup>Fm [2], <sup>257</sup>Fm [3], <sup>258</sup>Fm [14], and <sup>259</sup>Fm [18]. The solid curve for <sup>256</sup>Fm is a pre-neutron emission curve from ref. 2 while the dashed curve is a provisional mass analysis for <sup>256</sup>Fm measured in the same experimental set-up as used for <sup>258</sup>Fm [14].
- 3. TKE vs.  $Z^2/A^{1/3}$  for heavy actinide isotopes. Solid line represents linear fit of Viola [27]; dashed line is from Unik et al. [2]. The data for  ${}^{258}$ Fm and  ${}^{259}$ Fm are most probable TKE's [14, 18].
- 4. TKE vs. mass fraction for Cf isotopes. (Data from refs. 2 and 14.)
- 5. TKE vs. mass fraction for Fm isotopes. (Data from refs. 2, 3, 14.)
- Contour plots of TKE vs. mass fraction for <sup>254</sup>Cf, <sup>256</sup>Fm, and <sup>257</sup>Fm [41]. The contours are lines of relative numbers of events based on data groupings 5 MeV × 0.01 units of mass fraction.
- 7. Mass-yield curves for  $^{256}$ Fm<sup>\*</sup> and  $^{256}$ Fm [31].
- 8. Mass-yield curves for <sup>258</sup>Fm<sup>\*</sup> [4] and <sup>258</sup>Fm [14].
- 9. Experimental values of  $\overline{\nu}_T$  as a function of A of the compound nucleus. Data for SF are shown by +. Measurements for  $\overline{\nu}_T$  for (n,f) fission have been corrected to zero excitation energy using  $d\overline{\nu}_T/dE_x = 0.11$ MeV<sup>-1</sup> and are shown by 0. (Data from refs. 6, 31, 38.)
- 10. Variances of the neutron multiplicity distributions,  $\sigma_V^2$ , plotted as a function of Z and A of the fissioning nucleus. (Data from refs. 36, 38, 40, 41.)
- 11.  $P_t(v)$  for <sup>250</sup>Cf, <sup>252</sup>Cf, and <sup>257</sup>Fm for the fission events having the highest TKE's [40].



-18-



Fig. 2





-21-



- 2 2 -





-24-



- 25 -



-26-



Fig. 10

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