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**TITLE:** FISSION PROPERTIES OF EINSTEINIUM AND FERMIUM

**AUTHOR(S):** Darleane C. Hoffman

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## FISSION PROPERTIES OF EINSTEINIUM AND FERMIUM

Darleane C. Hoffman  
Los Alamos Scientific Laboratory

Since the discovery of einsteinium and fermium in the Mike debris in the form of the isotopes  $^{253}\text{Es}$ ,  $^{255}\text{Es}$ , and  $^{255}\text{Fm}$ , some two dozen more isotopes of these elements have been identified, many of which spontaneously fission. Studies of the spontaneous fission (SF) decay properties, particularly of the fermium isotopes, have been especially fascinating to me. Most of my remarks will be concerned with the fission properties of the fermium isotopes, primarily because so much more is known about them than about the SF of the einsteinium isotopes, and because a dramatic change in SF properties has been found to occur with increasing mass of the fermium isotopes. A dramatic increase in total kinetic energy (TKE) release and a change in the mass division from predominantly asymmetric to predominantly symmetric has been found to occur between  $^{256}\text{Fm}$  and  $^{258}\text{Fm}$ , with  $^{257}\text{Fm}$  apparently being in a "transition" region.

Relatively little is known concerning the SF of the einsteinium isotopes, probably primarily because the half lives for SF of the isotopes known so far are very long ( $10^3$  to  $10^7$  years) compared to those for alpha decay, and it has been difficult to obtain sufficiently strong sources for study because of the extremely high alpha decay rates. Isotopes for which measurements of the kinetic energy release and mass distributions have been made<sup>1,2</sup> so far include  $^{253}\text{Es}(\text{SF})$  and  $^{254}\text{Es}(\text{n},\text{f})$ , i.e., excited  $^{255}\text{Es} (^{255}\text{Es}^*)$ . The properties are similar to those of the lower Z actinides although the peak-to-valley ratio is somewhat lower and the TKE release is somewhat higher for  $^{255}\text{Es}^*$  than expected, but this may be due to the effect of the extra excitation energy resulting from neutron capture. Studies of the heavier einsteinium isotopes, although difficult, should be pursued for comparison with the fermium isotopes having the same number of neutrons in order to try to ascertain the effect of having one less proton. Such investigations of the kinetic-energy and mass distributions as a function of excitation energy can be done for prompt fission of  $^{255}\text{Es}$  ( $N = 156$ ) via the  $^{254}\text{Es}(\text{d},\text{p})$  reaction, and of  $^{256}\text{Es}$  ( $N = 157$ ) via

the  $^{254}\text{Es}(t,p)$  reaction using direct reaction techniques. We believe this can be done at the LASL Tandem Van de Graaff Facility using an isotope-separated  $^{254}\text{Es}$  target, and in collaboration with coworkers from LLL hope to perform such experiments this spring. Prompt fission of  $^{255}\text{Fm}$  and  $^{256}\text{Fm}$  via the  $(^3\text{He},d)$  and  $(^3\text{He},p)$  reactions will also be investigated for comparison using the same target and techniques.

I would now like to turn my attention to the systematics of the low-energy fission of the fermium isotopes, and consider the half lives, mass division, kinetic-energy release, and accompanying prompt neutron emission.

The SF half lives for the even-even fermium isotopes show a maximum of about 100 years at the  $N = 152$  subshell with the half lives decreasing very rapidly for either higher or lower neutron numbers. Based on the half life of 125 years for SF of  $^{257}\text{Fm}$ , it was at first estimated that  $^{259}\text{Fm}$  might have an SF half life of about a month. However, it was not detected in debris from several underground nuclear test in which it should have been produced<sup>3,8</sup>, and limits of less than 5 hours or greater than 7.5 years were placed on its half life. With the discovery in 1971 by Hulet and coworkers<sup>4</sup> that the half life of  $^{258}\text{Fm}$  was only 380 microseconds, it appeared that a "disaster" in SF half lives had occurred in the fermium isotopes after  $N = 157$ . The half life of  $^{259}\text{Fm}$  might easily then be much less than the 5 hour limit we had set earlier. If the reduction in half life between  $^{257}\text{Fm}$  and  $^{259}\text{Fm}$  was the same as that of  $4 \times 10^{-8}$  between  $^{256}\text{Fm}$  and  $^{258}\text{Fm}$ , a half life of 2 to 3 minutes could be estimated for  $^{259}\text{Fm}$ . However, calculations of Randrup et al.<sup>5</sup> indicated that disappearance of the second fission barrier at  $^{258}\text{Fm}$  could account for its very short half life. Thus  $^{258}\text{Fm}$  and  $^{259}\text{Fm}$  might have about the same half life except for the special hindrance<sup>6</sup> associated with the odd neutron and  $^{259}\text{Fm}$  might have a half life of only a few-tenths of a second. Finally, Weber, Wilhelmy, and I at IASL in cooperation with Hulet, Loughheed, Landrum, and Wild from LLL succeeded in 1975, after several attempts, in producing<sup>7</sup>  $^{259}\text{Fm}$  and measuring its half life as 1.5 seconds. This is the most neutron-rich nuclide so far identified and although its half life is perhaps not as short as the most pessimistic estimates, neither is it long enough to indicate a reversal of the

"disaster". Chances for producing still more neutron-rich fermium isotopes appear very remote at the present, even if their half lives were sufficiently long for measurement.

As early as 1963, Brandt et al.<sup>1</sup> reported double-kinetic energy measurements of the SF of  $^{254}\text{Fm}$  which showed properties not unlike those of the lower Z actinides. The mass division was still strongly asymmetric and the TKE release was not anomalously high, being roughly consistent with a linear extrapolation of TKE values for other actinides as a function of  $Z^2/A^{1/3}$ . However, even though it was commonly believed that all low energy fission, i. e., SF and thermal neutron-induced fission, always resulted in mass distributions which were strongly asymmetric, exhibiting the familiar double-humped mass distribution, some of us continued to speculate that the more neutron-rich fermium isotopes might show an increased yield of symmetric mass division as the fragment configurations could more closely approach the stable, doubly magic  $^{132}\text{Sn}$  core. Closed-shell effects usually become prominent only within a few nucleons of shell closure and, therefore, the effect might not show up at  $^{254}\text{Fm}$ , but might become apparent at  $^{257}\text{Fm}$ . Although  $^{257}\text{Fm}$  was not identified in the Mike debris, sources for study of its 0.2% SF branch were later obtained from debris from subsequent nuclear test conducted underground at the NTS, as well as from the AEC transplutonium production program at the high-flux isotopes reactor (HFIR) at Oak Ridge. (In fact, the largest source of  $^{257}\text{Fm}$  ever obtained was isolated at LLL from the Hutch debris<sup>8</sup>.) Initial double-kinetic energy measurements were performed on a source of  $^{257}\text{Fm}$  of only 0.8 SF/min obtained from Hutch debris and on a 2.7 SF/min source from the HFIR. Measurements on both types of sources showed greatly enhanced yields for symmetric mass division and an increase in TKE release--these results were reported<sup>9</sup> in 1970 and 1971, not without some trepidation! A monotonic increase in  $\overline{\text{TKE}}$  with approach to symmetry was also observed, in contrast to the measurements for  $^{252}\text{Cf}$ ,  $^{254}\text{Cf}$ , and lower Z actinides which show a decrease, as illustrated in Fig. 1. The high TKE release at symmetry was attributed to the sphericity of the fragments which would give rise to a larger TKE because of increased Coulomb repulsion. However, the variance of the TKE was also very large, indicating that some of the symmetric fragments were still ellipsoidal in shape, and that  $^{257}\text{Fm}$  might be in a "transition" region where

the fragments were still "soft" toward deformation. The mass distribution shown in Fig. 2 was obtained, but as is illustrated, the yields at symmetry were extremely sensitive to the neutron-emission function which was used in correcting the data. Since the TKE at symmetry for some of the fragments approached the total energy available from fission, we felt that neutron emission must be very low at symmetry and that this was a point that deserved experimental investigation. About the same time, thermal neutron-induced fission of  $^{257}\text{Fm}$  was measured by John et al.<sup>10</sup> and a still more symmetric, but very broad mass distribution with the most probable mass split being symmetric, was obtained. These observations indicated that there might indeed be something "new" in low energy fission as Glenn Seaborg had put it at the 1970 Houston meeting, and considerable interest was generated in additional measurements of the fermium isotopes. Kinetic-energy and radiochemical measurements were made for a number of fermium isotopes<sup>2,11-16</sup>. The results, which are shown in Fig. 3, indicated a clear trend toward more symmetric mass distributions and higher TKE's with increasing mass of the fermium isotopes. Our recent measurements of the SF of  $^{258}\text{Fm}$  (to be published) and  $^{259}\text{Fm}$  (Ref. 7) performed at LASL in collaboration with Hulet et al. from LLL, show that the most probable mass division for both isotopes is narrowly symmetric with a very high TKE release, consistent with the trends exhibited for the lighter fermium isotopes, but exhibiting a much greater change. The mass-yield curves shown in Figs. 4 and 5 show this effect.

Differences in mass distributions between SF and thermal neutron-induced fission caused by the excitation energy of about 6 MeV resulting from the neutron-binding energy can be seen by comparing the mass distributions for  $^{256}\text{Fm}(\text{SF})$  with  $^{256}\text{Fm}^*$  and  $^{258}\text{Fm}(\text{SF})$  with  $^{258}\text{Fm}^*$  shown in Figs. 6 and 7, Refs. 17, 10. The mass distribution for  $^{256}\text{Fm}^*$  shows increased yields for symmetric mass splits and a slightly higher  $\overline{\text{TKE}}$  than for SF of  $^{256}\text{Fm}$ . In the case of  $^{258}\text{Fm}^*$ , the effect is just the opposite--the mass distribution is less sharply symmetric and the  $\overline{\text{TKE}}$  has been reported<sup>10</sup> to be only 183 MeV compared to the most probable value of 238 MeV we have measured for  $^{258}\text{Fm}(\text{SF})$ . Perhaps these apparently conflicting results can be explained on the basis of fragment shell effects which tend to be washed out by the extra excitation energy in the case of neutron-induced fission. In the case of  $^{256}\text{Fm}$  and lighter actinides, the fragment shell

effects may tend to stabilize asymmetric mass division while in  $^{258}\text{Fm}$  they stabilize symmetric mass division. Thus the extra excitation energy, by outweighing the shell effects, makes the distribution more symmetric in the one case, and broader and more asymmetric in the other. Alternatively, the abrupt change in SF properties at  $^{258}\text{Fm}$  may be explained in terms of the potential energy surface of the fissioning nucleus. The potential energy surface being sampled during fission may be very shallow between the symmetric and asymmetric mass components. In  $^{256}\text{Fm}(\text{SF})$ , the potential energy minimum would be associated with a configuration giving asymmetric mass division and the addition of energy would allow a broader sampling of the surface which would permit some fission via symmetric mass division with a higher TKE. In  $^{258}\text{Fm}(\text{SF})$ , the potential energy minimum would be presumed to be associated with symmetric mass division and addition of energy to the system would allow the fissioning nucleus to sample mass asymmetric components of the potential energy surface which would also result in lower TKE release.

The measured and calculated values for the  $\overline{\text{TKE}}$  for low energy fission of the fermium isotopes and peak-to-valley ratios for the mass distributions are summarized in Table I. The calculated values of TKE, based primarily on liquid-drop considerations<sup>2,18</sup>, i.e., that the kinetic energy release is a linear function of  $Z^2/A^{1/3}$ , show a slight decrease with increasing A for a given Z, while the measured values for fermium show a slight increase between mass 254 and 256, 257 and a precipitous increase for 258 and 259. (See Fig. 8.)

A plot of average neutron emission,  $\bar{\nu}_T$ , as a function of A is shown in Fig. 9, and in general is seen to increase more or less linearly. However, as the total kinetic energy release increases and approaches the Q value for fission as in the fermium isotopes, the excitation energies of the fragments and neutron and/or gamma emission must of necessity decrease rather than continue to increase as shown in Fig. 9. From the data given in Table I, it can be seen that  $\bar{\nu}_T$  does indeed decrease as the TKE increases between  $^{254}\text{Fm}$  and  $^{256}\text{Fm}$  and  $^{257}\text{Fm}$ . The variance for neutron emission also goes up markedly between  $^{256}\text{Fm}$  and  $^{257}\text{Fm}$ , reflecting the large variance observed for the TKE for  $^{257}\text{Fm}$ . Studies of neutron emission in SF of  $^{257}\text{Fm}$  and of  $^{252}\text{Cf}$  showed<sup>19,20</sup> that for fission events with TKE greater than 240 MeV

(about 5% of the total fissions),  $\bar{\nu}_T$  was only 0.9 for  $^{257}\text{Fm}$  while for  $^{252}\text{Cf}$  it was still 2.2 for the 3% of the fissions having the highest TKE's. Some of our recent measurements<sup>21</sup> of neutron multiplicities (Fig. 10) also illustrate this point. The mass distribution for fission events from  $^{257}\text{Fm}$  having TKE greater than 235 MeV is shown in Fig. 11. It is narrowly symmetric with a  $\Gamma$ WHM of about 7 mass units and  $\bar{\nu}_T$  is only about 1 for these events. Similarly neutron emission for SF of  $^{258}\text{Fm}$  and  $^{259}\text{Fm}$  would also be expected to be extremely low since their most probable values of TKE are about 240 MeV and approach the total energy of about 250 MeV estimated to be available from fission.

In summary then, I have tried to show that the low energy fission of the fermium isotopes is a microcosm of the fission process, exhibiting a wide range of half lives, mass and kinetic-energy distributions and varying neutron emission. The trends in the fermium isotopes toward higher yields for symmetric mass division, higher total kinetic energies, and reduced neutron emission as the mass of the fermium isotopes is increased are consistent with the simple postulate that the closer the fragments resulting from symmetric mass division are to the doubly magic  $Z = 50$ ,  $N = 82$  configuration, the more highly favored symmetric fission becomes. The fragments become more spherical which results in increased kinetic energy, decreased neutron emission, and probably decreased gamma emission. The variances for the mass, kinetic-energy, and neutron distributions appear to be largest in the "transition" nucleus  $^{257}\text{Fm}$  where symmetric mass division apparently can result in fragment shapes ranging from rather deformed to nearly spherical as evidenced by the observation of symmetric mass division with both very high and very low TKE's. However, at  $^{258}\text{Fm}$  and  $^{259}\text{Fm}$  where symmetric mass division results in fragments with 79 or 80 neutrons, they are close enough to the 82-neutron shell so that most of them are quite spherical, resulting in the observed highly symmetric mass distributions, very high TKE's, low excitation energies, and presumably low neutron emission. If fermium isotopes up to mass 264 could be observed, one might speculate that nearly all symmetric mass division with the TKE approaching that available from the fission process might result. We have measured the SF properties for the even californium isotopes from  $N = 152$  to  $N = 158$  and see no such abrupt change or trend in fission properties as observed for the fermium

isotopes. It will be interesting to see whether the einsteinium isotopes with Z between californium and fermium show SF properties similar to the fermium isotopes or not.

I think it is important to try to correlate the SF properties of these nuclides with the theoretical predictions. Mustafa<sup>22</sup> has calculated potential energy surfaces for fission of the fermium isotopes which are consistent with our observations. His calculations indicate a preference for asymmetric mass division for  $^{256}\text{Fm}(\text{SF})$  and symmetric mass division for  $^{258}\text{Fm}(\text{SF})$ . The calculated potential energy region is rather flat in both cases and would be qualitatively consistent with the effects observed experimentally with the addition of excitation energy to the system via thermal neutron-induced fission.

Wilkins, Steinberg, and Chasman<sup>23</sup> also predict a rapid change from asymmetric to symmetric mass division in the vicinity of  $^{258}\text{Fm}$ . Their calculations imply a symmetric division consisting of one spherical and one elongated fragment, resulting in a lower TKE release than for two spherical fragments. A  $\overline{\text{TKE}}$  of about 225 MeV would be predicted while our data give a most probable value of 238 MeV for  $^{258}\text{Fm}$  and show substantial yields for fission splits with still higher TKE. Thus, our results suggest somewhat more compact, spherical shapes for the fragments at scission and/or a larger amount of pre-scission kinetic energy than do their predictions. Their calculated mass distribution is triple-peaked while our results for  $^{258}\text{Fm}(\text{SF})$  show little, if any, of this effect. However, the experimentally observed effects must be extremely sensitive to small changes in the potential energy surfaces and may be outside the accuracy which can reasonably be expected even from realistic static calculations. Ultimately, dynamic calculations incorporating both the potential energy surface of the fissioning nucleus and the shell effects in the fragments will probably be required in order to completely explain the phenomena associated with the fission process.

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## FIGURE CAPTIONS

- Figure 1. Contour diagrams showing pre-neutron-emission total kinetic energy distributions for  $^{257}\text{Fm}$  and  $^{254}\text{Cf}$  as a function of mass fraction. The contours are lines of relative numbers of events, based on data groupings 5 keV x 0.01 units of mass fraction. (from Ref. 9).
- Figure 2. Pre-neutron-emission mass-yield curve for  $^{257}\text{Fm}$  from Ref. 9.
- Figure 3. Normalized mass-yield distributions for SF of  $^{254}\text{Fm}$  (RC, Ref. 2),  $^{256}\text{Fm}$  (RC, Ref. 13),  $^{257}\text{Fm}$  (SS, Ref. 9), and for  $^{258}\text{Fm}^*$  (SS, Ref. 10).
- Figure 4. Normalized mass-yield distributions for SF of  $^{254}\text{Fm}$  (RC, Ref. 2),  $^{257}\text{Fm}$  (SS, Ref. 9), and  $^{259}\text{Fm}$  (SS, Ref. 7).
- Figure 5. Mass distributions for SF of fermium isotopes.
- Figure 6. Mass-yield curves for  $^{256}\text{Fm}$ (SF) and  $^{256}\text{Fm}^*$  from Ref. 15.
- Figure 7. Provisional mass distributions for  $^{258}\text{Fm}$ (SF) and  $^{258}\text{Fm}^*$  from Ref. 10.
- Figure 8. Systematics for the average TKE release for SF of curium through nobelium isotopes. The solid line represents the systematic trend of Viola (Ref. 18) and the dashed line is from the analysis of Unik et al. (Ref. 2).
- Figure 9. Experimental values of  $\bar{\nu}_T$  as a function of A of the compound nucleus. Data for SF are shown by (+). All measurements of  $\bar{\nu}_T$  for (n,f) are corrected to zero excitation energy and are shown by (o). (Figure from Ref. 11).
- Figure 10. Multiplicity distributions for  $^{250}\text{Cf}$ ,  $^{252}\text{Cf}$ ,  $^{254}\text{Cf}$ , and  $^{257}\text{Fm}$  from Ref. 21.
- Figure 11. Mass distribution for fission events from  $^{257}\text{Fm}$  having pre-neutron TKE greater than 230 keV, from Ref. 19.

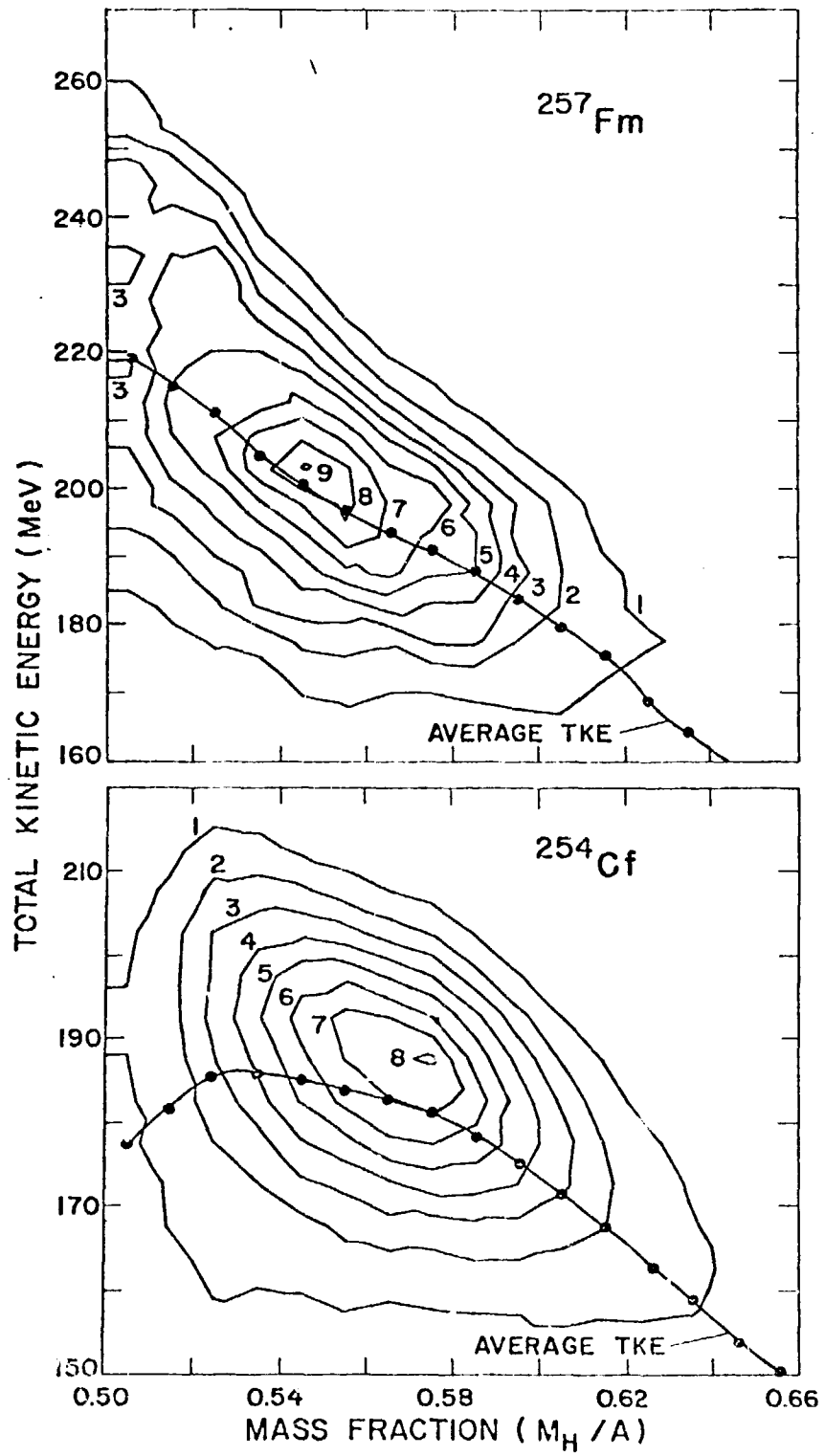


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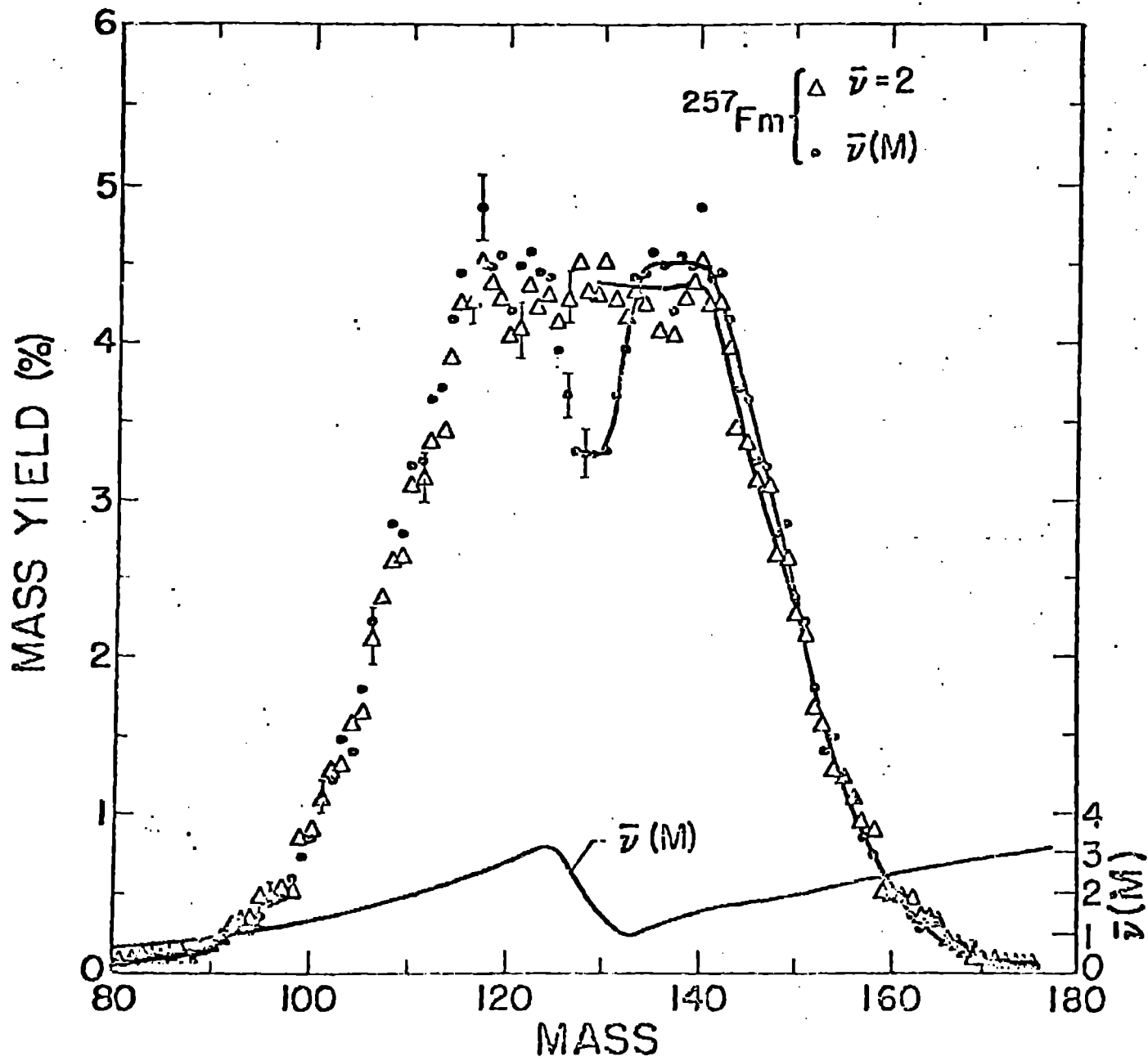


Figure 2

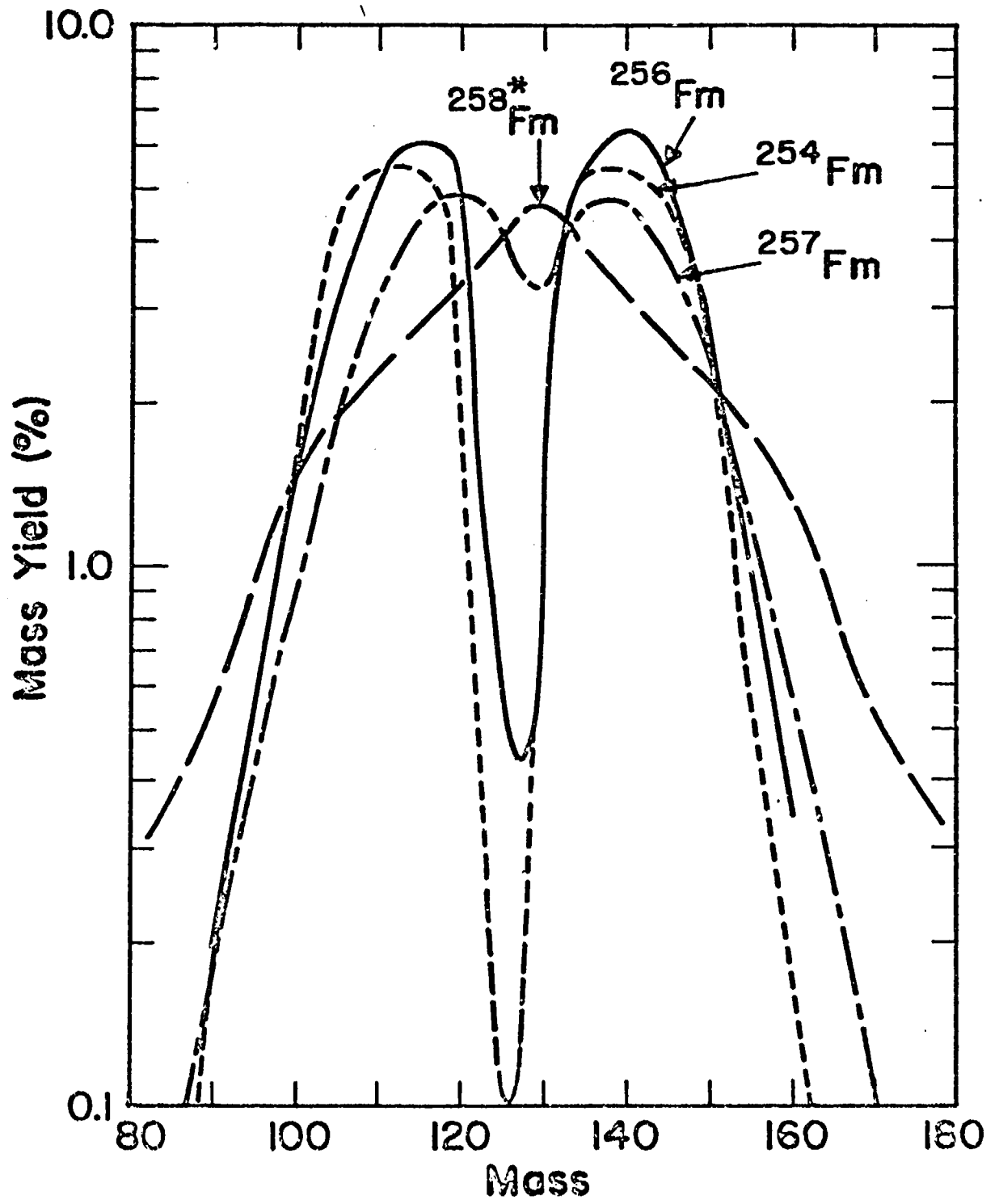


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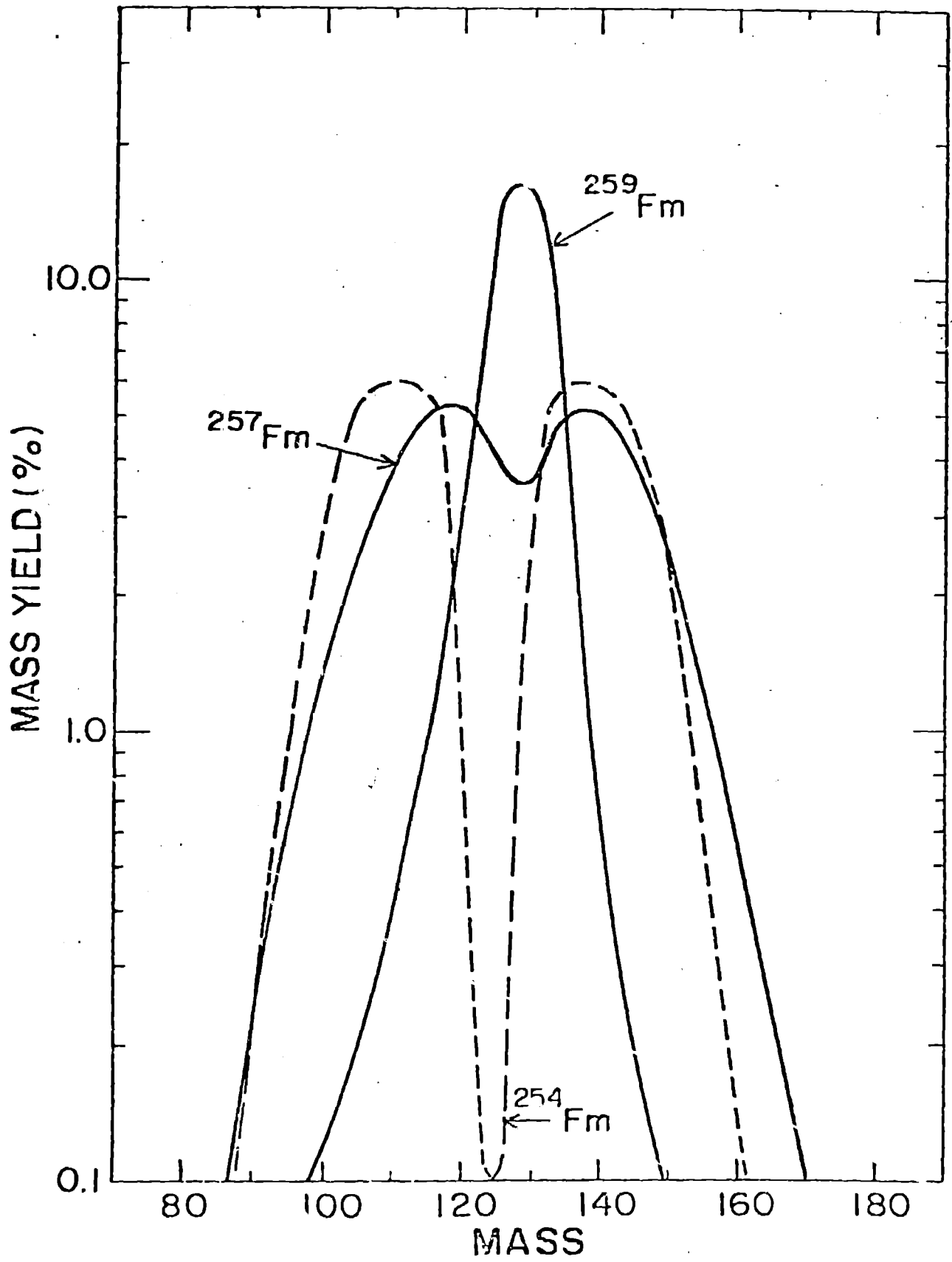


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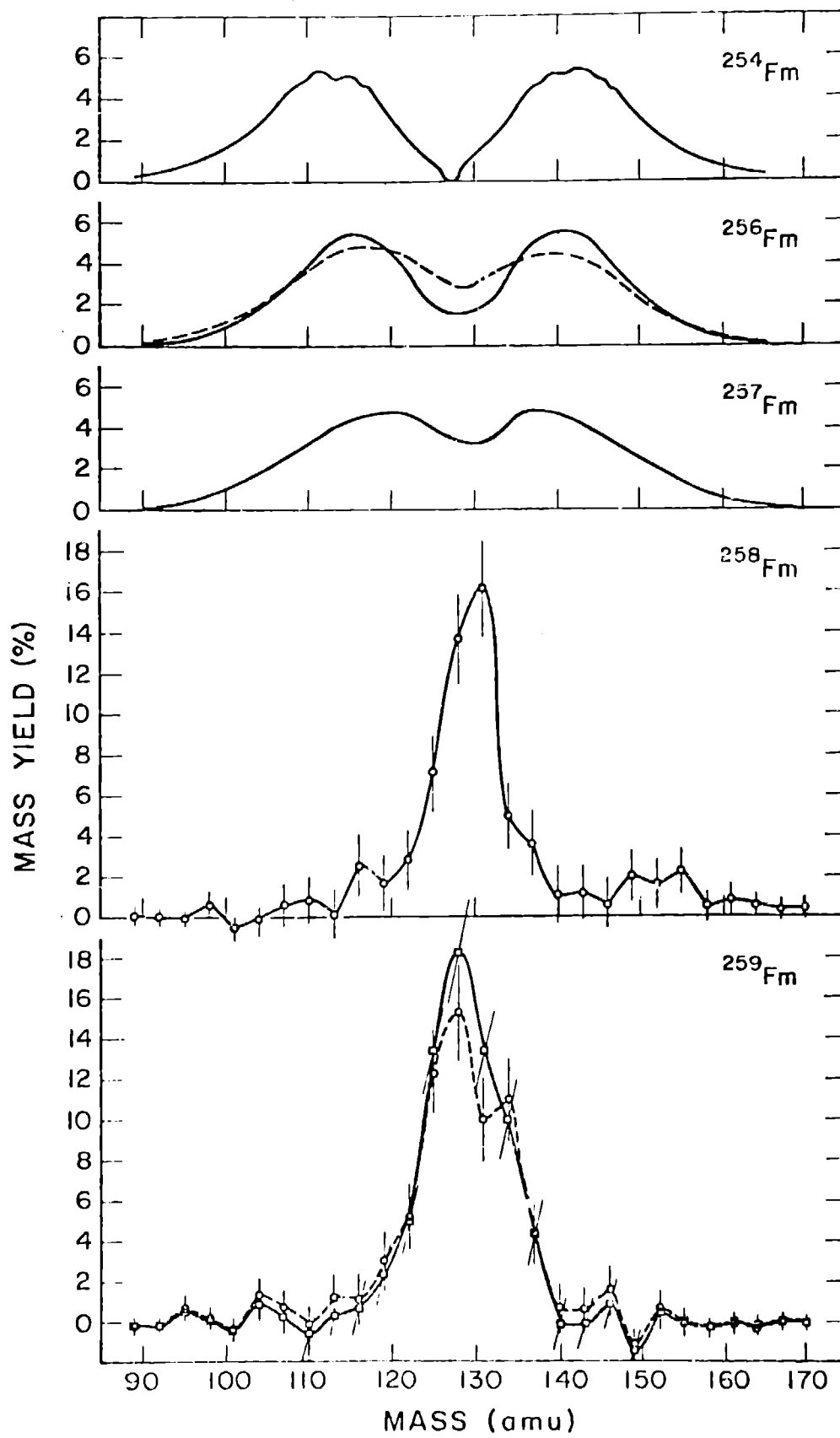


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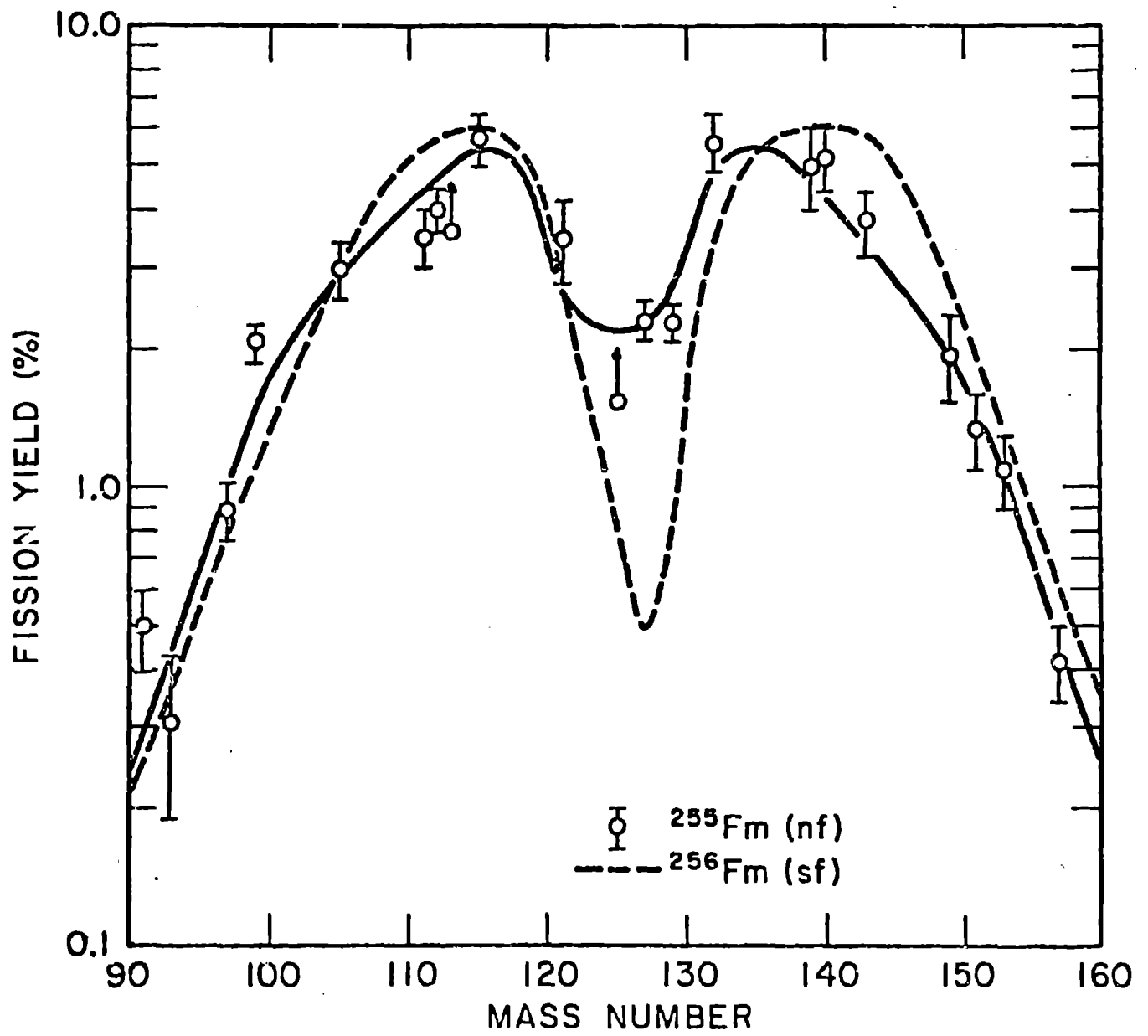


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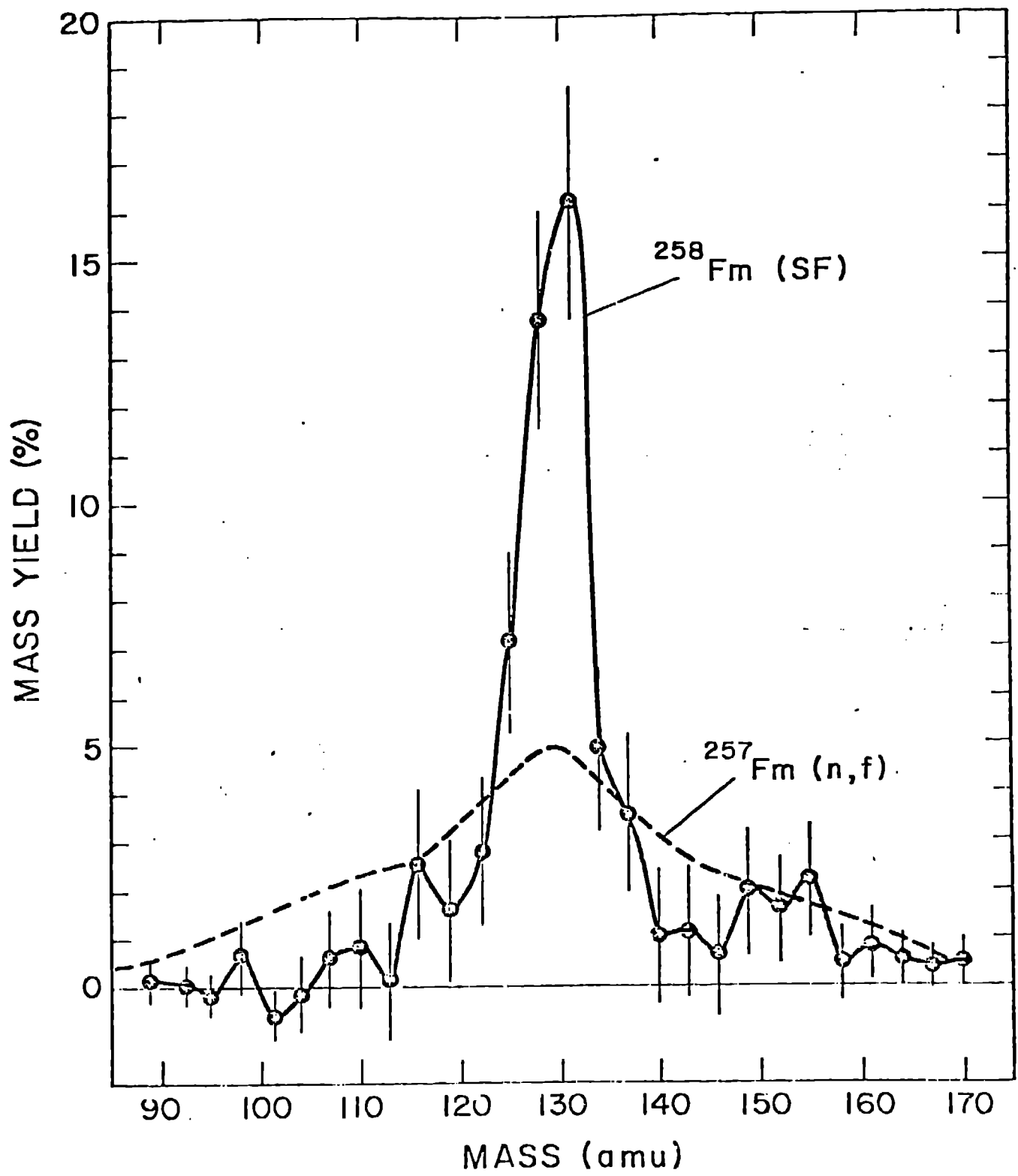


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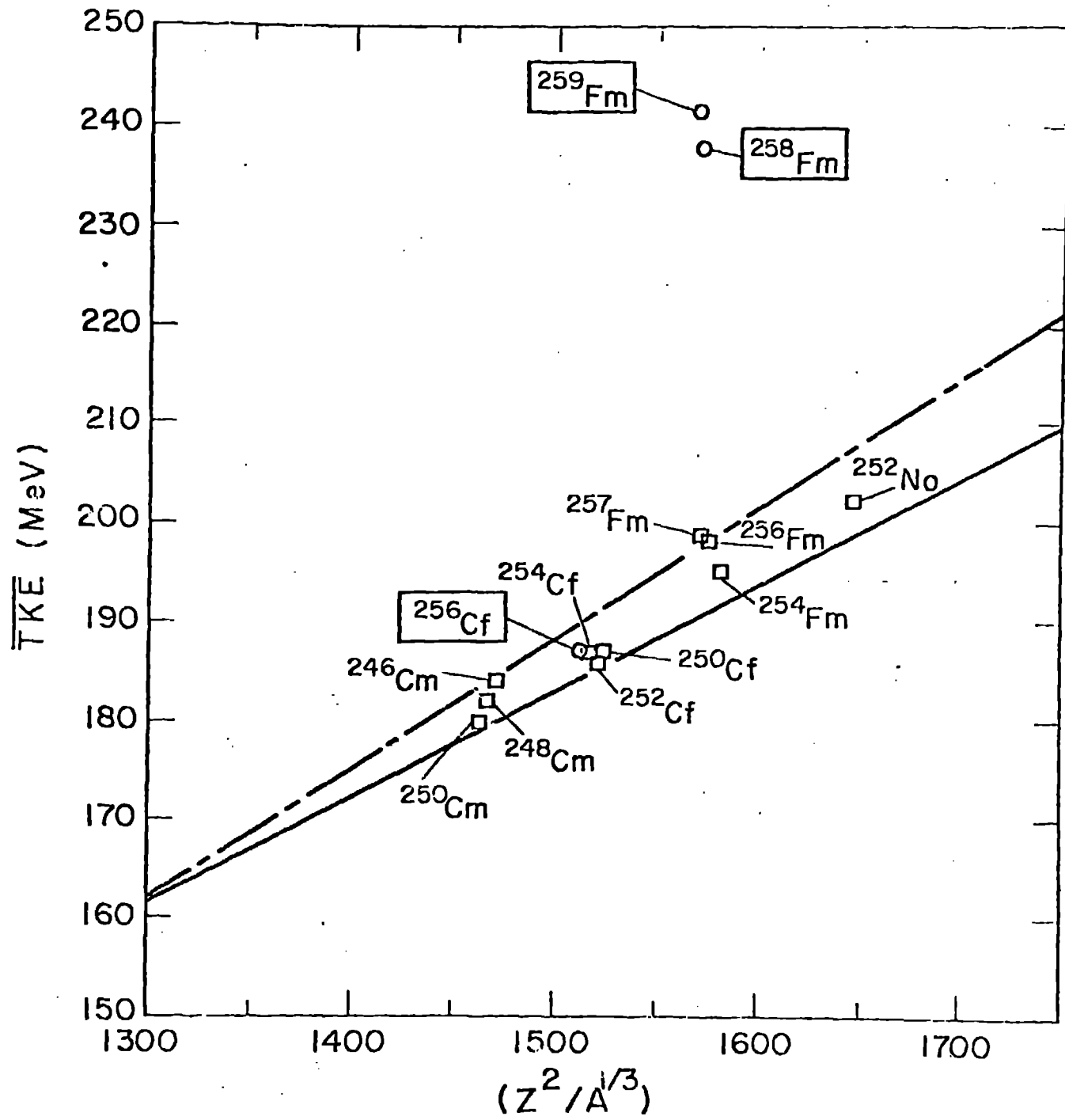


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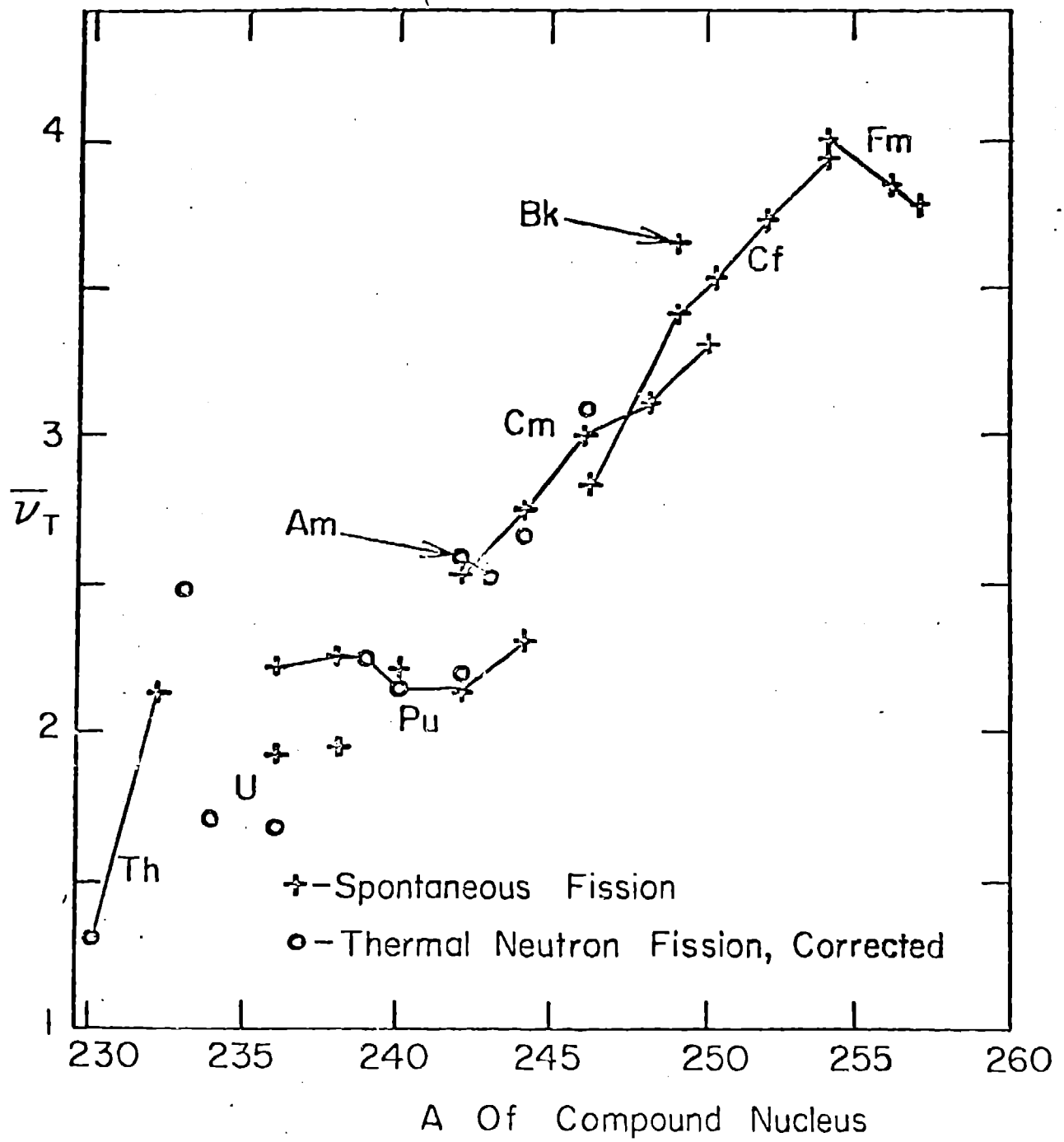


Figure 9.

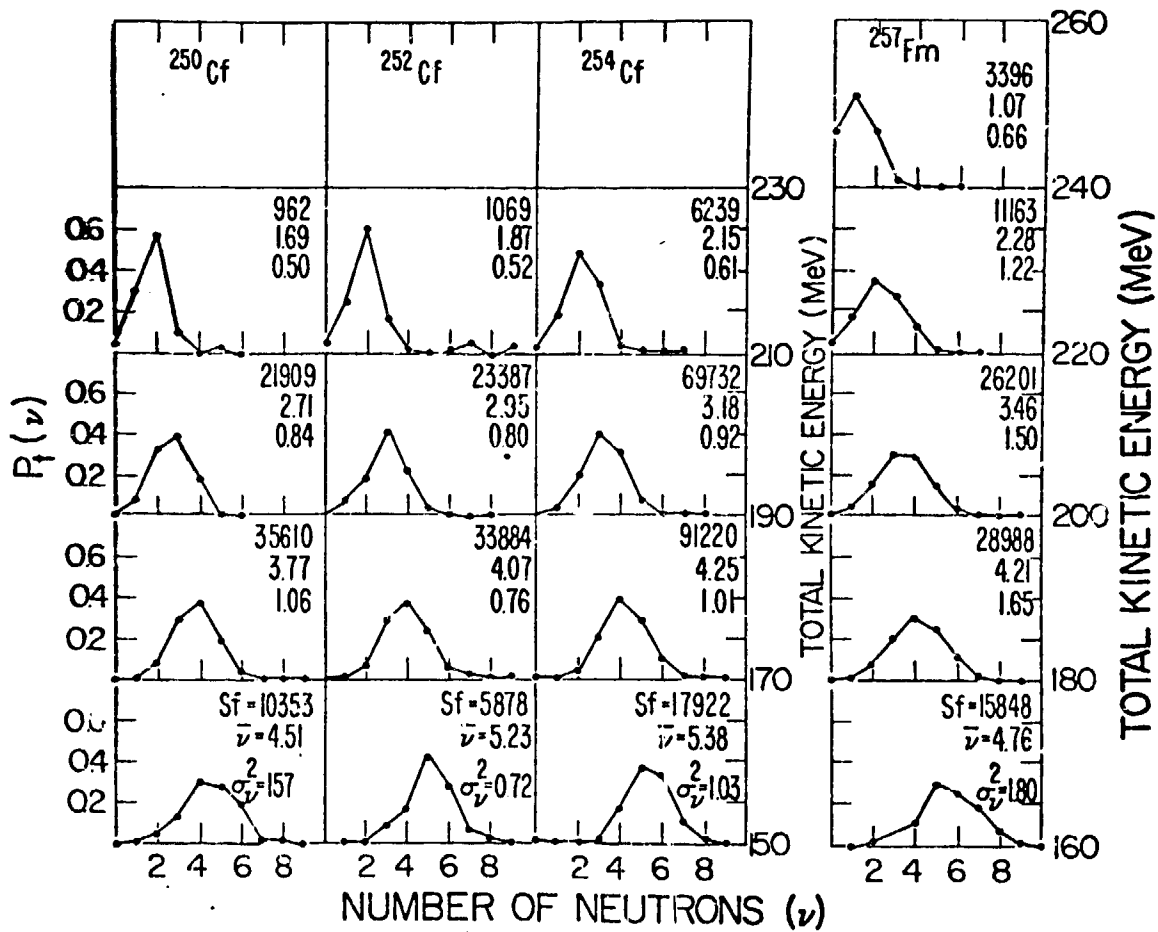
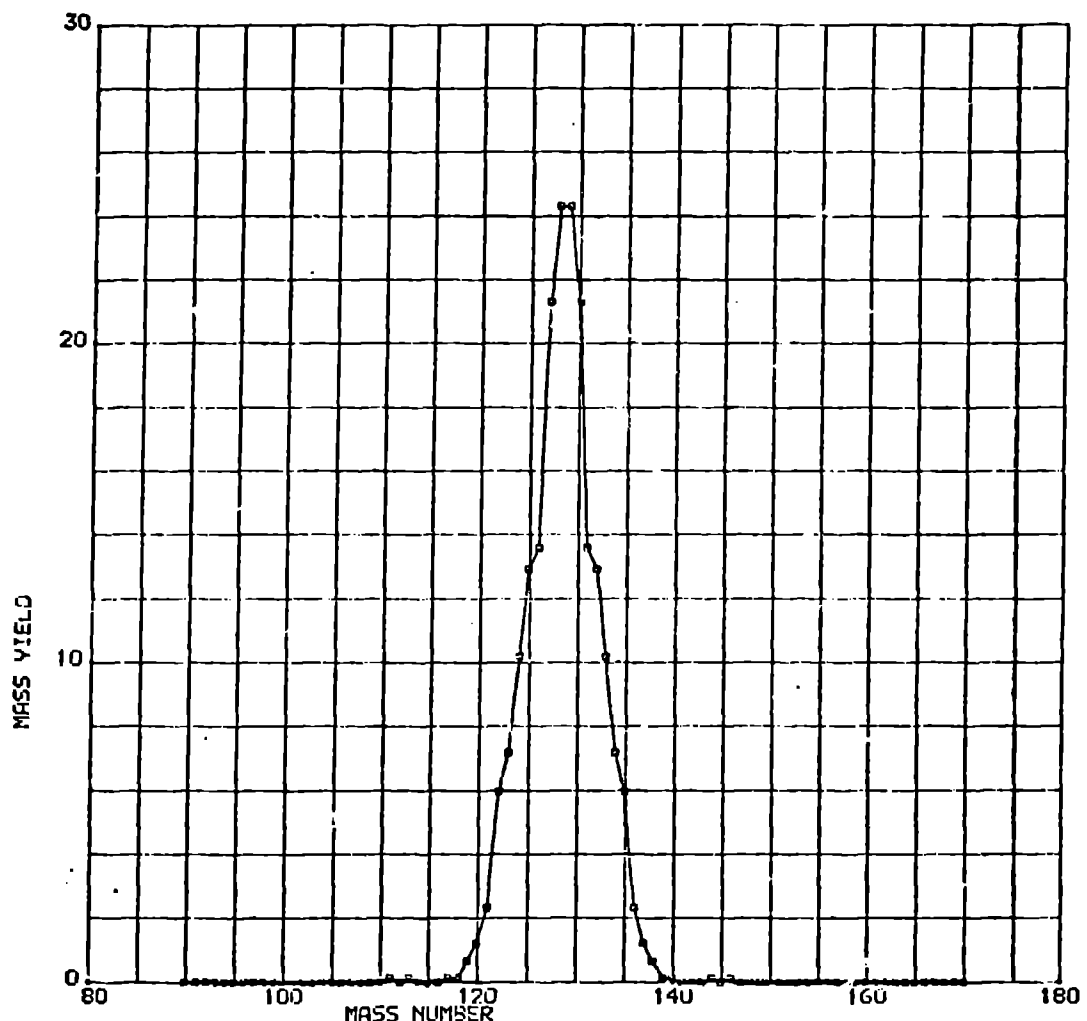


Figure 10.



257FM PRE-NEUTRON TOTAL KINETIC ENERGY GREATER THAN 235 MEV.  
 MASS YIELD

Figure 11.