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RESULTS FROM HEAVY-ION  
BOMBARDMENTS OF  $^{254}\text{Es}$

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# RESULTS FROM HEAVY-ION BOMBARDMENTS OF $^{254}\text{Es}$

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## 1. INTRODUCTION

Near to the limit of nuclei that can be made by intense neutron irradiations of lighter actinides,  $^{254}\text{Es}$  is the heaviest isotope producible in microgram amounts. When used as a target isotope for heavy-ion reactions, fewer nucleons need be added to  $^{254}\text{Es}$  to form unknown nuclei in a neutron-rich region lying above it on the nuclide chart. Much of this region is inaccessible by any other projectile/target combination. Because of these favorable factors, we have intensively explored the possibility of making new nuclides from heavy-ion reactions with  $^{254}\text{Es}$ . The nuclear reactions include transfer, incomplete- and complete-fusion with projectiles ranging from tritons to  $^{136}\text{Xe}$ . One of the major results has been the discovery of six new isotopes and an isomer of  $^{258}\text{Md}$  within the past 15 years, with four,  $^{260}\text{Md}$ ,  $^{261}\text{Lr}$ ,  $^{262}\text{Lr}$ , and  $^{262}\text{No}$  being found in the last few years. A portion of the nuclide chart, illustrated in Fig. 1, shows all the nuclides discovered from bombardments of Es targets shaded in gray. In addition, searches have been made for  $^{263}\text{[105]}$ ,  $^{264}\text{[105]}$ ,  $^{272}\text{[109]}$ , and superheavy elements, with these nuclide products shown in Fig. 1 in gray outline.

The potential for producing new nuclides from transfer reactions of  $^{18}\text{O}$  and  $^{22}\text{Ne}$  with  $^{254}\text{Es}$  were first investigated in 1981 and 1982. In these preliminary experiments, we measured the cross sections for the formation of a series of isotopes of Es, Fm, and Md by radiochemical methods. To extend these cross section measurements to shorter-lived isotopes of Md, No and, Lr, we employed a He-jet transfer system at the Gesellschaft für Schwerionenforschung, Darmstadt. The cross sections obtained from the He-jet experiments were normalized to those of  $^{255}\text{Fm}$  measured by the radiochemical methods. These transfer cross sections,<sup>1</sup> including ones for nuclides that have been recently discovered, are given in Figs. 2 and 3. Considering that the shapes of the isotopic yield curves are Gaussian for each element, it was clear from extrapolations of these curves to larger mass numbers that a number of new, neutron-rich isotopes of Md, No, and Lr could be produced with cross sections larger than 10 nb. However, transfer cross sections for producing any isotope of element 104 are expected to be below this level and, from all our studies, we have no evidence for their formation. Given that we could now predict the production cross section of an unknown nuclide with less than 20 to 30% uncertainty, the principal restraints to identifying new isotopes in this region were largely experimental. Because of the large (millibarns) cross sections for forming known nuclides near the target isotope, it was necessary to devise rapid separation methods in order to isolate new nuclides having much lower formation cross sections. Otherwise, the background radiation from known nuclei would overwhelm that from the unknown. No less important a problem

involves estimates of the half-lives and decay properties for the undiscovered nuclides in this region. Half-lives for  $\alpha$ ,  $\beta^-$ , and electron-capture (EC) decay can be predicted to within a factor of 2 to 3, but spontaneous fission, which is often the most competitive decay mode, has factors of 100 to  $10^5$  uncertainties in predicted half-lives. To improve the predictions, further experimental half-lives are needed but, on the other hand, obtaining spontaneous fission half-lives for new isotopes without the benefit of better estimates is mostly a blind search.

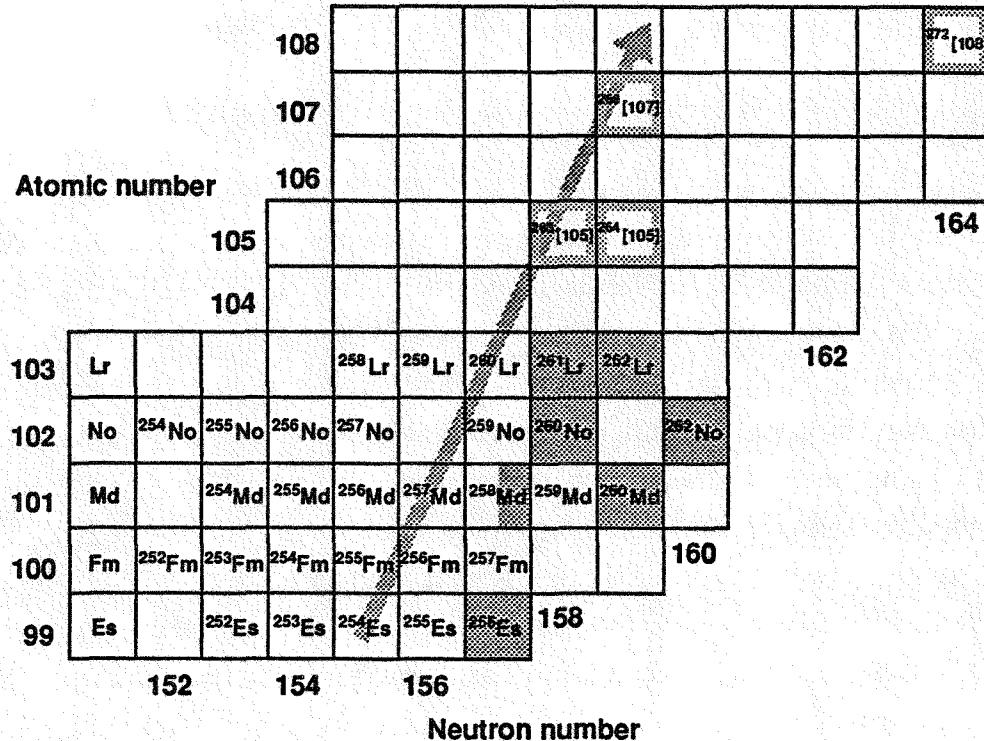


FIG. 1. A portion of the nuclide chart showing the isotopes produced in bombardments of  $^{254}\text{Es}$ . Those shaded in gray were discovered in  $^{254}\text{Es}$  bombardments and those enclosed in a border represent attempts to identify their formation. The long arrow indicates the peak of mass flow in transfer reaction.

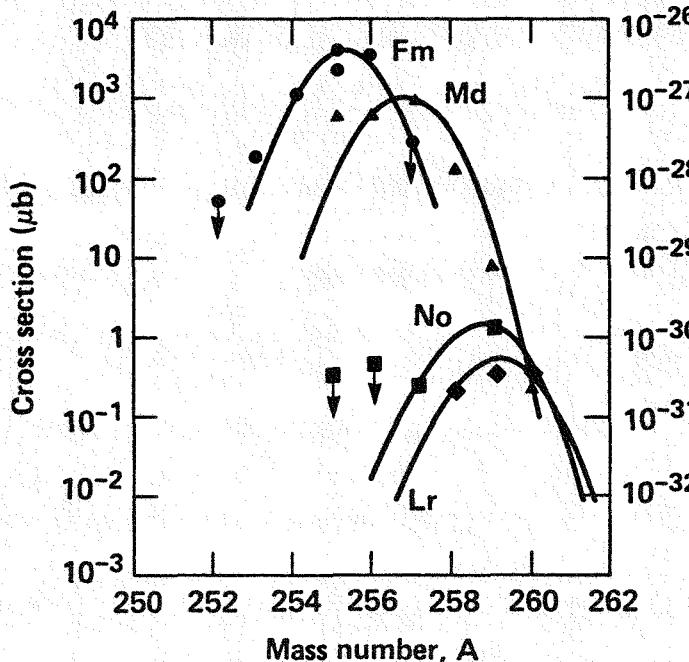


FIG. 2. Isotopic distributions measured for 105-MeV  $^{18}\text{O}$  reactions with  $^{254}\text{Es}$ . Cross sections taken from Ref. 1.

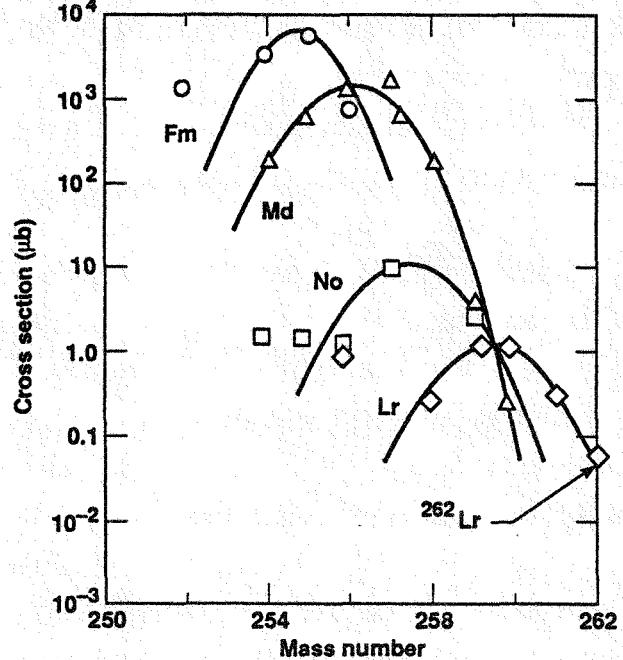


FIG. 3. Isotopic yield distributions measured for 125 and 126 MeV  $^{22}\text{Ne}$  reactions with  $^{254}\text{Es}$ . Cross sections from Ref. 1 with the addition of those for  $^{261}\text{Lr}$  and  $^{262}\text{Lr}$ .

In the following section, we report our experimental work and offer our most recent data on the decay properties for five new isotopes of Md, No, Lr, and for  $^{258m}\text{Md}$ . In addition to these successful experiments, we have also conducted searches for  $^{263}$ [105],  $^{264}$ [105],  $^{272}$ [109], and superheavy elements,<sup>2</sup> which are described in a subsequent section. The final part summarizes our conclusions based on what we have learned so far and what we believe are the most critical directions that should now be taken.

## 2. NEW ISOTOPES OF Md, No, AND Lr

### a. $^{258m}\text{Md}$

The first indications of an isomer of  $^{258}\text{Md}$  came from bombardments of  $3 \times 10^{11}$  atoms of  $^{255}\text{Es}$  with 26.3-MeV  $\alpha$  particles.<sup>3</sup> A spontaneous-fission (SF) activity decaying with a 43-min half-life was detected in the products of these bombardments. However, the amount produced was too low to chemically identify the source of the SF activity, but it was assumed on the basis of estimated Q values that  $^{258m}\text{Md}$  would decay mainly by EC to 370- $\mu\text{s}$   $^{258}\text{Fm}$ , which was known to decay by SF. These earlier results have been largely confirmed from our bombardments of  $^{254}\text{Es}$  with both  $^{18}\text{O}$  and  $^{22}\text{Ne}$  ions in which our production rate was about 4500 times greater than with the much smaller  $^{255}\text{Es}$  target.<sup>4</sup> From this new study, the half-life found for  $^{258}\text{Md}$  from following the decay of eight mass-separated samples was  $57 \pm 1$  min. Because these sources were prepared by electromagnetic mass separation, the mass number of the SF activity was established. Furthermore, we have determined with certainty the element identification of 370- $\mu\text{s}$   $^{258}\text{Fm}$  and the mother-daughter genetic relationship between  $^{258m}\text{Md}$  and  $^{258}\text{Fm}$  by the observation of time-correlated Fm K x-rays preceding SF by  $^{258}\text{Fm}$ .<sup>4</sup> The distribution of time intervals that we obtained is shown in Fig. 4. The larger peak near 290 ms is due to random background photons that happen to fall within our photon-energy window. The peak at 520  $\mu\text{s}$  corresponds to the lifetime of  $^{258}\text{Fm}$  following its formation by EC decay of  $^{258m}\text{Md}$  and equals a half-life of  $360 \pm 20$   $\mu\text{s}$ .

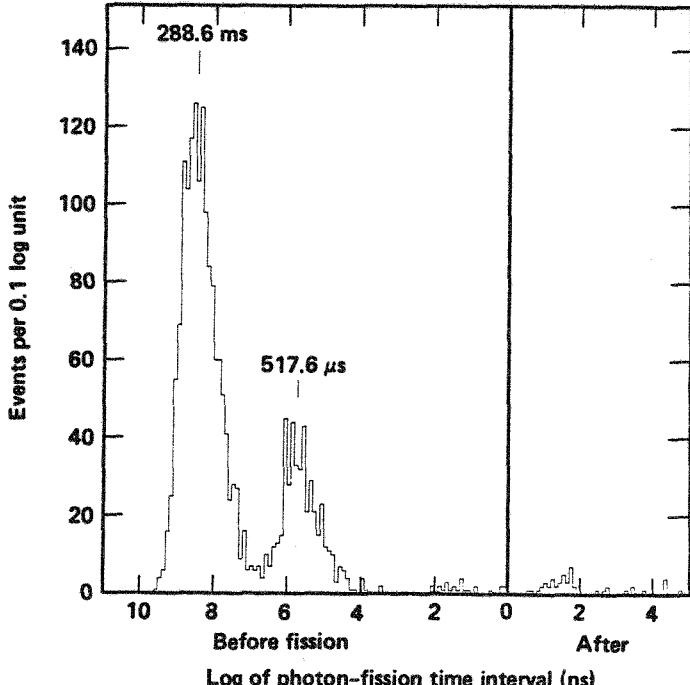


FIG. 4. Logarithmic time distributions for the last energy-windowed photon detected before SF of  $^{258}\text{Fm}$ . The photon energy window corresponds to the K x-ray region of Fm (112--145 keV). In this figure, the occurrence of fission is defined as zero time. The distribution around 520  $\mu\text{s}$  before fission is the lifetime of  $^{258}\text{Fm}$ .

b.  $^{260}\text{No}$ 

A SF activity with a half-life of  $106 \pm 8$  ms was produced with a cross section of  $1.1 \pm 0.2$   $\mu\text{b}$  in reactions of 99-MeV  $^{18}\text{O}$  ions with  $^{254}\text{Es}$ .<sup>5</sup> Activities with a similar half-life were made with cross sections of  $\leq 80$  and 200 nb in bombardments of  $^{254}\text{Es}$  with 125-MeV  $^{22}\text{Ne}$  ions and 73-MeV  $^{13}\text{C}$  ions, respectively. Because the formation cross section from the reaction  $^{18}\text{O} + ^{254}\text{Es}$  was consistent with an extrapolation of the No yield curve to mass 260, as can be seen in Fig. 2, it was suggested this new SF activity was associated with the decay of  $^{260}\text{No}$ . From recently compiled cross sections for 72-MeV  $^{13}\text{C}$  transfer reactions with  $^{254}\text{Es}$  (Fig. 5),<sup>6</sup> we conclude the 200-nb cross section reported by Somerville *et al.* for possibly making  $^{260}\text{No}$  from  $^{13}\text{C}$  appears an order of magnitude too large. Similarly, the same might be said for their  $^{22}\text{Ne}$  transfer reaction, where the upper limit for the cross section seems a factor of three to four too low. Therefore, we feel the assignment of this 100-ms activity is on shaky grounds, but it is still difficult to imagine any other alternative, especially since we have identified most of the surrounding nuclides. The large 1.1  $\mu\text{b}$  cross section from  $^{18}\text{O}$  reactions with  $^{254}\text{Es}$  precludes isomers or nuclei more distant than  $^{260}\text{No}$ . Everything considered, we would judge the assignment to be probable but, at the first opportunity, new experiments to prove the isotopic source of this 100-ms activity should be made.

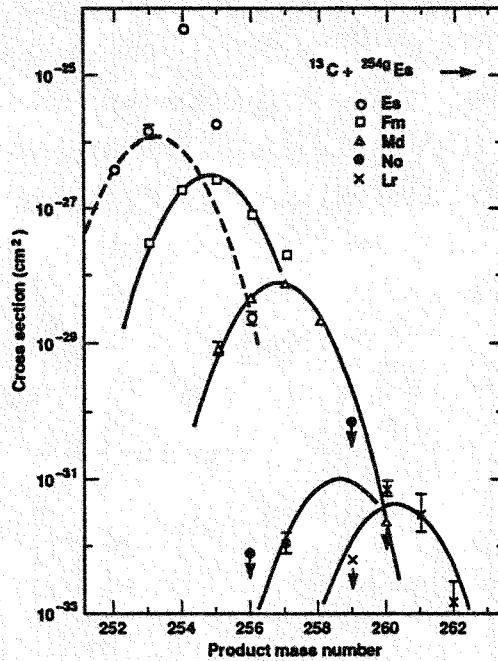


FIG. 5. Isotopic yields from transfer reactions in 72-MeV  $^{13}\text{C}$  bombardments of  $^{254}\text{Es}$  (Ref. 6).

c.  $^{260}\text{Md}$ 

From bombardments of  $^{254}\text{Es}$  with both  $^{18}\text{O}$  and  $^{22}\text{Ne}$  ions, we discovered  $^{260}\text{Md}$  in isotopically separated samples of the reaction products.<sup>7</sup> After collecting products recoiling from the target on Ta foils, we transported the foils from the 88-in cyclotron at the Lawrence Berkeley Laboratory to the Lawrence Livermore National Laboratory for off-line electromagnetic separation. The elapsed time from the end of bombardment until counting started after isotope separation was close to an hour. The mass-260 fraction decayed by spontaneous fission (SF) with a  $27.8 \pm 0.8$  day half-life. This same SF activity has been chemically identified many times over as belonging to the element Md. In view of the Q values for  $\beta^-$  decay of up to 1 MeV and, for EC, in the range of 0.5 to 1.4 MeV, we initially believed the SF activity probably originated from the possible daughters,  $^{260}\text{No}$  or  $^{260}\text{Fm}$ , in equilibrium with the

29-d parent. To check these possibilities, we constructed two specialized counting systems to measure the time intervals between  $\beta^-$  particles, K x-rays or L x-rays and a subsequent fission event. Because both daughter nuclides were predicted to have SF lifetimes of well under 1 s, these counting systems were designed to cover time intervals from about 10 ns up to 1 s. Maintaining the maximum geometrical efficiency for each type of radiation was necessary because only about 1500 atoms of  $^{260}\text{Md}$  could be prepared per bombardment. Total efficiencies, including geometry and detector efficiencies, were 60% for  $\beta^-$  particles up to 0.7 MeV, 34% for K and L x rays and, for fission fragments, 61% in the  $\beta^-$  system and ~90% in the x-ray system. In the  $\beta^-$  system, surface-barrier (Au) detectors depleted to a depth of 1 mm were used to detect and measure the energies of both  $\beta^-$  particles and fission fragments. In a separate counting system, K- and L-x-ray energies were measured with intrinsic-Ge detectors with a resolution averaging 1.1 keV between 100 and 150 keV. We made two to three bombardment to produce  $^{260}\text{Md}$  for each of the three counting experiments.

Analysis of our time-correlated data was complex because of background radiation whose random time distribution before fission partially overlapped the 100-ms lifetime region of potential daughters. Only in the case of  $\beta^-$  decay was the background rate sufficiently low to not interfere with detecting a SF daughter having a half-life of 106 ms, the period expected for  $^{260}\text{No}$  (see above). From these experiments, our  $2\sigma$  upper limit for a possible  $\beta^-$ -decay branch by  $^{260}\text{Md}$ , assuming a daughter lifetime of 100 ms, is 7.2%. The statistical analysis of the time intervals and photon energy data for possible K- or L-electron capture is incomplete, but the probabilities for a decay branch by either of these modes appears to be less than 10%. There was no obvious excess of photons above background with Fm K $_{\alpha 1}$  or K $_{\alpha 2}$  energies occurring 100 ms before fission and the same was also true for Fm L x-rays. Thus, we are assured that most if not all of the SF activity we observed in the mass-260 fractions arises directly from the SF decay of  $^{260}\text{Md}$ .

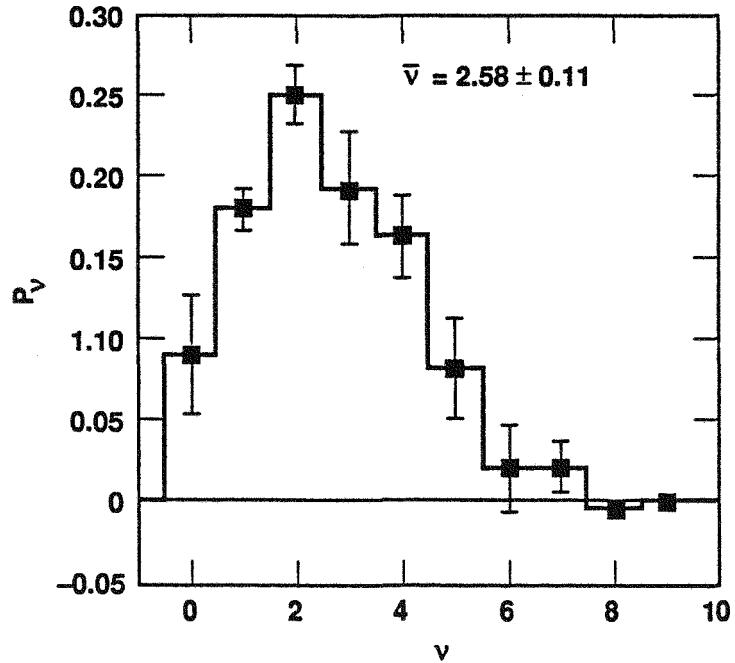


FIG. 6. Total neutron multiplicity distribution from the SF of  $^{260}\text{Md}$ , corrected for background, dead time, and counter efficiency.

During the search for possible daughters, we also measured correlated fission fragment energies and have derived the mass and kinetic-energy distributions for SF by  $^{260}\text{Md}$ .<sup>8</sup> Altogether, we have determined the mass and kinetic-energy distributions from the SF of  $^{258}\text{Fm}$ ,  $^{259}\text{Md}$ ,  $^{260}\text{Md}$ ,  $^{258}\text{No}$ ,  $^{262}\text{No}$ , and  $^{260}\text{[104]}$ .<sup>8,9</sup> All except for  $^{260}\text{[104]}$  fission by two distinct modes (bimodal) with each mode having about equal abundance. The reader is referred to Ref. 8 for a further description of this unusual fission process. Because of the uncommon total-kinetic-energy (TKE) distribution, we measured the neutron multiplicity from the fission of  $^{260}\text{Md}$  in collaboration with colleagues at Philipps University, Marburg, FRG.<sup>10</sup> The measured neutron-multiplicity distribution is shown in Fig. 6 where it is clear there are several fewer neutrons emitted per fission by this isotope than by  $^{252}\text{Cf}$ . The large drop in the number of neutrons emitted is due to cold fragmentation in fission, in which there is little excitation energy left in the fragments after scission to overcome the neutron binding energy. The high-energy mode of fission in  $^{260}\text{Md}$ , averaging 234 MeV, leaves only  $\sim 21$  MeV of excitation to be divided between the two fragments.

#### d. $^{261}\text{Lr}$ and $^{262}\text{Lr}$

Experiments conducted in 1984 gave us the first indication of the existence of  $^{261}\text{Lr}$  and  $^{262}\text{Lr}$  from the presence of excess SF activity in the mass-261 and -262 fractions, following electromagnetic separation of the recoil products from  $^{22}\text{Ne}$  bombardments of  $^{254}\text{Es}$ . These fission activities decayed with roughly a 1-h half-life and disappeared entirely after about 4 hours. The cross sections and decay periods were consistent with those expected for new Lr isotopes, as illustrated in Fig. 7. With this evidence in hand, we later performed a new series of bombardments to chemically separate and identify the source of these activities. Because of the 1000-fold larger cross sections for making 2.6-h  $^{256}\text{Fm}$ , an interfering SF activity, it was necessary to perform two cation-exchange-column separations to achieve complete decontamination of the Lr isotopes. We made five bombardments ranging in length from 2 to 3 h, resulting in a new 39-min SF activity being found in the chemically separated Lr fractions. Decay analysis of the SF activity from these bombardments indicated there were two activities present, as shown in Fig. 8a. To confirm that the long-lived component was also a lawrencium activity and not from SF by  $^{256}\text{Fm}$ , we performed three longer bombardments ranging from 5 to 21 h. The decay curve from one of these bombardments, shown in Fig. 8b, yielded a half-life of  $212 \pm 18$  min, while the average from the three experiments was 216 min. Later work indicated a half-life closer to 4 h.

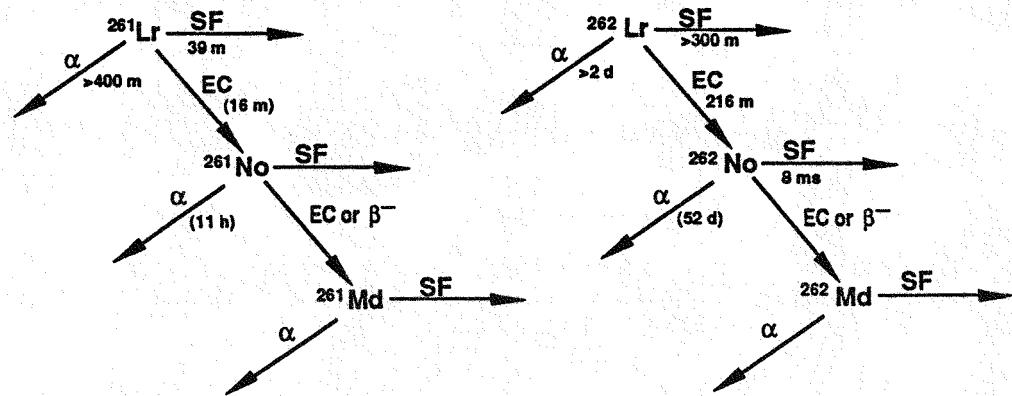


FIG. 7. Decay properties we estimate for  $^{261}\text{Lr}$  and  $^{262}\text{Lr}$  but with the inclusion of our measured values for the SF half-life of  $^{261}\text{Lr}$  and the EC half-life of  $^{262}\text{Lr}$ .

The assignment of the 39-min activity to direct SF by  $^{261}\text{Lr}$  rather than to its EC daughter,  $^{261}\text{No}$ , is tenuous. Recent Q-value estimates for EC decay of  $^{261}\text{Lr}$  span from 0.8 to 1.3 MeV. From the most reliable of these estimates, we would judge the EC half-life to be  $\sim$ 16 min. To test this possibility, we performed a series of chemical milking experiments in which nobelium was separated at 10-min intervals from a sample of purified lawrencium. No SF events were found in the No samples and, from this result, we calculate that the half-life for SF by  $^{261}\text{No}$ , arising from the EC decay of  $^{261}\text{Lr}$ , is less than 10 min. Unfortunately, a SF half-life for  $^{261}\text{No}$  shorter than 10 min is still within expectations. Without the observation of  $^{261}\text{No}$ , we were unable to demonstrate that the 39-min half-life was associated with EC decay by  $^{261}\text{Lr}$ . As noted below, our efforts to establish the decay mode for  $^{262}\text{Lr}$  were more successful.

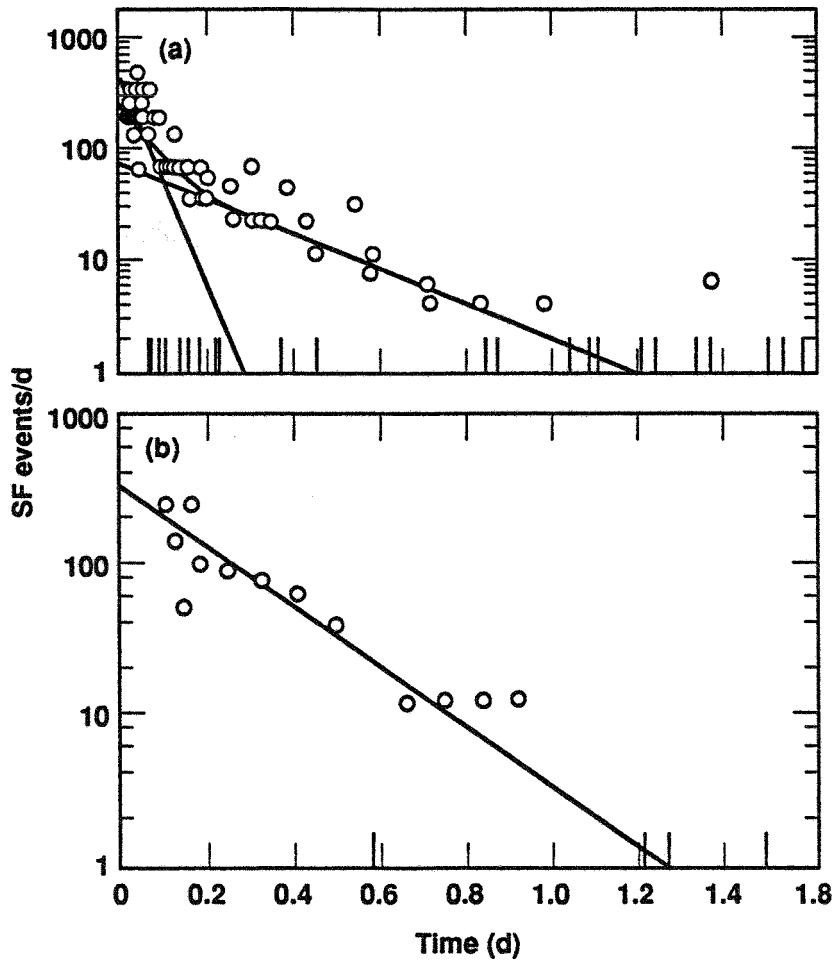


FIG. 8. (a) Composite SF decay curve from chemically isolated Lr produced from four, 2-h bombardments of  $^{254}\text{Es}$  with 127-MeV  $^{22}\text{Ne}$  ions. A 39-min and a 200- to 300-min component were observed. (b) Decay curve for the longer-lived SF activity found in Lr fractions from a 10-h bombardment.

#### e. $^{262}\text{No}$

Estimated Q values for EC decay by 4-h  $^{262}\text{Lr}$  are about 2 MeV, making EC decay to  $^{262}\text{No}$  a definite possibility. Because SF by  $^{262}\text{No}$  is unhindered by odd nucleons, its SF half-life was predicted to be as short nanoseconds or as long as a fraction of a second. Therefore, the 4-h decay period ascribed to  $^{262}\text{Lr}$  might well represent EC decay by  $^{262}\text{Lr}$  to produce  $^{262}\text{No}$ , with the later being in secular equilibrium with its much longer-lived Lr parent. Thus, our experimental goals were to determine if  $^{262}\text{Lr}$  decayed by EC and to measure the half-life and SF properties of it or its daughter,  $^{262}\text{No}$ , by measuring the time intervals between nobelium K x-rays and subsequent SF events in samples of chemically purified lawrencium. For this purpose, we used the same specially constructed, high-

geometry counter and attendant electronic and computer systems as were employed in searching for EC decay by  $^{260}\text{Md}$  and in the successful finding of this decay mode for  $^{258\text{m}}\text{Md}$ .<sup>4</sup>

A series of 38 bombardments, lasting from 2 to 9 h, of a 57- $\mu\text{g}\cdot\text{cm}^{-2}$  target of  $^{254}\text{Es}$  with 127-MeV  $^{22}\text{Ne}$  ions was made to produce  $^{262}\text{Lr}$ . Recoil products caught on a Ta foil were chemically processed to remove fission products and nearby actinides from lawrencium. The pure Lr fraction was then electroplated on 27- $\mu\text{g}\cdot\text{cm}^{-2}$  polyimide foils that were overcoated with 25  $\mu\text{g}\cdot\text{cm}^{-2}$  of gold. The foils were placed between two large, unmounted surface-barrier detectors in a vacuum chamber with beryllium windows placed directly behind the surface-barrier detectors. Two large, intrinsic germanium detectors were inserted in back of these windows for detecting photons in the No K x-ray region.

The mean lifetime of  $^{262}\text{No}$  and confirmation of  $^{262}\text{Lr}$  EC decay to  $^{262}\text{No}$  were derived from the distribution of logarithmic time intervals between photons having No K x-rays and subsequent fission events as shown in Fig. 9. Two distributions, which follow Poisson statistics, are resolved in the figure. The larger peak, with a mean lifetime of 1480 ms, is due to background photons preceding a fission event. We attribute the smaller peak at 6 ms to the mean lifetime of  $^{262}\text{No}$  and some tailing of background events.<sup>11</sup> From the distribution of time intervals, we calculate a weighted average of 5 ms for the SF half-life of  $^{262}\text{No}$ .

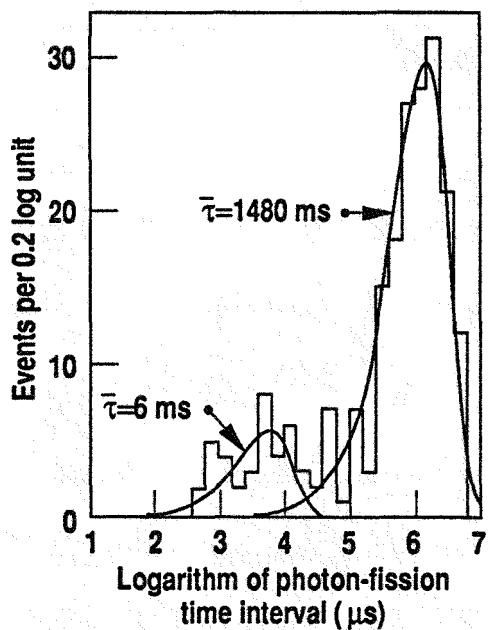


FIG. 9. The logarithmic distribution of time intervals between the last photon with No K x-ray energies preceding a SF event. The smooth curves are exponential fits to the measured data shown by the histogram.

Correlated fission-fragment energies were obtained from the surface-barrier detectors during these experiments. From these, fragment mass and kinetic-energy distributions were calculated for  $^{262}\text{No}$ , as shown in Figs. 10 and 11. The mass distribution is sharply symmetrical but the TKE distribution is highly asymmetrical, peaking at 235 MeV, with a lower-energy component at 199 MeV. These features are characteristic of bimodal fission and are very similar to those found for  $^{258}\text{Fm}$  and  $^{260}\text{Md}$ .<sup>8</sup> Observation of such an appreciable amount of the high-energy component is significant because this mode is thought to arise from the favorable division into  $Z=50$ , closed-shell Sn isotopes that are also near the 82-neutron closed shell. However, this nuclide lies two atomic numbers away from Fm, so the proton division is not optimum. To account for the abundance of the high-energy mode, we believe the

fragments' approach to the 82-neutron shell compensates for the unfavorable proton division. We might, then, expect heavier nuclei, provided they are neutron-rich, to feature a significant "cold-fission" mode.

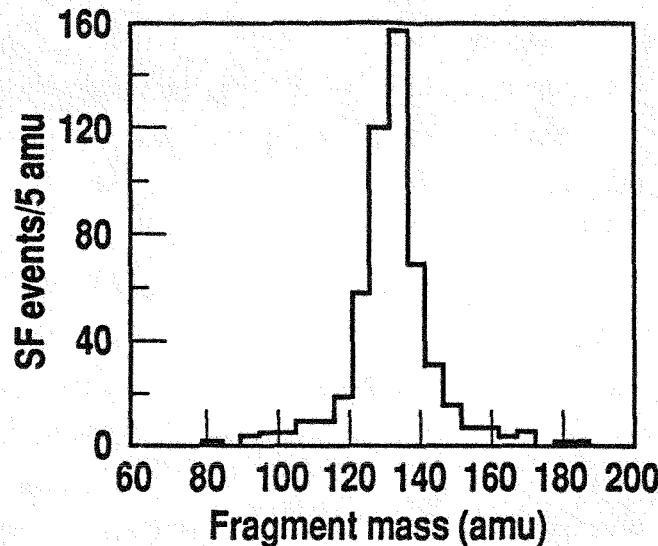


FIG. 10. Provisional mass distribution obtained for 5-ms  $^{262}\text{No}$ .

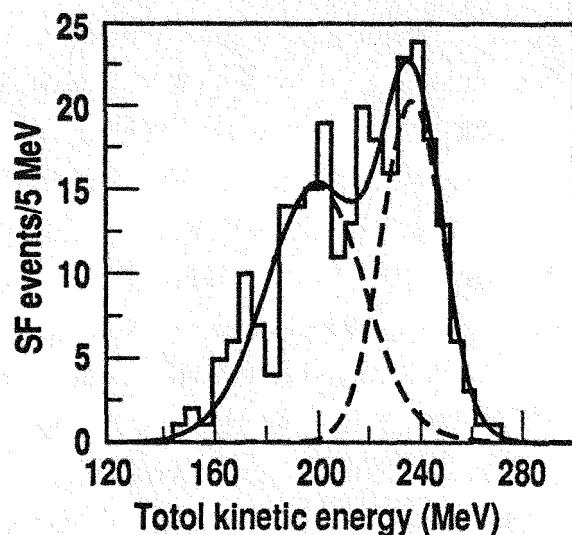


FIG. 11. Total-kinetic-energy distribution from the spontaneous fission of  $^{262}\text{No}$ . Dashed curves are unfolding of the TKE distribution into two Gaussians by least-mean-squares fitting.

### 3. ATTEMPTS TO IDENTIFY ISOTOPES OF ELEMENTS 105 AND 109

#### a. $^{263}[105]$

Targets of  $^{254}\text{Es}$  offer one of the very few possibilities of extending our knowledge concerning the limits of nuclear stability beyond the border of the known nuclei toward the neutron-heavy region. Accordingly, we have performed a number of experiments aimed at identifying  $^{263}[105]$  and one attempt to detect  $^{264}[105]$ . Figure 12 shows our recent estimates of the partial half-lives for possible decay paths. Those for  $\alpha$  and  $\beta$  decay were derived from a set of mass prediction and level schemes prepared by Moody.<sup>12</sup> Our estimates of the SF half-lives were made from extrapolations of systematic trends.

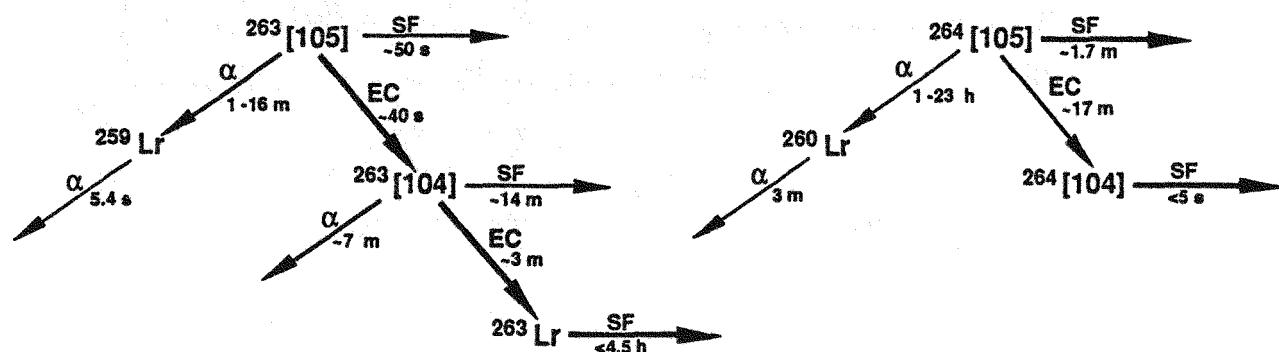


FIG. 12. Half-lives predicted from mass tables and SF systematics for the decay chains originating from  $^{263}[105]$  and  $^{264}[105]$ . Dominate branches are indicated by the heavier arrows. The SF estimates are very uncertain.

Our first experiments to detect  $^{263}[105]$  were based upon the expectation that  $\alpha$  decay would be the dominant decay path. In those experiments, we bombarded  $^{254}\text{Es}$  with 98.5-MeV  $^{16}\text{O}$  ions to produce  $^{263}[105]$  by an incomplete fusion reaction and searched for a mother-daughter sequence of  $\alpha$  decays.<sup>13</sup> Within a 95% confidence limit, such a sequence was not observed in the half-life range of 2 s to tens of

minutes, provided the formation cross section was greater than 5 nb. This cross section is well below the 15 to 20 nb we expected from  $(\text{Hf},\alpha\text{3n})$  reactions (25 to 30 nb for heavy No and Lr isotopes).

Still believing that  $\alpha$  emission would be the main decay mode, we made a further attempt at searching for  $\alpha$  decay by  $^{263}\text{Lr}$ [105]. The strategy was the same, namely, looking for  $\alpha$  decay of its daughter, the known isotope 4-s  $^{259}\text{Lr}$ , within 20 s after observing an  $\alpha$  particle in the appropriate energy range from decay of the parent  $^{263}\text{Lr}$ [105]. However, the reaction to produce  $^{263}\text{Lr}$ [105] was changed to one of complete fusion by using 71.8-MeV  $^{13}\text{C}$  ions as the projectile. After bombarding with a total of  $1.07 \times 10^{17}$  particles of  $^{13}\text{C}$ , we found no evidence for  $\alpha$  decay by  $^{263}\text{Lr}$ [105] to  $^{259}\text{Lr}$  within a half-life range of several seconds to tens of minutes. No mother/daughter pairs of events were observed in the case where the daughter was deposited on the face of an opposing detector by recoil from the  $\alpha$  particle emitted by the parent. In the case where the daughter did not leave the sample by recoil, the background from energy-degraded fission fragments essentially equaled the number of pairs of time-correlated events with  $\alpha$  energies within our preset range. Assuming the observation of three pairs, we established an upper limit for the formation cross section of 2.7 nb, whereas we would expect a 6 to 10 nb cross section from similar  $(^{13}\text{C},4\text{n})$  reactions.

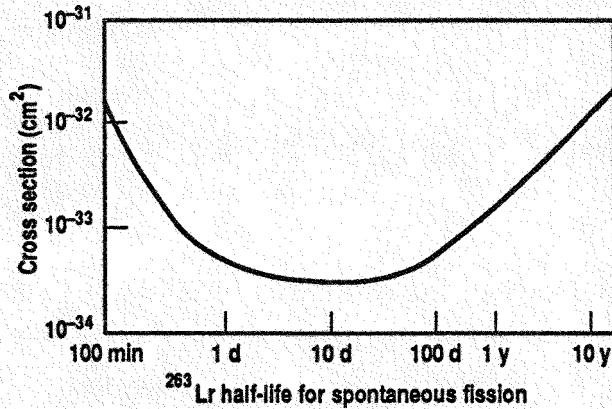


FIG. 13. Upper limits for the formation cross section of  $^{263}\text{Lr}$ [105] as a function of the SF half-life of  $^{263}\text{Lr}$ . The curve was derived on the basis of  $^{263}\text{Lr}$ [105] decaying by two electron-capture steps to  $^{263}\text{Lr}$ .

Having ruled out  $\alpha$  emission as a major decay mode for  $^{263}\text{Lr}$ [105] and now finding that EC seemed more likely (Fig. 12), we performed an experiment to search for possible EC decay. Assuming that the daughter,  $^{263}\text{Lr}$ [104], primarily decays by EC and not rapidly by SF, we might detect SF decay from chemically separated lawrencium containing the granddaughter,  $^{263}\text{Lr}$ . Our half-life estimate of 4.5 h for SF decay by  $^{263}\text{Lr}$ , given in Fig. 12, can be compared with 40 min measured for  $^{261}\text{Lr}$  and  $\geq 8$  h for  $^{262}\text{Lr}$ . From a single 12-h bombardment with  $^{13}\text{C}$  ions, we obtained cross-section limits for the formation of  $^{263}\text{Lr}$ [105] that were dependent on the assumed SF half-life of  $^{263}\text{Lr}$  (Fig. 13). The curve in this figure was derived by assuming we counted three SF events (95% confidence). This experiment suggests that EC decay by  $^{263}\text{Lr}$ [105] is doubtful. Having virtually eliminated  $\alpha$  and EC as possible decay modes for  $^{263}\text{Lr}$ [105], we conclude the most probable mode is SF with a half-life of well under 1 min.

#### b. $^{264}\text{Lr}$ [105]

We attempted to identify  $^{264}\text{Lr}$ [105] through SF decay by it or its possible EC daughter,  $^{264}\text{Lr}$ [104].<sup>14</sup> We would expect, with a high degree of certainty, that the even-even daughter isotope would decay within a few seconds by SF. Our experiment consisted of a series of short (30 min) bombardments of

$^{254}\text{Es}$  with 71.8-MeV  $^{13}\text{C}$  ions, followed by fast (10 to 15 min) chemical separations of element-105 and then counting the separated samples for SF activity. The chemical separations included a cation-exchange column where the VB series of elements (Nb, Ta, element-105) were eluted from the cation resin with 0.5M HF, which was then followed by extraction of the VB elements from a mixture of 6M HCl/6M HF into 4-methyl-2-pentanone. Based on added tracers, niobium and tantalum were separated with a 50 to 60% yield while interfering actinides were reduced by a factor of about  $10^8$ . We did not find any fission activities in the samples which could be attributed to the SF decay of  $^{264}\text{[105]}$  or its daughter,  $^{264}\text{[104]}$ . Assuming the chemical yield for element 105 is the same as for Nb and Ta and  $^{264}\text{[105]}$  has a 17-min EC half-life, we arrived at an upper limit of  $\sim 6$  nb for the formation of  $^{264}\text{[105]}$ . This limit is too high to be conclusive in rejecting EC as major decay mode for  $^{264}\text{[105]}$ . Moreover, we cannot exclude a SF half-life shorter than  $\sim 5$  min for this isotope because our chemical separations required at least 10 min, allowing time for a short-lived  $^{264}\text{[105]}$  to fully decay.

c.  $^{272}\text{[109]}$

Microscopic calculations of potential energy surfaces by Möller *et al.*,<sup>15</sup> Böning *et al.*,<sup>16</sup> and Cwiok *et al.*<sup>17</sup> show an improved stability for nuclides with N near 162 and atomic numbers between 106 and 110. Their calculations show the fission barriers increasing from about 5 MeV in the heavy actinides to 8.5 MeV in the center of this region because of a neutron subshell that is stabilized by a hexadecapole shape of the nucleus. It is especially important to determine whether these long theoretical extrapolations of nuclear properties are correct; if they are not, substantial changes in our understanding of the microscopic features of the nucleus would become necessary. The nuclide  $^{272}\text{[109]}$ , with 163 neutrons, lies in the zone of predicted maximum stability. Furthermore, it has the virtue of being an odd-odd nucleus, a property that normally promotes about a millionfold increase in SF half-lives. For the experimentalist attempting to produce and identify nuclei in this new region, these are very fortunate qualities. Lastly, this nuclide is accessible only through heavy-ion reactions with  $^{254}\text{Es}$ , which may be an adverse factor, considering the limited quantities of  $^{254}\text{Es}$  available.

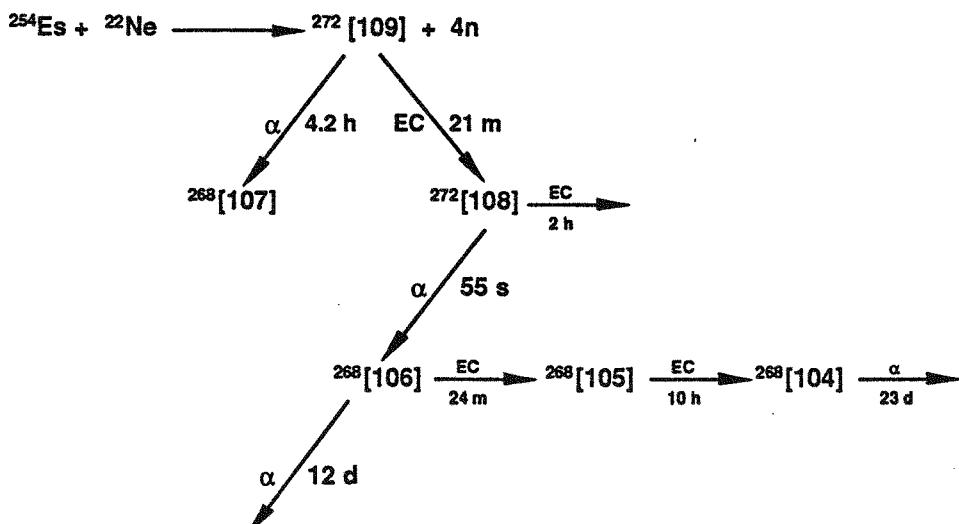


FIG. 14. Production and decay sequence anticipated for  $^{272}\text{[109]}$ .

Figure 14 indicates the production method and possible decay sequences for  $^{272}\text{[109]}$ . Because of the long extrapolations from known nuclides, these estimates are, perhaps, little better than best guesses. Although more recent estimates of SF half-lives are available now, at the time we accepted this decay

scheme as the basis of our experiment to test nuclear stability in this very heavy region. We bombarded  $^{254}\text{Es}$  with 117.5-MeV  $^{22}\text{Ne}$  ions for about 120 h, concentrating our search on detecting  $\alpha$  particles with distinctive energies from the decay of  $^{272}\text{[108]}$ , following EC decay by  $^{272}\text{[109]}$ .<sup>18</sup> To rapidly isolate element 108 from grossly larger quantities of actinides produced in transfer reaction, we used a He-jet system in combination with gas-phase separation chemistry. This continuous on-line method provided separation factors approaching  $10^8$  from nearly all other reaction products within a time scale of 2 s. The gas-phase separation was based on decomposition of the KCl aerosol by passing it through quartz wool heated to 650 °C while simultaneously introducing a small quantity of oxygen into the helium stream to produce a volatile tetroxide of element 108. From tests with a  $^{165}\text{Ho}$  target to produce  $^{182}\text{Ir}$  and its EC daughter  $^{182}\text{Os}$ , we obtained  $\geq 35\%$  yield for osmium, the 5d-shell chemical homolog of element 108. We are certain the yield is larger because some volatile  $\text{OsO}_4$  was lost during transfer of the samples to gamma counters. The  $\alpha$  spectrum obtained from 91.6 h of bombardment is presented in Fig. 15. Our analysis shows 3 or fewer  $\alpha$  decays in any 100-keV bin between 8.9 and 9.2 MeV, the energy range we expected for the  $\alpha$  decay of  $^{272}\text{[108]}$ . The presence of Rn isotopes in the spectrum (several at energies  $< 8$  MeV) showed they were condensed on the  $-155$  °C collecting surface, indicating the experiment was performing properly. From the few possible events in the  $\alpha$ -energy range expected for  $^{272}\text{[108]}$ , we calculate a formation cross section of less than 1 nb. We conclude either the stability predicted for these nuclides is less than anticipated or their formation cross section is lower than our limit.<sup>18</sup> Also the SF half-life of  $^{272}\text{[108]}$  may be considerably less than 2 s, in which case we would not have observed an  $\alpha$ -decay branch in these experiments. But this, too, would indicate a failure of the theoretical predictions.

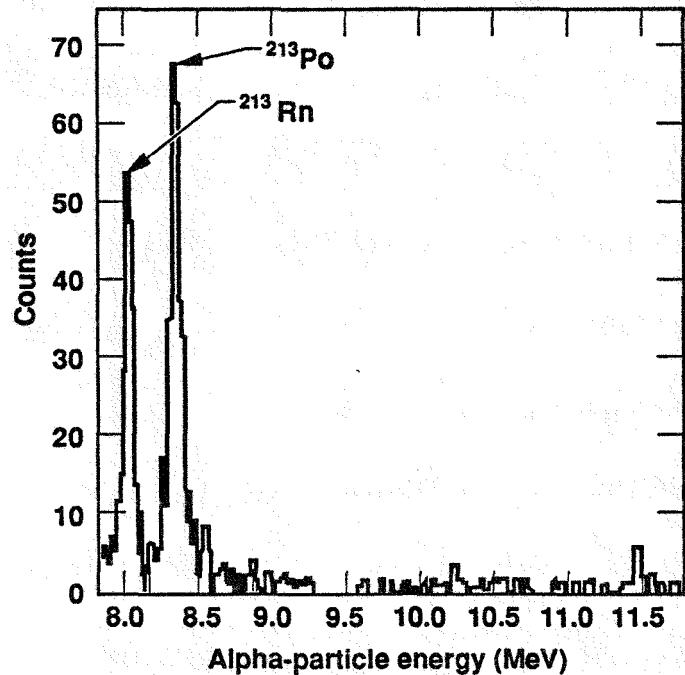


FIG. 15. Alpha-particle spectrum from the volatile species produced in 91.6 h of bombarding  $^{254}\text{Es}$  with  $^{22}\text{Ne}$  ions. The peak at 8.375 MeV is due to residual  $^{213}\text{Po}$  from calibration of the detectors with a  $^{229}\text{Th}$  source.

#### 4. SUMMARY

A surprising amount of information has resulted from the bombardment of  $^{254}\text{Es}$  with heavy ions. Moreover, with the discovery of 7 isotopes, we have pushed back the frontier in the transpermium region toward the neutron-rich side, a particularly difficult quarter to otherwise reach. A summary of these results should highlight the most significant consequences, chief among these being the discovery of

bimodal fission. This finding has resulted in new insights into the fission process, greatly altering our perception of the physics involved, and some have thought it to be the most important discovery in fission within the last 20 years. For the near future, we believe the primary goal is to determine if the very large increase in stability predicted for nuclei around the 162-neutron subshell exists. An experimental demonstration of this forecast is pivotal in verifying the accuracy of the long theoretical extrapolations of nuclear properties. If they are incorrect, the supposed existence of the superheavy elements is in great jeopardy because of the even longer theoretical extrapolations that predict their stability.

Our many experiments using  $^{254}\text{Es}$  as a target have provided much needed information on a group of known and previously unknown nuclides that were in a critical region with respect to tests of theory and to forming better predictions for the properties of even more distant nuclei. Along the way, there were several surprising results, including the discovery of bimodal fission, the finding of a reversal in the systematic trend of SF half-lives for nobelium isotopes, and, with each new isotope discovered, we have found SF half-lives to be many orders of magnitude longer than anticipated ten years ago. These revelations would not have appeared if appreciable quantities of  $^{254}\text{Es}$  had not been systematically produced by the U.S Transplutonium Production Program. We have more than touched the surface of the possible research that can be performed by bombarding  $^{254}\text{Es}$  with heavy ions, but further progress depends upon the availability of larger amounts of  $^{254}\text{Es}$  for targets and new instruments with much greater discrimination against unwanted background radiation from uninteresting nuclides coproduced in heavy-ion reactions with  $^{254}\text{Es}$ . Plans for producing up to ten times the current amounts of  $^{254}\text{Es}$  have been made; in concert with them, we and others have proposed several new instruments to overcome the discrimination problem and to attack entirely new problems concerning relativistic effects in these very heavy atoms.<sup>19</sup> Much of the expectations and promises projected by these earlier proposals have been confirmed in the results reported here. However, the possibilities for significant progress are far from being exhausted. The time has now arrived to build new instruments and to extend the studies using  $^{254}\text{Es}$  targets into an even heavier and more difficult realm of nuclides. Perhaps, then, we could hope to disclose further crucial results five years from now.

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