Spontaneous fission of rutherfordium isotopes

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We have found several new spontaneous fission (SF) activities and suggested assignments for some of them to rutherfordium (element 104) isotopes. Their half-lives and production cross sections have been measured by collecting recoils on a moving nickel or steel tape and transporting them past stationary mica track detectors at known velocities. The following tentative assignments are based on several cross bombardments and comparisons between experimental and calculated production cross sections: ^{256}Rf(9±2 ms), ^{257}Rf(3.8±0.8 s, 14±9% SF), ^{258}Rf(13±3 ms), ^{259}Rf(3.4±1.7 s, 9±3% SF), ^{260}Rf(21±1 ms), and ^{262}Rf(47±5 ms). Presently we are unable to assign several other new spontaneous fission activities with half-lives of ~15 ms, ~22 ms, ~100 ms ($Z < 104$), ~1.6 s, ~5 s, ~5 s, ~30 s, and ~47 s. Contrary to other observations, we have not found any indication for an 80-ms spontaneous fission activity, earlier credited to the spontaneous fission decay of ^{260}Rf, in reactions in which we expected to produce this isotope. Our measurements support a shift in the spontaneous fission half-life systematics at element 104, first proposed by Flerov and Oganessian *et al.* and later predicted by Randrup *et al.* and Baran *et al.* and attributed to the disappearance of the second hump of the double-humped fission barrier and a weakening of the 152-neutron subshell.

I. INTRODUCTION

Based on the charged liquid-drop model of the nucleus, nuclei tend to be stable with respect to spontaneous fission (SF) decay if the ratio of Coulomb energy to twice the surface energy is less than one, or equivalently, $Z^2/A < 50$. Since the Coulomb energy increases roughly as $Z^2/A^{1/3}$ while the surface energy increases more slowly as $A^{2/3}$, fission becomes an increasingly important decay mode for the heaviest elements. This importance and the different categories of SF decay are apparent from a color wall chart (available from one of the authors, L.P.S.) showing the 121 SF activities for which assignments have been established or suggested. Nuclei near closed shells are expected to be more stable against SF than neighboring nuclei; nuclei with odd numbers of protons and/or neutrons are generally more stable against SF than neighboring even-even nuclei. An understanding of the shell effects and the systematic variation in partial half-life for SF decay as a function of the number of neutrons and protons is essential in predicting the half-lives for heavier, unknown nuclei and, in particular, for superheavy elements.

This paper is primarily concerned with the SF half-lives of rutherfordium isotopes (element 104). At the beginning of our experiments there were already some half-life measurements by a group from the Joint Institute of Nuclear Research (JINR) indicating that a shift in the SF half-life systematics might be occurring at element 104.[1,2] This conclusion was largely based upon half-life and cross section measurements from several cross bombardments aimed at producing rutherfordium isotopes. More recent measurements using the velocity filter SHIP (Ref. 3) (separator for heavy ion products) by Münzenberg *et al.* have confirmed a half-life of ~8 ms for ^{256}Rf.[4,5]

Several authors have since made calculations that indicate a drastic modification of the SF half-life systematics at element 104 (Refs. 6–8) or at element 102 and beyond.[9] Randrup *et al.* attributed this change in half-life systematics to the disappearance, below the ground state, of the second hump of the double-humped fission barrier in rutherfordium isotopes and a weakening of the 152-neutron subshell effect on the SF half-lives. Mustafa and Ferguson have also predicted an absence of the second barrier for even-even rutherfordium isotopes with mass numbers of 260 or higher.[10]

In our attempts to determine the partial SF half-lives of rutherfordium isotopes and, therefore, to see if there is indeed a change in the SF half-life systematics at element 104, a recoil tape-transport system with high sensitivity was employed. The reactions used, together with the measured production cross sections and half-lives, are listed in Table I. In addition, we have found other new SF activities and measured their production cross sections. Most of these activities could not be assigned and are briefly discussed at the end of Sec. III. A more detailed discussion of these unassigned activities is found in Ref. 11. Preliminary reports of our results can also be found in Refs. 12–14, 16, and 90. The details of the experimental apparatus and the methods used to produce these new SF activities are described in the following section.

II. EXPERIMENTAL

The search for new SF activities with half-lives between milliseconds and seconds and cross sections in the

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TABLE I. Observed spontaneous fission activities with possible assignments to isotopes of element 104.

Mass	$T_{1/2}$ (ms)	Estimated fission branch (%)	Cross section (nb) Expt.[a]	Calc.[b]	Reaction	Projectile energy (MeV)
256	9±2	97[c]	7±3	0.8	^{249}Cf(^{12}C,5n)^{256}Rf	85
257	3800±800	14±9	2±0.4	30	^{249}Cf(^{12}C,4n)^{257}Rf	75
258	13±3	100?	10^{+10}_{-3}	3	^{246}Cm(^{16}O,4n)^{258}Rf	95
259	3400±1700	9±3	0.6±0.2	7	^{245}Cm(^{18}O,4n)^{259}Rf	93
260	20±1.2	100?	14±2	10	^{249}Bk(^{15}N,4n)^{260}Rf	80
	21±1.1		6±1	4	^{248}Cm(^{16}O,4n)^{260}Rf	92
	19±1.4		9±1[d]		^{249}Cf(^{18}O,α3n)^{260}Rf	96
262	53±4	100?	5±1	5	^{248}Cm(^{18}O,4n)^{262}Rf	89
	50±16		1±1	1.5	^{244}Pu(^{22}Ne,4n)^{262}Rf	113

[a]Experimental partial cross section for *fission* decay, assuming compound nucleus ranges in the target.
[b]Calculated *total* production cross section using the JORPLE code for a rutherfordium isotope including all decay modes (Refs. 29 and 30).
[c]A 3-percent α branch in ^{256}Rf with 8.81-MeV α particles was reported in Ref. 88.
[d]The unknown range of the ^{260}Rf recoils produced in this reaction is likely to be less than the compound nucleus range assumed in determining this cross section. If so, this cross section should be multiplied by $639/\{[^{260}\text{Rf range } (\mu g/cm^2) \text{ in } CfF_3] - 107\}$.

nanobarn range is hampered by the production through transfer reactions of the SF emitter ^{256}Fm (2.6 h) and of ^{256}Mo (77 min), which decays by electron capture to ^{256}Fm. This yielded a substantial SF background in previous experiments where we measure short half-lives in the millisecond range using a wide, rotating drum.[13–15]

A major improvement was the development of a fast tape system. Thus, for most of the experiments reported in this work a long tape collected and transported the recoils past one meter of stationary mica track detectors,[94] as shown in Fig. 1. With one to two km of tape the linear density of ^{256}Fm and/or ^{256}Md nuclei was so low that the SF background was reduced a factor of 20 compared to the scanning, rotating drum system described in Refs. 13 and 14. The tape was either 1 km of 25-μm-thick nickel or 2 km of 12.5-μm-thick stainless steel and cooled by contact with a water-cooled post or by a helium gas jet directed toward the back side of the tape. The target, tape, and everything in electrical contact with the tape acted together as a Faraday cup for measuring the beam intensity. The beam from the 88-inch cyclotron was collimated to a diameter about one millimeter smaller than the target diameter as shown in Fig. 1. The target diameters were 6.4 mm except for the ^{254}Es target which was concentrated onto a 3-mm diam area because of the very small amount of target material available.

Typically, the tape moved at speeds of 0.05 to 1.3 meter per second, as determined from the angular velocity of the capstan drive. The tape could be moved left and right at different speeds, regulated to within one percent. During the slowdown, reversal, and acceleration of the tape motion at the end of each reel cycle, the cyclotron beam was turned off electronically. With one meter of micas on

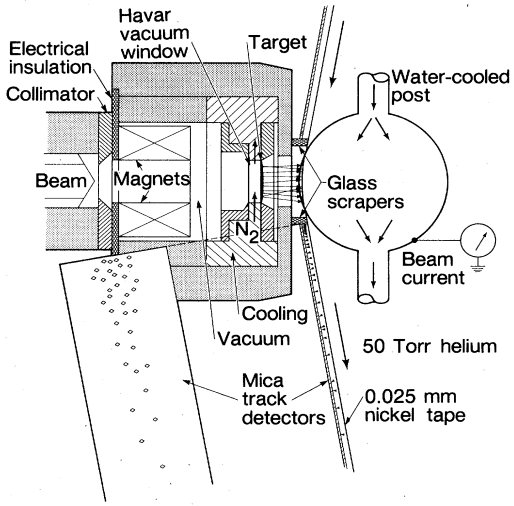

FIG. 1. Recoil tape-transport system with large background suppression for the investigation of short-lived SF activities. Nuclei produced at the target are shown recoiling forward. These recoil nuclei stop in a moving nickel tape, 1 km in length, which transports them past stationary mica track detectors. The SF decays from nuclei deposited in the tape, shown as tiny arrowheads, register as the diamond-shaped tracks in the mica shown on the inset of the figure. The number of tracks or SF decays per unit length of mica decreases with the time after production. Knowing the tape speed, the time associated with each track is measured by the distance from the target along the mica detectors. Not shown is a typical 30-$\mu g/cm^2$ aluminum cover foil over the target to reduce the transfer of target material to the tape. The drawing is approximately to scale, except for the mica inset showing the tracks, with the scale set by a target diameter of 6.4 mm.

each side of the target and different left and right tape speeds, effects independent of tape speed, such as beam scattering and neutron-induced fission, could be checked. Scattered ions from the beam could damage the mica surface near the edges of the first micas and, if the density of scattered tracks was very high, they could be confused with SF tracks. Glass "scrapers" were therefore placed in contact with the tape surface on both sides of the target to shield the mica from scattered ions. Unfortunately, the tape often became warped with use so that good contact between the scrapers and tape could not always be maintained; therefore, the data from the first several millimeters of mica were usually rejected, making it difficult to detect activities with half-lives of 5 ms or less.

After each bombardment, the mica plates were etched for 50 min in 48-percent hydrofluoric acid at 60 °C. The SF tracks were then visible under a microscope with $\times 100$ magnification. From the known speed of the tape, the linear distances from the target position to the tracks in the mica corresponded to the times of SF decay for nuclei after their production at the target. From these decay times, decay curves were constructed and the half-lives and cross sections were determined with a multicomponent, maximum-likelihood code.[17] The overall detection efficiency, which included the geometrical efficiency for emitting SF fragments from the tape, the formation of acceptable tracks in the mica detectors, and the scanning efficiency, was determined to be ~ 70 percent. This efficiency was obtained by comparing the SF rate from an uncovered ^{252}Cf source measured in a gaseous ionization chamber with the rate measured with mica track detectors from a source covered by a thin layer of nickel, which simulated SF fragments from recoils deposited in the tape. Subjective differences in scanning efficiency, determined by two observers scanning the same micas and by the same observer scanning the same micas twice, one year apart, were found to be as large as ten percent.

The India-ruby muscovite mica used in the experiments offered a convenient method for detecting SF fragments because of its relative insensitivity to light ions and its low uranium content.[94] To eliminate the historical background due to SF of natural uranium in the mica, we annealed the micas for several days at 500 °C and preetched them with hydrofluoric acid for ~ 80 min at 60 °C. After each bombardment, the micas were etched again for 50 min. Preetching caused the historical (old) tracks to be larger in size because of a longer etching time than the tracks from SF of nuclei produced in the bombardment. The necessary preannealing of the micas also helped to break down the walls of the older tracks. With these procedures, we could readily distinguish the SF tracks from nuclei produced in the bombardment from the larger, "faded" tracks from SF of natural uranium nuclei in the mica.

We also estimated upper limit cross sections for neutron-induced fission of uranium in the mica. The spatial flux distribution of fast neutrons away from a 113-MeV ^{22}Ne beam striking a ^{244}Pu target was measured to have an exponential decay constant of ~ 0.7 cm^{-1} for the experimental setup of Fig. 1. This measurement was done by using micas to detect the number of neutron-induced fissions from a thin layer of uranium nitrate. Assuming that all of our experiments had this same spatial distribution of neutrons, the background we measured in certain controlled experiments with bombarding energies near the peaks of (HI,4n) excitation functions gave us an effective cross section of less than 0.1 nb due to neutron-induced fission of uranium in the mica.

The targets used in the experiments were prepared either by vaporizing the actinide fluoride onto a heated beryllium backing foil[18] or by electroplating the actinide onto the target backing in a solution of isopropyl alcohol and nitric acid.[19] In the case of the ^{249}Bk target, the berkelium was chemically separated from its californium daughter four days prior to the first bombardment and then volatilized as the fluoride BkF_3; the einsteinium chemical separation took place thirteen days prior to the first bombardment. Decay of the target material over the course of several experiments was taken into account in computing cross sections. In most cases the targets were coated and covered with a total of 25 to 60 μg/cm^2 of aluminum to reduce the transfer of target material to the tape. Typical target thicknesses were 0.5 to 0.9 mg/cm^2 of the actinide fluoride or oxide, of which 0.4 to 0.7 mg/cm^2 was the actinide element. These thicknesses were equal to or slightly larger than the calculated ranges of compound-nucleus recoils produced in ^{15}N- to ^{18}O-induced reactions.

The projectile energy at the target was determined from the resonant frequency of the cyclotron and from the calculated energy loss[20] through the arrangement of a Havar vacuum-separation foil, nitrogen cooling gas, and beryllium target backing shown in Fig. 1. After passage through the target, measurements of the mean beam energy using solid-state detectors were found to agree with the energy calculated from the range-energy tables of Northcliffe and Schilling to better than 0.5 MeV for ~ 111-MeV ^{18}O ions. Small effects such as energy straggling [≤ 0.9 MeV, estimated[21,22]] and the finite energy width of the cyclotron beam [≤ 0.1 MeV (Ref. 23)] have been neglected in plotting excitation functions; taking them into account would slightly reduce the widths of the excitation functions in Figs. 2(a) and 5.

We used the ranges of compound nucleus recoils in the target compound plus aluminum cover foils to determine all of the cross sections in the paper. These ranges were calculated from the energies of compound nucleus recoils using range tables[20] and an approximation for ranges in compounds derived from the Bragg additivity rule for stopping powers in the elements of the compounds.[24] Ranges calculated from these tables agreed to within 10 percent with the experimental range measurement of Hahn *et al.*[25] for the reaction

$$83.8\text{-MeV } ^{12}\mathrm{C}+^{238}\mathrm{U}\rightarrow ^{245,244}\mathrm{Cf}+5\mathrm{n},6\mathrm{n}\ ,$$

using a U_3O_8 target. Since the corrections to the recoil yield from the targets due to range straggling were calculated to be less than 10 percent,[11,26] excluding the unknown effect of localized variations in thickness, they were not included in determining the cross sections.

If our suggested assignments to compound nucleus

TABLE II. Cross bombardments consistent with the assignments ^{260}Rf(~20 ms) and ^{262}Rf(~50 ms).

Compound-nucleus reaction	Projectile energy (MeV)	Cross section (nb) ^{260}Rf(~20 ms)	^{262}Rf(~50 ms)
$^{16}O + ^{244}Pu \rightarrow ^{260}No$	95	≤ 4	≤ 0.6
$^{13}C + ^{248}Cm \rightarrow ^{261}No$	81	≤ 6	≤ 6
$^{18}O + ^{244}Pu \rightarrow ^{262}No$	95	≤ 0.3	≤ 0.3
$^{15}N + ^{248}Cm \rightarrow ^{263}Lr$	97	≤ 1	≤ 1
$^{15}N + ^{248}Cm \rightarrow ^{263}Lr$	97	≤ 2 (15 ms)	
$^{15}N + ^{248}Cm \rightarrow ^{263}Lr$	83	≤ 4	≤ 3
$^{12}C + ^{249}Cf \rightarrow ^{261}Rf$	85	≤ 2	≤ 1.4
$^{16}O + ^{246}Cm \rightarrow ^{262}Rf$	95	≤ 7	≤ 2
$^{13}C + ^{250}Cf \rightarrow ^{263}Rf$	73	≤ 370	≤ 540
$^{16}O + ^{248}Cm \rightarrow ^{264}Rf$	92	6 ± 1	$\leq 0.6 \pm 0.3$
$^{15}N + ^{249}Bk \rightarrow ^{264}Rf$	80	14 ± 2	$\leq 0.3 \pm 0.7$
$^{18}O + ^{248}Cm \rightarrow ^{266}Rf$	89	≤ 26	5 ± 1
$^{22}Ne + ^{244}Pu \rightarrow ^{266}Rf$	113	≤ 2	1 ± 1
$^{15}N + ^{249}Cf \rightarrow ^{264}Ha$	86	$\leq 0.3 \pm 0.1$	≤ 0.2
$^{18}O + ^{249}Bk \rightarrow ^{267}Ha$	93	≤ 0.7	≤ 0.6
$^{13}C + ^{254}Es \rightarrow ^{267}Ha$	73	≤ 150	≤ 220
$^{18}O + ^{249}Cf \rightarrow ^{267}[106]$	96	9 ± 1	$\leq 0.0 \pm 1.5$
$^{18}O + ^{250}Cf \rightarrow ^{268}[106]$	99	$\leq 1500 \pm 300$	$\leq 13\,000$
$^{18}O + ^{254}Es \rightarrow ^{272}[107]$	99	$\leq 0.5 \pm 0.1$	≤ 0.5
$^{22}Ne + ^{250}Cf \rightarrow ^{272}[108]$	125	≤ 80	≤ 140
$^{22}Ne + ^{254}Es \rightarrow ^{276}[109]$	125	≤ 50	≤ 90

products in Table I are correct, the cross sections should also be correct, except for the transfer reaction ^{249}Cf(^{18}O,α3n)^{260}Rf, producing recoils with an unknown range. It is important to note that ranges for products of transfer reactions such as this can be significantly different from compound nucleus ranges.[25,27] Future experiments might show that some of the other SF activities listed in Tables I and II and discussed in the figures are actually transfer products with longer or shorter ranges than the compound nucleus ranges used in establishing the cross sections. If so, the cross sections can be corrected roughly by multiplying them by the ratio of the compound nucleus range to the actual range in the target material with ranges derived from integral range distributions.

Several points concerning the data analysis are worth mentioning. The multicomponent maximum-likelihood code maximizes a likelihood function for a set of event times $t_1, t_2, \ldots, t_n$ associated with each of the n tracks to determine the most probable half-life and number of counts for each component in the data.[17] To calculate upper limits for cross sections, a least-squares computer code FRANTIC (Ref. 28) was used to fit the data. The component of interest was assumed to be present in the data along with components similar to the maximum-likelihood ones. The half-lives and amounts of all components were variable except for the half-life of the component of interest. The weights for the points used in fitting the data were determined from maximum-likelihood results.

The errors quoted in Tables I and II for all of the cross-section measurements include the statistical error in the number of counts, a ten-percent uncertainty in the scanning efficiency, the uncertainty in resolving more than one activity, and, in one case, the uncertainty in the beam flux measurement. However, the half-life errors reflect only the statistical uncertainties. Some half-lives and cross sections which were measured repeatedly were found to be in statistical disagreement. As an example, two separate measurements of the half-life for the reaction 109-MeV $^{18}O + ^{248}Cm$—17 ± 2.2 ms and 22 ± 1.3 ms—have only a 5-percent statistical probability of differing by this much or more.

III. EXPERIMENTAL RESULTS

The half-life and the production cross section are the only two properties of a SF activity that can be deduced from a single tape experiment. Even these quantities can be uncertain if more than one component is present. In Table I we suggest assignments to isotopes of rutherfordium based on comparisons between calculated or estimated cross sections for production of a particular isotope and the experimental cross sections for several cross bombardments. We used the JORPLE code, a later version of the Jackson-Sikkeland model,[29,30] to calculate the production cross sections for neutron-evaporation residues. This model predicts the maximum cross sections for compound nucleus products with evaporation of four neutrons to within a factor of 3 of the experimental values for the heavy ions ^{12}C to ^{18}O bombarding actinide targets. The model also agrees with the projectile energies at which the excitation functions for neutron-evaporation products reach their peaks, although discrepancies of up to ± 3 MeV have been observed for some carbon- and ^{18}O-induced reactions[31] and for one case with ^{15}N ions,[32] as shown in Fig. 2(d). The widths of experimental excitation

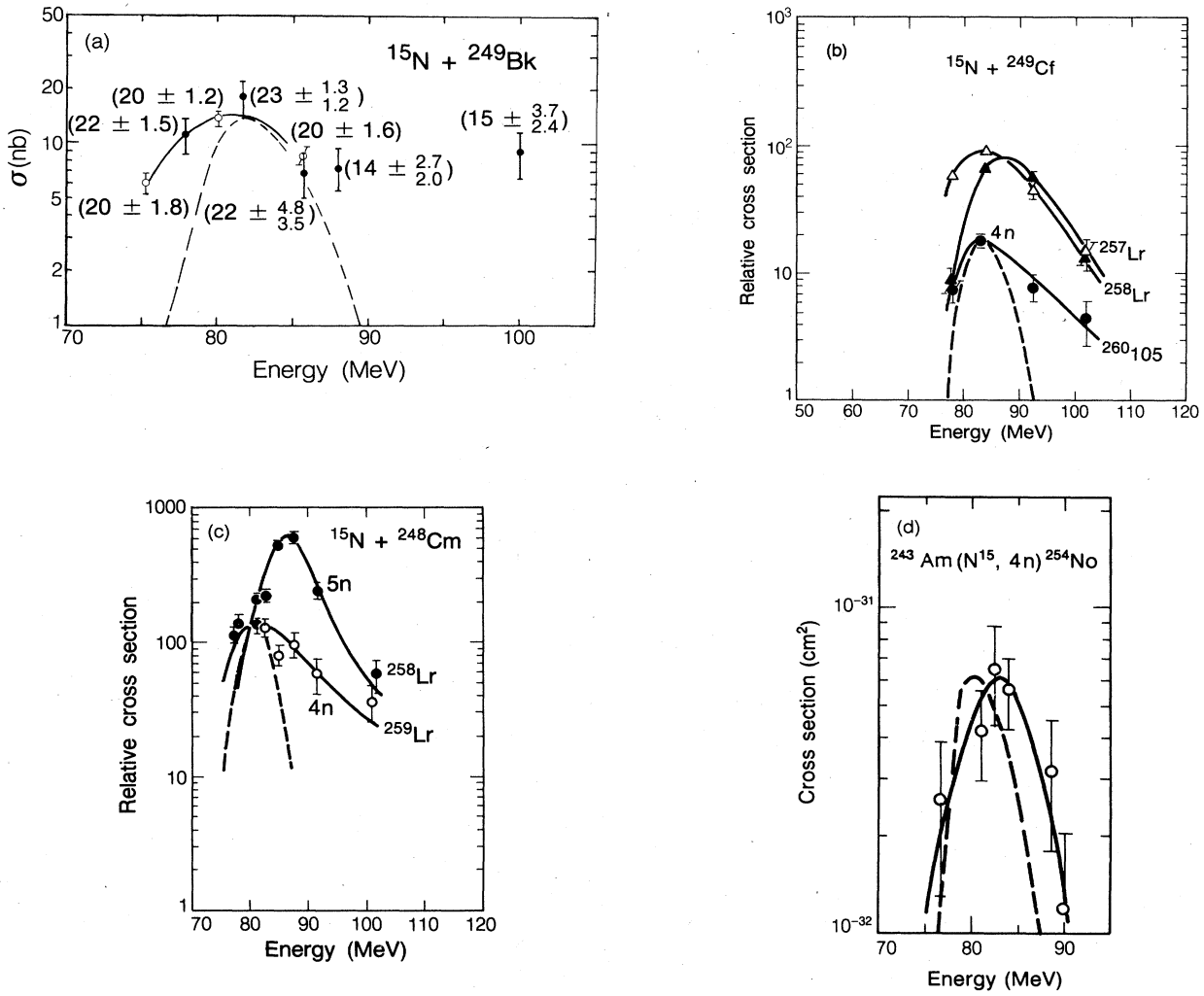

FIG. 2. (a) Excitation functions for the short-lived SF activities produced in the rotating-drum experiments (Ref. 13) (solid circles) and in the tape experiments (open circles) for the reaction $^{15}N + ^{249}Bk$. The values in parentheses give the half-lives with standard deviations or 68-percent confidence limits measured for each energy. The solid curve is drawn to guide the eye; the dashed curve is the excitation function calculated using the JORPLE code for production of ^{260}Rf. The cross sections for the rotating-drum experiments are 7 to 8 percent lower than those quoted in Ref. 13 due to small corrections in the calculation of recoil ranges (see Sec. II). (b)—(d) Experimental excitation functions for (^{15}N,4n) reactions. The dashed curves show the excitation functions for (^{15}N,4n) reactions calculated with the JORPLE code and normalized to the peak cross sections for each of the experimental excitation functions. Note that each experimental excitation function is broader than calculated. (b) $^{249}Cf(^{15}N,4n)^{260}Ha$ (Ref. 92), (c) $^{248}Cm(^{15}N,4n)^{259}Lr$ (Ref. 93), (d) $^{243}Am(^{15}N,4n)^{254}No$ (Ref. 32).

functions are, however, often greater than calculated, as shown in Figs. 2(a)—(d). Measured cross sections for similar reaction types, if available, have been used in estimating cross sections for noncompound nucleus products.

Often several isotopes were consistent with the available cross section data for production of a SF activity. But we knew the half-lives for some of these isotopes and they differed from the half-life of our SF activity. We also compared the half-life we measured with extrapolations of the known SF half-life systematics to other candidate isotopes. Thus, from a knowledge of the SF half-life systematics for $Z \leq 102$ we could narrow down the list of possible assignments for the activity. In the case where we observed SF activities with half-lives between 14 and 24 ms, it is difficult to explain all of the cross bombardments by production of a single isotope; two or more isotopes with similar half-lives may have been produced.

In the following we summarize our present knowledge of the fission properties of the rutherfordium isotopes and discuss several new SF activities that we observed but could not assign.

A. ^{253}Rf

A 1.8-s SF activity assigned to ^{253}Rf was found in the bombardment ^{50}Ti + ^{206}Pb.[33] Since the production cross section was half the calculated cross section, the SF branch was postulated to be 50 percent.

B. ^{254}Rf

In the reaction 245-MeV ^{50}Ti + ^{206}Pb, 15 SF events were found with a half-life of 0.5±0.2 ms and a production cross section of 7 nb.[34] Ter-Akopyan *et al.* assigned this 0.5-ms SF activity to ^{254}Rf based on the following arguments:

(1) The calculated production cross section for ^{254}Rf agreed with the measured cross section for the 0.5-ms SF activity.

(2) The activity was absent in the reaction ^{48}Ti + ^{208}Pb which has a minimum excitation energy 6 MeV higher than for the reaction ^{50}Ti + ^{206}Pb.

(3) The partial α-decay half-life for ^{254}Rf was estimated to be 4—600 ms, consistent with SF being the dominant decay mode.

C. ^{255}Rf

A ~2-s SF activity produced in the reaction ^{207}Pb(^{50}Ti,2n)^{255}Rf was assigned to a 50-percent SF branch of ^{255}Rf by comparing experimental and calculated production cross sections.[33] In Refs. 2 and 35 the half-life was reported as approximately 4 s. Using the velocity filter SHIP at Gesellschaft für Schwerionenforschung (GSI) it was later found in the same reaction that 8.726-MeV α particles from ^{255}Rf were correlated in both time and position with the known ^{251}No daughter α-particle decays.[4,5] The half-life for the α-decay events from ^{255}Rf was $1.4^{+0.6}_{-0.3}$ s, with 68-percent confidence limits. SF events with the same half-life of $1.4^{+1.0}_{-0.4}$ s were also observed, implying a 45-percent SF branch and confirming the original work of Flerov *et al.*

D. ^{256}Rf

A 5-ms SF activity produced in the reaction ^{50}Ti + ^{208}Pb with a cross section of 6 nb was assigned to the isotope ^{256}Rf.[36] A later half-life measurement of $8.1^{+1.3}_{-0.8}$ ms from the same reaction using SHIP supported this assignment[4,5] based on discrimination against transfer products and the additional evidence of a narrow excitation function peaking near the optimum bombarding energy expected for producing ^{256}Rf.

From the reaction of 85-MeV ^{12}C with ^{249}Cf we produced a 9±2-ms SF activity with a cross section of 7±3 nb. This half-life agrees within error with the value of $8^{+1.3}_{-0.8}$ ms measured at GSI for ^{256}Rf, but the production cross section we measured is nine times larger than calculated from the Jackson-Sikkeland model. Having obtained this same high cross section in previous measurements, two of us (M.J.N. and A.G.) believed an assignment to ^{256}Rf was unlikely.[37] The Jackson-Sikkeland model correctly predicts the cross sections for the quite similar (^{12}C,5n) and (^{13}C,5n) reactions with targets of ^{235,236,238}U (Refs. 38—40), ^{239}Pu (Refs. 41 and 42), ^{241,243}Am (Ref. 43), and ^{244,246,248}Cm (Refs. 38, 39, and 44). Thus, the high cross section is still unexplained.

E. ^{257}Rf

^{257}Rf decays primarily by α-particle emission[45,46] with a complex α-particle spectrum and a half-life of 4.8±0.5 s.[47] It was the first α-emitting isotope of element 104 to be found,[15] and the name rutherfordium was proposed for this element,[48] based on the Z identification of its known daughter ^{253}No. The atomic number of this isotope was also well established by observing the coincidences of α decays from ^{257}Rf with nobelium daughter x rays.[46] In the bombardment 75-MeV ^{12}C + ^{249}Cf, we have produced a 3.8±0.8-s SF emitter with a cross section of ~2 nb. Assignment of this SF emitter to other known 3- to 5-second SF activities such as ^{256}No and ^{259}Rf can be ruled out on the basis of small fission branches and/or low estimated production cross sections. Since the measured half-life for this SF emitter and the half-life for α-particle decay of ^{257}Rf agree within the uncertainties of the measurements, we suggest that the 3.8-s SF activity is a SF branch in ^{257}Rf. Although an uncertainty in the partial cross section for the production and subsequent α decay branch was not quoted in either Ref. 46 (~12 nb when produced with 73-MeV ^{12}C ions), or Ref. 45 (~10 nb when produced with 68-MeV ^{12}C ions), these measured cross sections taken together with the excitation function measurements described in Ref. 45 would suggest that the uncertainty in the cross section for production with 75-MeV ^{12}C ions is probably not larger than 50 percent. Comparing the measured partial cross sections for the production of ^{257}Rf radioactivities including α decay (~12 nb when produced with 73-MeV ^{12}C ions) and SF decay (2±0.4 nb with 75-MeV ^{12}C ions), these data would imply a possible fission branch of 14±9 percent.

With 237- to 242-MeV ^{50}Ti ions bombarding a target of ^{208}Pb, Münzenberg *et al.* recorded 18 α-particle decays from ^{257}Rf and three SF events which were position correlated with the stopping of the corresponding recoils 0.1 s or more earlier.[5,49] Considering that their efficiency for detecting SF events was twice that of α particles in those experiments, we calculate that the SF branch of ^{257}Rf is less than or equal to 8±5 percent. Since ^{255}Rf, a probable 1.4-s SF emitter, can also be produced in the reaction ^{208}Pb(^{50}Ti,3n)^{255}Rf, the three SF events they observed with 237- to 242-MeV ^{50}Ti ions cannot be unambiguously assigned to ^{257}Rf or ^{255}Rf. However, based on the four SF events they assigned to ^{255}Rf produced with 252- to 257-MeV ^{50}Ti ions and an extrapolation of the excitation function for ^{255}Rf measured from its unambiguous α-decay branch, we would expect only 0.3 SF events with 242-MeV ^{50}Ti ions, whereas three SF events were observed. Therefore, we consider it more likely that these three SF events are associated with ^{257}Rf than with ^{255}Rf. In this case, the fission branch calculated from the data of Münzenberg *et al.* would be 8±5 percent, which is consistent with our estimate of 14±9 percent.

F. ^{258}Rf

Previously, excitation functions for a SF activity with a half-life of 13±2 ms were measured for products of the

reactions $^{12,13}C + ^{249}Cf$ and their excitation functions were found to be consistent with an assignment to ^{258}Rf.[15,45,50]

In the GSI experiments producing elements 107 (Ref. 51) and 109 (Ref. 52) both α decay and electron capture decay followed by SF events were assigned to ^{258}Ha. From the agreement in the measured respective half-lives of $1.8^{+1.1}_{-0.7}$ s and $1.6^{+0.6}_{-0.5}$ s for these two decay modes of ^{258}Ha, it follows that the SF half-life of ^{258}Rf is less than one second, in agreement with the previous half-life measurements of 13 ± 2 ms.[15,45,50]

We also have produced a 13 ± 3-ms SF activity in the reaction 95-MeV $^{16}O + ^{246}Cm$. This activity is probably also due to ^{258}Rf. Although the production cross section of 10^{+10}_{-5} nb listed in Table I is larger than the calculated value of 3 nb, the difference may not be significant. The large uncertainty in the experimental cross section reflects mainly the uncertainty in the fluence of the ^{16}O beam using the early rotating drum system.[15]

G. ^{259}Rf

^{259}Rf is mainly an α-particle emitter[45] with a half-life of $3.0 + 1.3$ s.[53] Ratios of 0.07—0.12 have been measured for the yield of $\sim$3-s SF events versus the measured or calculated yields of ^{259}Rf, in the reactions $^{13}C + ^{249}Cf$ (9 ± 5 events),[53] $^{18}O + ^{246}Cm$ (31 events),[54] and $^{22}Ne + ^{242}Pu$ ($\sim$60 events).[1] All of the authors assigned these 3-s SF events to a SF branch in ^{259}Rf.

We have observed 20 SF events with a half-life of 3.4 ± 1.7 s in the reaction 93-MeV $^{18}O + ^{245}Cm$. The measured production cross section of 0.6 ± 0.2 nb corresponds to 9 ± 3 percent of the calculated cross section to produce ^{259}Rf, in agreement with the SF branching ratios for ^{259}Rf reported by the previous authors. Since the 3.2-s half-life of ^{256}No, with a 0.3-percent fission branch, is very close to that of ^{259}Rf, we cannot distinguish these two isotopes. Although the production cross section for ^{256}No in our reaction $^{18}O + ^{245}Cm$ is unknown, a very rough estimate based on the cross section for the quite similar reaction $^{248}Cm(^{18}O,\alpha3n)^{259}No$ would account for only 10 percent of the events we observed.

Bemis *et al.*[53] compared the measured number of α particle decays from both ^{259}Rf and ^{256}No, produced in the reaction 86.5-MeV $^{13}C + ^{249}Cf$, with the SF decays expected from the fission branch of ^{256}No and found that 5 ± 1 fission events should be due to ^{256}No. This can be compared to the remaining 9 ± 5 fission events assigned to ^{259}Rf. Although few SF events were observed in each of the studies cited, a consistent SF branching ratio of 0.07 to 0.12 is estimated or obtained from four different reactions.

H. ^{260}Rf

In 1964 a group at the Joint Institute for Nuclear Research (JINR) in Dubna[55] assigned a 300-ms SF activity to the isotope ^{260}[104], claimed the discovery of element 104, and named it kurchatovium. This half-life has since been revised by the JINR group to 100 ms,[56,57] and after later experiments to 80 ms.[58–61] However, searches by several of the present authors for the 100-ms SF activity in the reactions 94-MeV $^{16}O + ^{248}Cm$ and 92-MeV $^{18}O + ^{246}Cm$,[15] and later for the 80-ms SF activity in the reaction 78- to 86-MeV $^{15}N + ^{249}Bk$ (Refs. 13 and 14) have yielded negative results. We have since searched for the 80-ms SF activity with improved sensitivity using the tape system. Table III summarizes the results derived from the data shown in Figs. 3(a)—(c) for the reactions $^{15}N + ^{249}Bk$, $^{16}O + ^{248}Cm$, and $^{18}O + ^{249}Cf$. The limits

TABLE III. Cross sections for SF [upper limits (nb)] for an *assumed* half-life of 80 ms and for a range of half-lives from 60 to 100 ms of ^{260}Rf.

Reaction	Projectile energy (MeV)	*Assumed* half-life 80 ms	60—100 ms
$^{249}Bk(^{15}N,4n)^{260}Rf$	80	$\leq0.3\pm0.4$	$\leq0.3\pm0.5$
$^{248}Cm(^{16}O,4n)^{260}Rf$	92	$\leq0.4\pm0.2$	$\leq0.5\pm0.2$
$^{249}Cf(^{18}O,\alpha3n)^{260}Rf$	96	$\leq0.0\pm0.5$	$\leq0.0\pm1.0$

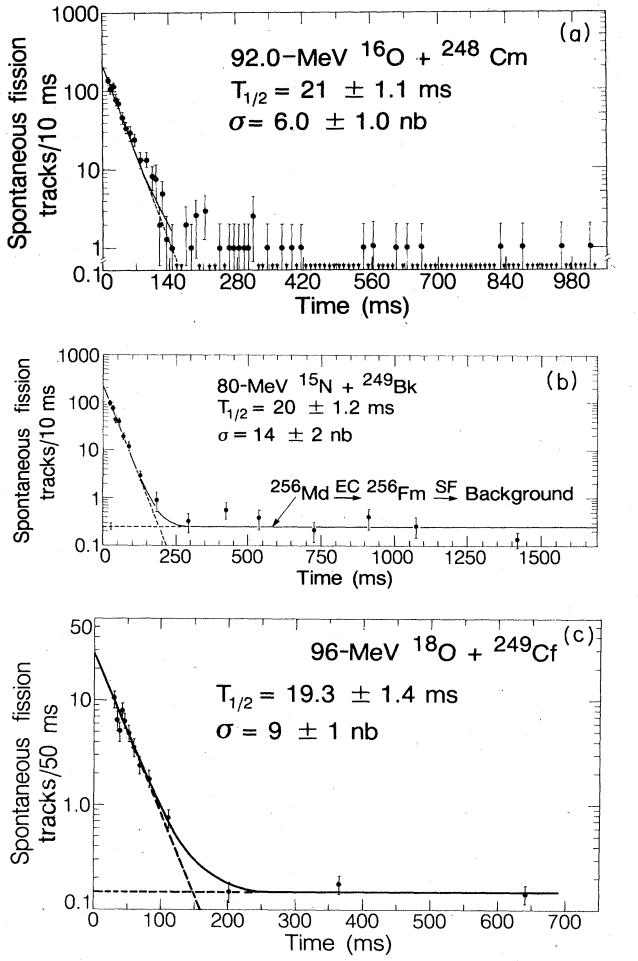

FIG. 3. Decay curves showing $\sim$20-ms SF activities observed in the reactions (a) 92-MeV $^{16}O + ^{248}Cm$, (b) 80-MeV $^{15}N + ^{249}Bk$, and (c) 96-MeV $^{18}O + ^{249}Cf$. The dashed lines show the amounts of ^{256}Fm backgrounds and short-lived components determined in the fits; the solid curves show the sums of these components. The cross-section upper limits for a possible 80-ms SF activity, listed in Table III were established from these same data.

for the reactions 80-MeV $^{15}N + ^{249}Bk$ and 92-MeV $^{16}O + ^{248}Cm$ are, respectively, 30 and 10 times below the calculated production cross sections listed in Table I for producing ^{260}Rf. For the reaction 80-MeV $^{15}N + ^{249}Bk$ our cross section upper limit is 24 times lower than the value of 8 ± 2 nb reported by the JINR group for the same reaction using 82-MeV ^{15}N ions,[59] even though the cross sections measured by each group for the production of ^{256}Md agree.[13,59]

In 1969 a SF activity with a half-life between 20 and 30 ms was first produced in the reaction of ^{16}O with ^{248}Cm by Ghiorso *et al.*[15] They considered the assignment of ^{260}Rf a possible, but unlikely one, because a half-life of the order of 10^{-6} s was expected, based on an extrapolation of the existing SF half-life systematics for $Z \leq 102$. A $\sim$20-ms SF activity was later produced in the reaction $^{15}N + ^{249}Bk$ and an assignment to ^{260}Rf was suggested, based on several cross bombardments, in Refs. 13 and 14. The JINR group argued that this $\sim$20-ms SF activity produced in the reaction $^{15}N + ^{249}Bk$ was actually composed of a mixture of 13.7-ms $^{242}Am^f$ and the 80-ms SF activity which they assigned to $^{260}[104]$.[59,62] This interpretation was shown to be highly improbable compared to the interpretation of a single $\sim$20-ms SF activity.[11,13,63,64]

The assignment of the $\sim$20-ms SF activity to ^{260}Rf is supported by the following data:

(1) The calculated production cross sections for ^{260}Rf agree with the measured values from the data in Figs. 3(a) and (b) for the reactions $^{16}O + ^{248}Cm$ and $^{15}N + ^{249}Bk$, as shown in Table I; the estimated cross sections for other candidate isotopes do not agree with the set of values measured for the SF activity for all the reactions listed in Tables I and II.

(2) For the reaction $^{15}N + ^{249}Bk$ the bombarding energies for the experimental and calculated maxima of the excitation functions agree, as shown in Fig. 2(a).

(3) The cross sections for other (HI,α3n) and (HI,α4n) reactions[15,65–67] are consistent with a value of $\sim$9 nb obtained from the data in Fig. 3(c) for the reaction $^{249}Cf[^{18}O(96\ MeV),\alpha3n]^{260}Rf$.

(4) The $\sim$20-ms SF activity was absent in the other reactions listed in Table II in which low cross sections were expected for production of ^{260}Rf.

(5) Symmetric SF events observed for the $\sim$20-ms SF activity are so far only associated with neutron-rich nuclei with $Z \geq 100$.[64]

(6) The ranges of the $\sim$20-ms SF activity recoils in the reaction $^{15}N + ^{249}Bk$ (Ref. 64) are consistent with the ranges calculated for the compound nucleus product ^{260}Rf.

A $\sim$20-ms SF activity was also produced in the reaction 109-MeV $^{18}O + ^{248}Cm$, as shown in Fig. 4, with a cross section of $\sim$10 nb. Its half-life is indistinguishable from the $\sim$20-ms SF activity which was produced in the reaction $^{15}N + ^{249}Bk$ and assigned to ^{260}Rf ($^{18}O + ^{248}Cm$—22 ± 1.3 ms; $^{15}N + ^{249}Bk$—20 ± 1.2 ms). But the production cross section of 50 times the calculated value for ^{260}Rf, the broad excitation function width of 12 MeV shown in Fig. 5 (8 MeV calculated for ^{260}Rf), and the excitation function peak at 109 MeV (103 MeV calculated for ^{260}Rf) are all inconsistent with the compound nu-

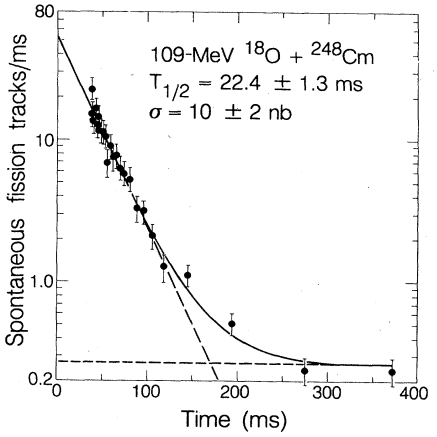

FIG. 4. Decay curve for the reaction 109-MeV $^{18}O + ^{248}Cm$ showing a 22-ms SF activity. This half-life is indistinguishable from the half-lives deduced from the data in Figs. 3(a)—(c). The dashed lines show the amounts of the ^{256}Fm background and the short-lived component determined in the fit; the solid curve shows the sum of these components.

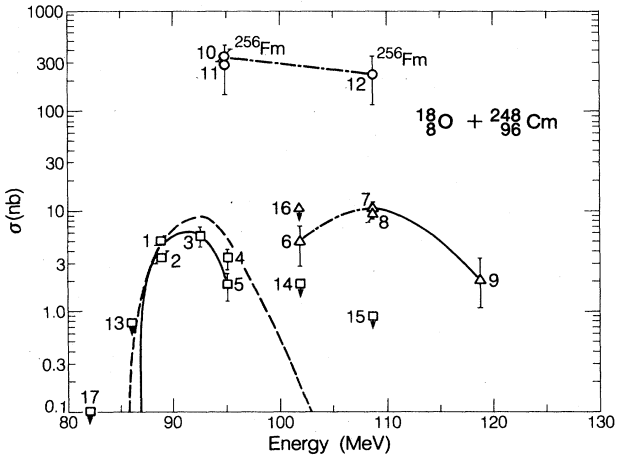

FIG. 5. Excitation functions for the SF activities observed in the reaction $^{18}O + ^{248}Cm$. The following are measured half-lives listed according to the identifying number beside each point in the figure: (1) 55 ± 5 ms, (2) 50 ± 6 ms, (3) 34 ± 6 ms, (4) 51 ± 9 ms, (5) 29 ± 22 ms, (6) 35 ± 28 ms, (7) 22.4 ± 1.3 ms, (8) 17.1 ± 2.2 ms, (9) 15 ± 4 ms, and (10) $\sim$2.67 h. Point numbers 13 to 15 and 17 are cross-section upper limits for a $\sim$50-ms SF activity and point number 16 is a cross-section upper limit for a $\sim$20-ms SF activity. The $\sim$50-ms SF activity is probably due to ^{262}Rf; the $\sim$20-ms SF activity is unknown but probably is a transfer product with $Z < 104$. The long-dashed curve represents the excitation function calculated using the JORPLE code for production of ^{262}Rf. The solid curves and the dot-dashed curves are meant to guide the eye through the experimental points. A dot-dashed curve was chosen to connect point number 6 because that point has a large uncertainty in both the cross section and half-life. The cross sections for ^{256}Fm have been determined by measuring the SF-decay rates from aluminum catcher foils.

cleus product ^{260}Rf; while the reactions $^{15}N + ^{249}Bk$, $^{16}O + ^{248}Cm$, and $^{18}O + ^{249}Cf$ produced a ~20-ms SF activity consistent with an assignment to ^{260}Rf. Our ^{248}Cm targets were composed of 96.5% ^{248}Cm, 3.3% ^{246}Cm, and 0.0004—0.0013 % ^{244}Cm. Although ^{260}Rf can be produced in the reaction $^{18}O + ^{246}Cm$, the ^{246}Cm fraction of our target is small and the calculated production cross section using ^{18}O ions with such a high energy of 109 MeV is completely negligible. Consequently, a single ~20-ms SF activity could not have $Z=104$ and be consistent with all of our cross bombardments. The ~20-ms SF activity produced in the reaction $^{18}O + ^{248}Cm$ is probably a different isotope from ^{260}Rf, that is, a transfer product with $Z<104$. Using the reaction $^{18}O + ^{248}Cm$ to show that its recoil angular distribution is not typical for compound nucleus reactions and/or that its SF mass distribution or total kinetic energy for SF differ from the fission properties measured for ^{260}Rf (Ref. 64) would prove that two distinct ~20-ms SF activities exist.

I. ^{261}Rf

This isotope, with a half-life of 65 s, decays by emitting α particles with energies of 8.22—8.32 MeV. These α particles have been correlated with the decays of the recoil daughter ^{257}No in a separate detector. This isotope has also been used to show that rutherfordium behaves chemically like its homolog hafnium, as expected from the periodic table of elements.[15,68,69] An upper limit of 10 percent for SF branching in ^{261}Rf(65 s) has been established.[70]

J. ^{262}Rf

As seen from the decay curve of Fig. 6, SF activities with half-lives of 55 ms and 1.3 s were produced in the reaction 88.8-MeV $^{18}O + ^{248}Cm$. The origin of the 1.3-s component is unknown and this activity is briefly discussed in Sec. III K 3. The narrow excitation function for the ~50-ms SF activity shown in Fig. 5 and the maximum production cross section of 6 nb are consistent with identifying it as the compound-nucleus evaporation product ^{262}Rf. Points 3 and 5 of Fig. 5 have poorly determined half-lives due to critical regions of mica which had to be rejected in the half-life analysis because they were either scratched or of unacceptable quality. The weighted average of our half-life measurements for the reaction $^{18}O + ^{248}Cm$ gives 47±5 ms. We have probably also produced this activity in the reaction 113-MeV $^{22}Ne + ^{242}Pu$, although only 77 SF events were observed. The corresponding production cross section of ~1 nb for this last reaction is close to the calculated cross section of ~1.5 nb for ^{262}Rf. The results from other cross bombardments in Table II are consistent with the assignment of the ~50-ms SF activity to ^{262}Rf. It is unlikely that the ~50-ms SF activity could be a transfer product because, including the upper limit points 13—15 and 17 of Fig. 5, the excitation function for its production in the reaction $^{18}O + ^{248}Cm$ appears quite narrow.

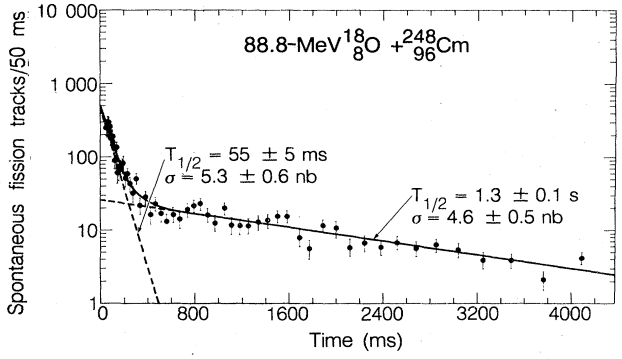

FIG. 6. Decay curve for the SF events observed in the reaction 88.8-MeV $^{18}O + ^{248}Cm$. The dashed lines show the amounts of the 55-ms and 1.3-s components computed in the fit; the solid curve is the sum of these components. The 55-ms component is probably due to ^{262}Rf. The 1.3-s component has not yet been identified but is briefly discussed in Sec. III K 3.

K. Unassigned SF activities or activities with $Z \neq 104$

1. ~15-ms and ~22-ms SF activities

Based solely on their half-lives these two activities could be ^{258}Rf(13 ms) and ^{260}Rf(~20 ms). However, the ~15-ms SF activity produced in the reaction 88- to 100-MeV $^{15}N + ^{249}Bk$ and the 15- to 22-ms SF activity produced in the reaction 109- to 119-MeV $^{18}O + ^{248}Cm$ both have production cross sections which exceed those calculated for the (HI,6n) reaction products ^{258}Rf(13±2 ms) and ^{260}Rf(~20 ms) by factors of ~130 and ~50, respectively. Furthermore, the excitation functions for these activities shown in Figs. 2(a) and 5 are considerably broader than calculated for ^{258}Rf and ^{260}Rf and are more characteristic of transfer products. Since no mechanism is known that would enhance the (^{15}N,6n) or (^{18}O,6n) reaction cross sections we cannot assign these ~15- and ~22-ms SF activities to ^{258}Rf and ^{260}Rf(~20 ms), respectively. This is based on the fact that no evidence of any special enhancement was found in any of the reactions $^{238}U(^{18}O,6n)^{250}Fm$,[71] the quite similar reaction

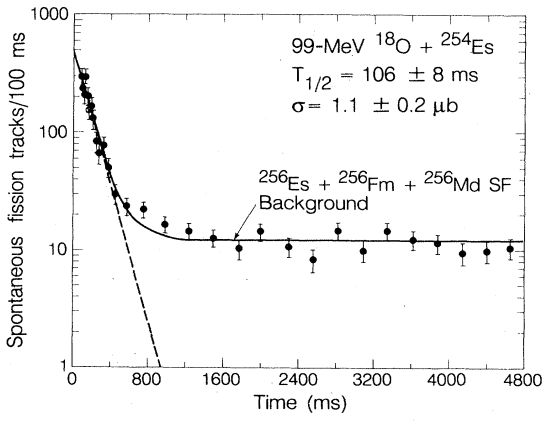

FIG. 7. Decay curve for the reaction 99-MeV $^{18}O + ^{254}Es$ indicating that a 106-ms SF activity is produced. The dashed line shows the amount of the 106-ms component computed in the fit; the solid curve is the sum of this component and SF background. Although this ~100-ms activity could not be assigned, the production cross section is very large compared to our other SF activities.

^{248}Cm(^{18}O,5n)^{261}Rf (Ref. 70), ^{238}U(^{16}O,6n)^{248}Fm (Ref. 72), or ^{238}U(^{14}N,6n)^{246}Es (Ref. 73).

2. ~100-ms SF activity

We have found a 106±8-ms SF activity shown in Fig. 7 with a production cross section of 1.1±0.2 μb in the reaction 99-MeV ^{18}O + ^{254}Es.[90] An activity with a similar half-life of 73±24 ms was produced in the reactions of 73-MeV ^{13}C ions with a target composed of 68 to 75 percent ^{254}Es and 25 to 32 percent ^{250}Cf. Assuming this activity was produced from the ^{254}Es portion of the target its production cross section was ~200 nb. In the reaction 125-MeV ^{22}Ne + ^{254}Es only 29 SF events were observed, but these were consistent with a ~100-ms half-life and a cross-section upper limit of 80 nb. ^{260}Rf is a very unlikely assignment for this ~100-ms SF activity because of its absence in other reactions listed in Table III in which we should have easily produced that nuclide. But ^{260}No is a possible assignment because it is the only even-even nuclide whose production cross section of 1.1 μb would fit an extrapolation of the yield curve for transfer products from the reaction ^{18}O + ^{254}Es.[74] Because of a displacement of the nobelium yield curve toward lower neutron numbers in transfer reactions of ^{22}Ne with ^{254}Es, the cross section for ^{260}No in this reaction would be expected to be much lower, also in agreement with experiment. However, a ~100-ms half-life for ^{260}No would be surprisingly long, based on an extrapolation of the known nobelium half-lives in Fig. 8 and a known half-life of only 1 ms for ^{258}No. Thus, an assignment to ^{260}No is supported by our cross bombardments but would be surprising in view of the nobelium half-life systematics.

3. ~1.6-s SF activity

The ~1.3-s SF activity produced in the reaction ^{18}O + ^{248}Cm with a maximum cross section of 16 nb and shown in the decay curve of Fig. 6 has properties similar to ^{259}Fm. The most precise half-life of 1.6±0.1 s obtained from our tape experiments, the symmetric mass distribution, and the mean total kinetic energy for fission[75] all agree with the same properties measured for ^{259}Fm, which was discovered in the reaction ^{257}Fm(t,p)^{259}Fm.[76] The excitation function in Fig. 10 of Ref. 11 is broad, as expected for a transfer product. But other more favorable transfer reactions analogous to ^{248}Cm(^{18}O,X)^{259}Fm have measured peak cross sections or upper limits that are either lower or no larger than the value of 16 nb measured for the ~1.6-s SF activity.[11,77–79] These data would suggest that either there is another ~1.6-s SF activity with fission properties similar to ^{259}Fm, or that the original assignment of ^{259}Fm is incorrect.

Recently the JINR laboratory reported the production of SF activities with half-lives of 1–3 s and cross sections of the order of 10 nb in the reactions of ^{20}Ne and ^{22}Ne ions with targets of ^{249}Cf and ^{249}Bk.[80] Possible assignments of ^{252,256}No, ^{246}Fm, ^{257}Es, and ^{260}Md were suggested.

4. ~5-s SF activities

A half-life of 4.5±0.5 s and a production cross section of ~7 nb were measured for a SF emitter produced in the reaction 70-MeV ^{13}C + (80%^{249}Bk/20%^{249}Cf). A one-percent electron-capture branch in ^{258}Lr(4.4 s) to the spontaneously fissioning isotope ^{258}No(1.2 ms) is a possible explanation. Another 5.5±1.5-s SF activity which could not be due to either ^{258}Lr or ^{257}Rf was produced with a cross section of 3 nb in the reaction 98-MeV ^{18}O + (82–88% ^{249}Bk/12–18% ^{249}Cf).

5. SF activities with half-lives between 30 and 50 s

A 47±13-s SF activity, produced with a cross section of ~9 nb in the reaction 75-MeV ^{12}C + ^{249}Cf, could not be a SF branch in ^{254}No, based on our limit of 0.05 percent for this fission branch obtained in the reaction ^{245}Cm(^{13}C,4n)^{254}No. This limit is in agreement with the limit we derived from Refs. 32 and 81, giving the lower limit for the partial half-life for SF of ^{254}No shown in Fig. 8. A 30±10-s component in the decay curve for the reaction

98-MeV ^{18}O+(82–88 % ^{249}Bk/12–18 % ^{249}Cf)

might be due to a SF branch in ^{262}Ha(34.1 s).[82–84] However, assuming the 30-s component is a 78-percent SF branch of ^{262}Ha,[85] the total production cross section of 6 nb is much larger than the calculated cross section of 1 nb.

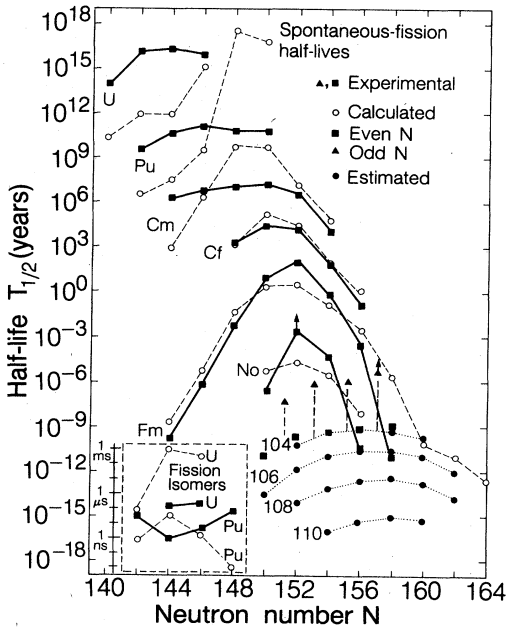

FIG. 8. Comparison between the experimental half-lives and the partial half-lives calculated or estimated by Randrup *et al.* (Ref. 7) for SF of even-even nuclei. [For rutherfordium the experimental partial half-lives for the odd-A isotopes are included. ^{253}Rf with a partial SF half-life of 3.6 s (Ref. 33) is not shown.] Lower limits on the partial SF half-lives of ^{256}No (this work and Refs. 32 and 81) and ^{261}Rf (Ref. 70) are shown as upward-pointing arrows. The inset at the lower left shows a comparison between calculated and experimental half-lives for fission isomers.

IV. DISCUSSION OF SF HALF-LIFE SYSTEMATICS

In Fig. 8 the tentative half-lives for even-even element-104 isotopes have been plotted together with the predictions of Randrup *et al.*[7] In the calculations the partial SF half-lives for even-even nuclei up to element 102 were first fitted by adjusting the renormalization constant for irrotational flow in the macroscopic inertial mass. Then the half-life calculations were extrapolated to elements 104 and beyond. In the case of uranium and plutonium nuclei with poorly-known potential barriers at large deformations,[7] the agreement for both ground states and isomeric states (inset of Fig. 8) is worse than for heavier nuclei with less extended barriers. The half-lives for activities with proposed assignments to rutherfordium isotopes are remarkably close to the predictions. At element 104 Randrup *et al.* predicted a weakening of the 152-neutron subshell effect and a disappearance of the second hump of the double-humped fission barrier.[7] Baran *et al.*[8] also have estimated the partial half-lives for SF of even-even nuclei without any adjustable parameters by using the cranking approximation to calculate the inertial mass parameters for the fission modes. However, the agreement with the tentative rutherfordium half-lives is not as good as the calculations shown in Fig. 8.

The shape of the distribution of half-lives versus neutron number is very sensitive to whether the second barrier is above or below the ground state for each isotope of element 104. For thorium to curium nuclei the height of the second hump of the fission barrier has been observed to monotonically decrease with increasing proton number.[86] This trend would suggest that for some proton number the second barrier drops below the ground state, in agreement with the theoretical predictions. A possible example of this effect can be seen in Figs. 8 and 9. For fermium isotopes the drastic decrease in partial SF half-life from $^{256}_{100}$Fm ($T^{SF}_{1/2}=2.86$ h) to $^{258}_{100}$Fm ($T^{SF}_{1/2}=0.38$ ms) may be the result of a lowering of the second barrier below the ground state. With a sudden decrease of the effective width of the fission barrier, the fission half-life is decreased by a factor of $\sim 10^7$ (Refs. 6 and 7). As shown in Fig. 9, this effect was predicted to occur between ^{258}Fm and ^{260}Fm,[6] whereas the experimentally observed drop in half-lives occurs between ^{256}Fm and ^{258}Fm in Fig. 8. The 152-neutron subshell might lower the ground state below the second barrier, which would drastically increase the half-life. According to the calculations of Randrup *et al.* the second barrier lies one or two MeV below the ground state for all even-even element-104 isotopes with neutron numbers between 150 and 158.[87] Therefore, even if the 152-neutron subshell effect in the ground state is as large in rutherfordium isotopes as it is in nobelium isotopes [~ 0.75 MeV (Ref. 11)], the calculated half-lives would still be determined mainly by the penetration of only one barrier and show no dramatic increase at neutron-number 152.

Mustafa and Ferguson[10] have also predicted the second barrier to be below the ground state for even-even rutherfordium nuclei with $A \geq 260$. These calculations were made using the asymmetric two-center shell model, which minimizes the potential energy of deformation with respect to the neck radius, the ratio of nuclear volumes on either side of the neck plane, and other physical dimensions. For $A < 260$ these authors predict that the second barrier is *above* the ground state. If correct, there would be an increase in the SF half-lives as the mass number decreases below 260. However, the probable assignments of an 8-ms half-life to ^{256}Rf and a 13-ms half-life to ^{258}Rf would suggest that the 152-neutron subshell effect upon these SF half-lives is weak and that the second barrier is below the ground state for these even-even rutherfordium isotopes.

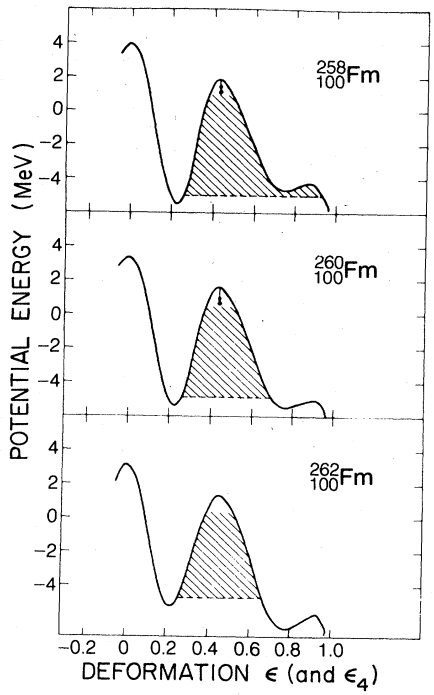

FIG. 9. Calculated fission barrier potentials plotted against the deformation parameter ϵ for heavy isotopes of fermium (Ref. 6). Beyond ^{258}Fm the second peak and second minimum are below the ground state energies shown as the horizontal dashed lines at the bases of the cross-hatched areas. This leads to a drastic decrease in the SF half-lives.

In Fig. 8 the tentative partial half-lives for the SF of odd-mass rutherfordium isotopes are plotted as solid triangles. The SF hindrance factors derived from these half-lives for odd-mass isotopes of elements 104 and 106 relative to the even-even isotopes are summarized in Table IV. For the references listed under each isotope in Table IV a preference has been made to experiments in which SF and α decays were measured simultaneously, i.e., entries with references labeled "(SF + α)." For other references the precise SF branches and hindrance factors are less certain because either the SF or α decays were measured in separate experiments, as for example ^{263}[106] (Refs. 67 and 84), or only the partial cross section for SF decays has been measured and compared to the calculated total production cross section, as for example ^{253}Rf.[33] But these hindrance factors are generally smaller than those observed for odd-mass nuclei of lighter elements such as nobelium, fermium, and californium. For odd rutherfor-

TABLE IV. Tentative odd-mass SF hindrance factors for elements 104 and 106.

Isotope	Half-life from α decay (s)	SF branch (%)	SF hindrance factor	References SF branch	References Half-life from α decay
^{253}Rf[a]	1.8	50	>7000	33 (SF)[c]	33[g]
^{255}Rf	$1.4^{+0.6}_{-0.3}$	45±20	~ 700	(4,5) (SF + α)[b]	4,5
		50	~ 700	33 (SF)[c]	
^{257}Rf	4.8±0.5	8±5[d]	~ 6000[d]	(5,49) (SF + α)[b]	47
		14±9	~ 3000	This work (SF), (45,46) (α)[e]	
^{259}Rf	3.0±1.3	6.3±3.7	~ 3000	53 (SF + α)[b]	53
		9±3[f]	~ 2000	This work (SF)[c]	
		7	~ 3000	54 (SF)[c]	
		6—12	1500—3000	1 (SF)[c]	
		≤ 20	~ 900	91 (SF + α)[b]	
^{261}Rf	65±10	≤ 10	≥ 20000	70 (SF + α)[b]	70
^{263}[106]	0.9±0.2	~ 70	1000[h]	84 (SF), 67 (α)[e]	67

[a]Not shown in Fig. 8.
[b]SF branch derived from a simultaneous measurement of SF events and α particles.
[c]SF branch derived from a comparison of the measured partial cross section for SF and the calculated total production cross section.
[d]This is an interpretation we made in Sec. III E of the data from Refs. 5 and 49.
[e]SF branch derived from a comparison of the measured partial cross sections for SF and α decay in separate experiments.
[f]Based upon a calculated cross section, using the JORPLE code, of 6.7 nb for the reaction ^{245}Cm[^{18}O($\sim$93 MeV),4n]^{259}Rf.
[g]Half-life determined from SF of ^{253}Rf. α decay has not been observed for this isotope.
[h]Determined by comparing the estimated partial SF half-life for ^{263}[106] with the predicted half-lives of even-even isotopes of element 106 in Fig. 8.

dium isotopes this can be qualitatively understood in terms of the disappearance of the second barrier. In the calculations of Randrup *et al.*[6] the SF barrier of a nucleus with an odd neutron number is obtained by raising the potential energy over that for an even-even nucleus by a "specialization energy" due to the required conservation of total spin and parity with deformation. The addition of the odd neutron increases both the thickness and height $V(r)$ of the barrier [i.e., the action integral over the fission coordinate r, $\int \sqrt{2MV(r)}/\hbar\, dr$, where M is inertial mass function]. But as Figs. 11 and 12 of Ref. 6 show, the calculated increases in barrier thickness and height are much greater when starting with a double-humped fission barrier than with a single-humped barrier. This explains why rutherfordium isotopes with predicted single-humped barriers may be hindered less than lighter elements with double-humped barriers.

As seen from Fig. 8 the upper limit of 10 percent on the SF branching of ^{261}Rf may imply some added stability associated with 157 neutrons relative to the other rutherfordium isotopes. This added stability associated with 157 neutrons has also been observed in longer than expected *α-decay* half-lives for isotopes of elements 100 to 106.

While the predictions of Randrup *et al.* agree quite well with the SF half-lives for even-even isotopes of element 104, the GSI group recently measured a partial SF half-life of 7 ms for the isotope ^{260}[106], which was considerably longer than the predicted value of $\sim$0.2 ms in Fig. 8.[88,89]

V. CONCLUSIONS

Our SF half-life measurements for isotopes with probable rutherfordium assignments support a change in the SF half-life systematics at element 104 (Refs. 1 and 2) that has been attributed to the disappearance of the second barrier and a weakening of the 152-neutron subshell effect.[7,8] We have presented evidence for the tentative assignment of a $\sim$50-ms SF activity to the new isotope ^{262}Rf. We have again not observed the 80-ms SF activity attributed by the JINR group[55—62] to ^{260}Rf in three favorable reactions for production of that isotope. However, our cross sections for the production of a $\sim$20-ms SF activity in these same three reactions ^{15}N + ^{249}Bk, ^{16}O + ^{248}Cm, and ^{18}O + ^{249}Cf support our previous assignment to ^{260}Rf(20 ms).[13,14] The excitation functions and absolute production cross sections for the $\sim$22-ms SF activity in the reaction 109-MeV ^{18}O + ^{248}Cm and the $\sim$15-ms SF activity[13] in the reactions 88- to 100-MeV ^{15}N + ^{249}Bk are consistent with unknown transfer products with $Z < 104$, but with half-lives indistinguishable from those of the isotopes ^{258}Rf($\sim$13 ms) and ^{260}Rf($\sim$20 ms). Several other new SF activities have been produced in the course of this work, most of which have not yet been assigned. But our results support previous assign-

ments of ^{256}Rf($\sim$8 ms),[36,4,5] ^{258}Rf($\sim$13 ms),[15,45,50] and ^{259}Rf($\sim$3 s, 7% SF).[1,53,54] From the reaction $^{12}C + ^{249}Cf$ our SF data along the others' data[45,46] for production of the α emitter ^{257}Rf support the possible assignment of a 3.8-s SF activity to a 14±9-percent SF branch of ^{257}Rf (4.8 s). Our measurements of the partial half-lives of the odd-A isotopes ^{257,259}Rf, together with the JINR (Refs. 1, 2, and 35) and GSI (Refs. 4 and 5) results on ^{255}Rf, imply odd-neutron SF hindrance factors of 700 to 6000. The fact that these hindrance factors are lower than for lighter elements can be understood in terms of the disappearance of the second barrier.[6] Although the measured half-lives for the SF of even-even isotopes of rutherfordium agree quite well with the predictions of Randrup *et al.*, the calculations will have to be modified to reproduce the longer than expected half-lives of element 106 and beyond.[88,89]

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