Decay properties of ^{257}No, ^{261}Rf, and ^{262}Rf

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(Received 5 July 2000; published 13 November 2000)

In bombardments of ^{244}Pu targets with 114- and 120-MeV ^{22}Ne projectiles we detected 69 α-α correlations linking α decays of ^{261}Rf and ^{257}No. We observed one α peak with $E_\alpha = 8.30 \pm 0.06$ MeV for ^{261}Rf and peaks with α-particle energies 8.07–8.40 MeV for ^{257}No. The half-life of ^{257}No was measured to be 25 ± 3 s. No correlations were found between α decays and subsequent spontaneous fission events, from which we calculated an upper limit of 1.5% for the fission branch of ^{257}No and estimated an upper limit of 3% for the α-decay branch of ^{262}Rf. The cross section of the ^{244}Pu(^{22}Ne,5n)^{261}Rf reaction was measured to be about 4 nb at both ^{22}Ne energies used. We also report on some results from ^{242}Pu+^{22}Ne and ^{238}U+^{26}Mg bombardments.

PACS number(s): 25.70.Gh, 23.60.+e, 25.85.Ca, 27.90.+b

I. INTRODUCTION

The nuclides ^{261}Rf and ^{262}Rf were identified and generally characterized in the discovery experiments (Refs. [1] and [2], respectively). However, more detailed studies of their decay properties are definitely important. First, these nuclides are descendants of neutron-rich α-decaying species with Z,N around the predicted shell closures $Z=108$ and $N=162$ (see, e.g., Refs. [3–6]), whose existence was established in a series of experiments on identification and decay studies of the new nuclides ^{265}Sg, ^{266}Sg, ^{267}Hs, and ^{273}110 [2,7–9]. Since the identification of such new species is based on establishing genetic links between their α decay and subsequent α or spontaneous-fission (SF) decays of the descendant nuclides, a complete knowledge of the decay properties of the latter facilitates decay-sequence searches and prevents overlooking decay chains from heavier precursors. Second, the $N=157-158$ nuclides ^{261}Rf and ^{262}Rf are, by themselves, close to the $N=162$ shell, which can significantly influence their decay properties, especially those of ^{262}Rf [2].

The nuclide ^{261}Rf was characterized [1] to be an α emitter with a half-life $T_{1/2}$ of 65 ± 10 s and the main α-particle energy group at $E_\alpha = 8.28 \pm 0.02$ MeV; its identification was based on physical separation followed by detecting the known 25-s α-decaying daughter, ^{257}No [10,11]. An upper limit of 10% for SF branching in the decay of ^{261}Rf has been reported [1] and an electron-capture (EC) branching of $\leqslant 10\%$ has been estimated [12] from beta half-life systematics. The isotope ^{261}Rf has been repeatedly used in experiments performed to study the chemical properties of Rf (see, e.g., Refs. [13–15]). More recently ^{261}Rf was produced in on-line gas chromatographic studies of Rf chlorides, in which a new, presumably more accurate $T_{1/2}$ value of 78^{+11}_{-6} s has been measured [15]. In all of the previous experiments, the ^{261}Rf was produced via the ^{248}Cm(^{18}O,5n) reaction with an estimated cross section of about 5 nb [1]. A common feature of these experiments was the use of gas (He)-jet techniques to purge the recoil products from the reaction chamber and transport them to α-counting stations, either with or without chemical processing. Because of Pb and Bi impurities in the targets, α-particle spectra in the E_α range of ^{261}Rf and ^{257}No decays were often [1,15] contaminated by α activities, e.g., from ^{211m}Po, ^{212m}Po, or ^{213}Po.

Another feature which hindered previous experiments is the fact that the α energies of ^{257}No (8.22 to 8.32 MeV [10,11]) overlap those of ^{261}Rf; this made the two nuclides indistinguishable by α-energy analyses with the experimental techniques used. Although the technique used in Ref. [1] allowed the physical isolation of the daughter α activity on separate detectors, there was no possibility of following the one-to-one correspondence between two consecutive α decays of a particular mother-daughter pair. In Ref. [15], the very fact of detecting $\alpha-\alpha$ correlations of ^{261}Rf to ^{257}No was shown, but true $\alpha-\alpha$ correlations could not be disentangled from random $\alpha-\alpha$ chains produced by genetically unrelated decays from ^{261}Rf and ^{257}No.

We report here on our experiments (presented preliminarily in Ref. [9]) in which we used the technique of kinematic separation to isolate the $Z=104$ recoils from transfer-reaction products, to implant the separated recoils in a position-sensitive silicon detector, and to observe, for individual α-decay events of ^{261}Rf, a subsequent time- and position-correlated α-decay event of its genetically related daughter ^{257}No. Another goal of our experiments was to discover the α-decay branch of the even-even nuclide ^{262}Rf [2] by searching for time and position correlations between α decays of ^{262}Rf and subsequent SF events from its short-lived spontaneously fissioning daughter ^{258}No.

The first unambiguous identification of the spontaneously

*Deceased.

TABLE I. Summary of the bombardments of ^{244}Pu, ^{242}Pu, and ^{238}U with ^{22}Ne and ^{26}Mg beams. Here W is the average target thickness, E is the beam energy in the middle of the target, E^* the excitation energy of the compound nucleus, D the total beam dose, and σ the production cross section (with an estimated accuracy of a factor of ~ 2) for the indicated nuclide.

Target	W (mg cm^{-2})	Ion	E (MeV)	E^* (MeV)	D ($\times 10^{18}$)	Nuclide	σ (nb)
^{244}Pu	0.41	^{22}Ne	114	46	3.1	^{261}Rf	4.4
			120	51	2.1	^{261}Rf	3.8
^{242}Pu	0.30	^{22}Ne	114	45	3.8	^{259}Rf	1.7
						^{260}Rf [a]	0.9
^{238}U	0.28	^{26}Mg	134	46	0.45	^{260}Rf [a]	0.24
			140	51	1.7	^{259}Rf	1.1
^{238}U	0.25	^{22}Ne	117	51	1.3	^{255}No	200 [b]

[a]Tentative assignment is based on literature data [12].

[b]A cross section value from Ref. [20] corrected by applying the nuclear data from Ref. [12] regarding the EC branchings in the decays of ^{255}No and ^{251}Fm.

fissioning isotope ^{262}Rf was reported in Ref. [2]; it was observed as the α-decay daughter of ^{266}Sg. From time intervals for six detected α-SF correlation chains linking α decays of ^{266}Sg with subsequent SF decays of ^{262}Rf, the total half-life of the ground state of ^{262}Rf was measured to be $1.2^{+1.0}_{-0.5}$ s, 25 times longer than a tentative value of 47 ms ascribed to ^{262}Rf previously [16]. The stability of ^{262}Rf against SF decay proved to be higher by a factor of 10^2-10^3 than that of several nuclides with lower Z or N values, such as ^{258}Fm, ^{262}No, or ^{256}Rf (see also Ref. [17]). This significant stability increase for ^{262}Rf is due to a strong effect of the shell closures $N=162$ and $Z=108$. The observation of α decay of ^{262}Rf would yield important information for improving predictions of masses, shell corrections, and radioactive decay properties of unknown heavy nuclei including those in the vicinity of $N=162$ and $Z=108$. The detection of α decay of ^{262}Rf would also allow the unequivocal identification of ^{258}No, which is reported to be a spontaneously fissioning nuclide with $T_{1/2}=1.2\pm 0.2$ ms [12,18].

II. EXPERIMENTAL TECHNIQUE

To produce ^{262}Rf and ^{261}Rf we used the complete fusion reaction ^{244}Pu+^{22}Ne followed by the evaporation of four or five neutrons from the compound nucleus ^{266}Rf. Beams of ^{22}Ne projectiles were delivered by the Dubna U400 cyclotron. We chose ^{22}Ne bombarding energies of 114 and 120 MeV, resulting in excitation energies of the compound nucleus ^{266}Rf of about 46 and 51 MeV, respectively. Three plutonium targets (98.6% ^{244}Pu, 1.1% ^{242}Pu, and 0.3% ^{240}Pu) with average areal densities of 0.41 mg cm^{-2} ^{244}Pu and a total area of 11.7 cm^2 were arranged on a wheel whose rotation was synchronized to the 150-Hz frequency of the cyclotron so that a target was exposed to the $\sim$2.2-ms beam macropulse during each 6.7-ms beam cycle. The targets were electrodeposited on 0.70 mg cm^{-2} Ti substrates and covered with a 30-μg cm^{-2} carbon layer. For calibration purposes, we performed a bombardment of ^{238}U with ^{22}Ne projectiles. A summary is given in Table I, which also shows results of ^{242}Pu+^{22}Ne [19] and ^{238}U+^{26}Mg [9,19] bombardments (see below).

Evaporation residues (EVR's) recoiling out of the ^{244}Pu targets were separated in flight from beam particles and various transfer-reaction products by the Dubna Gas-filled Recoil Separator, described in Ref. [21]. To set the field B of the separator's dipole magnet for $Z=104$ EVR's, we used prior measurements [2,7,19,21] of the average charge states for slow EVR's with $Z=89-104$ moving in 0.7 Torr of hydrogen, cf. Fig. 1 in Ref. [7]. The average charge states of Rf and No isotopes were calculated to be 1.9, 2.6, and 2.2 for EVR's produced in the ^{242,244}Pu+^{22}Ne, ^{238}U+^{26}Mg, and ^{238}U+^{22}Ne reactions, respectively. The separated EVR's passed through a time-of-flight (TOF) measurement system composed of two (start and stop) multiwire proportional chambers in a 1.5-Torr pentane-filled module and were implanted in a position-sensitive detector (PSD) array composed of three 40×40-mm^2 silicon *Canberra Semiconductor* detectors, each with four 40-mm high ×9.7-mm wide strips. We obtained horizontal (x) positions for the reaction products from the 12 strips and vertical (y) positions from the 40-mm high resistive layer of the detectors. Top and bottom or y-position signals from each strip were divided into a signal for α or implant events ($\sim 1-14.5$ MeV) and a signal for SF events ($\sim 20-250$ MeV). We also recorded the energy of the α/implant events; we determined the total energy of SF events by off-line summing of their y-position signals. With each detected energy event, we also recorded the strip number, TOF information, the time in μs from the beginning of each beam pulse to either α or implant or SF events, and the running time in 0.1-ms intervals. The dead time of the electronics system was ≈ 7 μs. The data were acquired in list mode.

Alpha-energy calibrations were performed periodically using α emitters produced in the ^{197}Au+^{22}Ne reaction. Most of the strips had α-energy peak full width at half maximum (FWHM's) of about 50–60 keV. By using known event sequences from the calibration reactions, we measured the FWHM y-position deviation Δpos to be 1.2 mm (3% of the strip height) for α-α sequences; for α-SF sequences we expected a similar FWHM Δpos value. The FWHM Δpos value for Rf EVR-α and EVR-SF correlations was estimated to be $\approx$6 mm due to the low measured energies of the EVR's.

We determined the detection efficiency of ^{216}Ac EVR's produced in the complete fusion reaction ^{197}Au+^{22}Ne, the product of the $3n$ evaporation channel. Comparing the total number of ^{216}Ac α particles and the observed number of EVR's correlated with them, we measured the EVR detection efficiency to be $\approx$75%. In this case, the initial ^{216}Ac EVR energy of 11 MeV was reduced to $\sim$5 MeV at implantation due to losses in the target, hydrogen gas and the TOF module. In the ^{244}Pu+^{22}Ne reaction the initial EVR energy of 9.5 MeV was reduced to $\sim$2.5 MeV of implantation energy. Furthermore, due to the pulse-height defect of the PSD array, most of the signals from the $Z=104$ implants were reduced below the detection threshold set at $\sim$1 MeV.

To test the collection efficiency of the separator, we performed a model bombardment of ^{238}U with ^{22}Ne, detecting the known nuclei ^{254,255,256}No [12] implanted in the PSD. In the α-energy range of 7.60–8.34 MeV, covering the known α spectrum of ^{255}No, we detected a total of 1708 α's during out-of-beam periods. We observed about 110 correlated α-α pairs of the ^{254}No$\rightarrow$ ^{250}Fm$\rightarrow$ ^{246}Cf chain in which mother α decays with $E_{\alpha 1}=7.97$–8.18 MeV were followed by daughter α decays with $E_{\alpha 2}=7.31-7.52$ MeV. The half-life of the daughter α activity was calculated to be 23^{+16}_{-8} min [22] in agreement with the literature value of 30 ± 3 min [12] for ^{250}Fm. From this we estimated that α decays of ^{254}No, the product of the $6n$ evaporation channel, resulted in about 290 detected α's in the range of 7.60–8.34 MeV. We also detected 38 α's in the α-energy range of 8.34–8.50 MeV belonging to the product of the $4n$ evaporation channel, ^{256}No. By comparing the detected yield of ^{255}No with that calculated from the cross section value of Ref. [20], corrected by applying more recent nuclear data [12] on the EC branchings in the decays of ^{255}No and ^{251}Fm, the collection efficiency was determined to be $(5.4^{+1.4}_{-1.2})$%. Yields of the products of the $4n$- and $6n$-evaporation channels relative to that of the $5n$ channel were calculated to be $0.016^{+0.004}_{-0.002}$ and $0.14^{+0.10}_{-0.05}$, respectively, in agreement with the results given in Ref. [20].

III. RESULTS AND DISCUSSION

A. Production and decay properties of ^{261}Rf and ^{257}No

The production of ^{261}Rf in the ^{244}Pu+^{22}Ne reaction was clearly observed in the energy spectra of α particles detected at both bombarding energies used. The group of α particles with $E_\alpha\approx8.20-8.40$ MeV from α decays of ^{261}Rf and ^{257}No was the only α group in the E_α range $\geqslant$7.6 MeV. Since the α-particle spectra measured at the bombarding energies 114 and 120 MeV were rather similar, in Fig. 1 we show the sum energy spectrum of α particles detected by the whole PSD array during the entire measurement time of 260 h. Note that most α particles with E_α below 7.6 MeV (e.g., ^{211}Po) originate in the decays of long-lived activities produced in the ^{197}Au+^{22}Ne reaction used for calibrations, rather than from reactions with impurities in the targets.

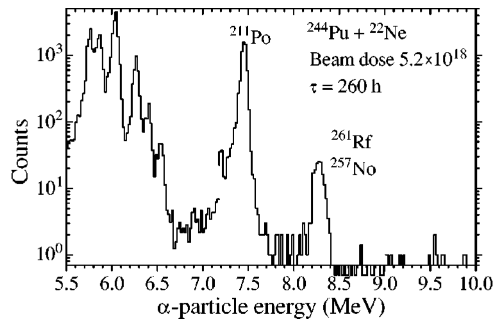

FIG. 1. Sum energy spectrum of α particles detected out of beam in the ^{244}Pu+^{22}Ne reaction at the bombarding energies 114 and 120 MeV.

We conducted an off-line search for correlated α-α event pairs with both α particles detected out of beam in the α-energy range $E_\alpha\geqslant$7.6 MeV within the time window $\Delta t=1800$ s; the y-position deviations between two α events were required to be within 1.2 mm. At the bombarding energy 120 MeV we observed 17 correlated α-α pairs in which mother α decays with $E_{\alpha 1}=8.22-8.41$ MeV were followed within time intervals of 0.4–181 s by daughter α decays with $E_{\alpha 2}=8.07-8.37$ MeV. At the bombarding energy 114 MeV we observed 25 pairs of α events with $E_{\alpha 1}=8.25-8.36$ MeV and $E_{\alpha 2}=8.21-8.40$ MeV, correlated within time intervals of 0.2 to 129 s. No α-α correlations were found in the Δt range of 181–1800 s, with the exception of one α-α pair with $E_{\alpha 1}=8.83$ MeV, $E_{\alpha 2}=8.09$ MeV, and $\Delta t=223$ s, which was observed at the bombarding energy 114 MeV and we attribute to the α-decay chain of ^{259}Rf produced in reactions with the 1.1% admixture of ^{242}Pu in the ^{244}Pu target material. From our observations, it follows that the number of α-α pairs from genetically unrelated α-decay events of ^{261}Rf and ^{257}No is less than 0.1 within the time window of 200 s, which should be compared with the observed total of 42 α-α chains formed by genetically linked α decays of these two nuclides. A total of 245 single out-of-beam α events detected in the E_α range of 8.06–8.42 MeV is in good correspondence with the number of 252 single α events from ^{261}Rf and ^{257}No expected on the basis of the 42 observed α-α correlations. The correlation times measured for the 42 α-α chains give a maximum-likelihood half-life of 25 ± 4 s for ^{257}No.

We also searched for correlated α-α event pairs from the ^{261}Rf$\rightarrow$^{257}No$\rightarrow$^{253}Fm chain, in which only one of the two α events was detected out of beam. To avoid a contribution from random α-α correlations, in this case we required the y-position deviations between the two α events to be within 0.5 mm; the search was made within the time window $\Delta t=500$ s. The α energies were allowed to vary between 8.00–8.70 MeV and 8.00–8.50 MeV for out-of-beam ^{261}Rf- and ^{257}No-like members of α-α chains, respectively. For in-beam events, the corresponding α energy ranges were restricted to 8.22–8.41 MeV and 8.19–8.40 MeV, thus limit-

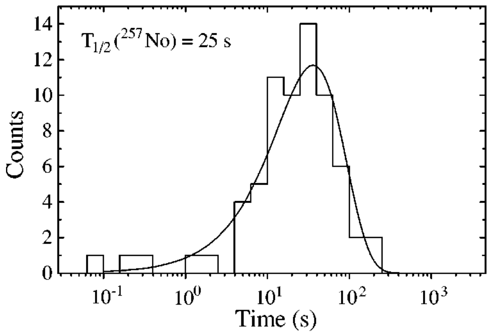

FIG. 2. Sum distribution of time intervals from correlated $^{261}Rf \rightarrow ^{257}No \rightarrow ^{253}Fm$ α-α pairs. The calculated distribution for $T_{1/2} = 25$ s is also shown.

ing the E_α values to those observed for ^{261}Rf and ^{257}No from the 41 α-α chains with both α events detected out of beam (with the exception of a single correlation including a ^{257}No event with $E_\alpha = 8.07$ MeV). As a result of the above-described selection, we observed an additional 27 α-α correlations in which α events with $E_{\alpha 1} = 8.24 - 8.40$ MeV were followed within time intervals of 0.08–192 s by daughter α decays with $E_{\alpha 2} = 8.19 - 8.38$ MeV. From the correlation times measured for these 27 α-α chains, we obtain a half-life value of 20^{+6}_{-5} s for ^{257}No. Two α-α pairs with correlation times above 300 s were found, indicating a possible presence of a few random pairs among the above 27 α-α correlations.

In Fig. 2 we show the distribution of time intervals from all of the 69 α-α correlations. In general, the distribution of the measured decay times fits the decay pattern of a single activity; however, three events in Fig. 2 show decay times of 0.08, 0.21, and 0.38 s, which are several hundred times shorter than expected from the ^{257}No lifetime. This observation might hint at the existence of a much shorter-lived state in ^{257}No decaying with similar α-particle energies, although the limited statistics do not allow more conclusive statements. Considering all of the measured decay times, we calculate a half-life of 25 ± 3 s for ^{257}No, in agreement with the accepted value of 25 ± 2 s [12].

The α-particle energy spectra of ^{261}Rf and ^{257}No based on the observed 69 genetically linked α decays of these two nuclides are plotted in Fig. 3. The correlation technique used in our work made it possible to measure for the first time the α-energy spectrum of ^{261}Rf without any contribution from α decays of its daughter. The measured α-particle energy spectrum of ^{261}Rf is relatively narrow and can be well fitted by a single Gaussian curve with a standard deviation $\sigma = 33$ keV, centered at $E_\alpha = 8.30$ MeV; from calibrations we estimated the averaged α-energy peak σ value to be 27 ± 6 keV. Our data (see Fig. 3) show no α-decays which correspond to the 8.52-MeV α events of ^{261}Rf, one observed by the GSI group in one of two $Z = 112$ decay sequences [23] and one reported from chemistry experiments with ^{265}Sg [24].

The α-particle energy spectrum that we measured for decays of ^{257}No is in good agreement with the α-energy pattern observed for this nuclide in previous experiments [11]. As shown in Fig. 3, this spectrum can be fitted by the sum of three Gaussian curves with $\sigma = 28$ keV, defining the three

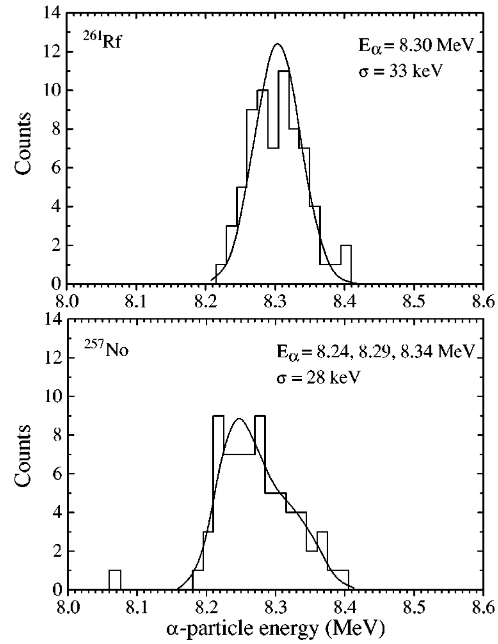

FIG. 3. Experimental α-particle spectra of ^{261}Rf and ^{257}No based on the genetically linked α decays with corresponding fitted spectra.

main α groups of ^{257}No with $E_\alpha = 8.24$, 8.29, and 8.34 MeV, with relative intensities of 55, 26, and 19%, respectively, according to Ref. [11]. One α-decay event was detected with $E_\alpha = 8.07$ MeV, implying the existence of a weak α group of ^{257}No; this 8.07-MeV α event followed a preceding 8.34-MeV α event from ^{261}Rf by 0.38 s. Note that lower α-particle energies of ^{257}No, down to $E_\alpha = 8.11$ MeV, were also observed in experiments [2,17], although using detectors with poorer energy resolution.

We searched for correlated α-SF events with out-of-beam α particles in the energy range $E_\alpha \geqslant 7.6$ MeV and SF events detected both out of beam and in beam. No correlations were found within the time window $\Delta t = 1000$ s. The y-position deviations between α and SF events were required to be within 1.2 mm. On the basis of the total number of α particles with $E_\alpha = 8.06 - 8.42$ MeV and 42 observed α-α correlations of ^{261}Rf and ^{257}No, we calculated a 68% confidence level upper limit of 1.5% for the SF branch of ^{257}No.

B. Spontaneous-fission activities from the $^{244}Pu + ^{22}Ne$ reaction and ^{262}Rf decay

In the reaction $^{244}Pu + ^{22}Ne$ we detected 239 out-of- and in-beam SF events at the ^{22}Ne energy of 114 MeV and 303 SF events at 120 MeV. Analyzing the EVR-SF correlation time distributions, we singled out two short-lived activities. At both energies a SF activity with $T_{1/2} = 1.4^{+0.3}_{-0.2}$ ms was observed, being the source of about 45 events at the ^{22}Ne energy of 114 MeV and of about 42 events at 120 MeV. Another SF activity, with $T_{1/2} = 26^{+10}_{-6}$ ms, contributed ~ 12 and ~ 22 events at 114 and 120 MeV, respectively. These numbers do not represent the complete contribution from the corresponding activities since the detection efficiency of

EVR's was less than 100%. The wide distribution of these events on the detector area also hints that the total yield of observed activities could be higher.

One of the probable sources of the 1.4-ms SF activity is ^{258}No produced in the $\alpha 4n$-emission channel. Note that recently the product of an $\alpha 3n$ channel, ^{259}No, was observed in this reaction with the cross section in the range of 6.4–3.1 nb [25] corresponding to the ^{22}Ne bombarding energies of 110.9–129.2 MeV. However, the average measured energy of the 1.4-ms recoils appeared to be two times higher than expected for ^{258}No, $\sim$2.3 MeV, which means $\sim$5-MeV implantation energy. The spontaneously fissioning isomer ^{244mf}Am produced in transfer reactions could possibly contribute to this activity but, as it will be shown below, we did not observe noticeable yield of a similar pn-transfer product — the isomer ^{242mf}Am in the ^{242}Pu+^{22}Ne reaction. One of the probable sources of 26-ms SF activity is spontaneous fission of ^{260}Rf, a $6n$-evaporation product, especially at the ^{22}Ne energy of 120 MeV (see Sec. III C). Contributions from the decays of hypothetical spontaneously fissioning $Z=104$ isomers cannot be excluded; the above assignments remain somewhat speculative.

To probe the α-decay branch of the even-even nuclide ^{262}Rf we searched for correlations between α decays of ^{262}Rf and subsequent SF events from its short-lived spontaneously fissioning daughter ^{258}No. The theoretical Q_α value for ^{262}Rf of $\approx$8.25 MeV [3] corresponds to a partial α-decay half-life of approximately 200 s. For potential ^{262}Rf decays, we considered the α-energy range $E_\alpha=7.8-8.5$ MeV, which corresponds to the partial α-decay half-life range of 10–3000 s [3]. We found no α-SF correlations with α particles and SF events detected both out-of- and in-beam within the time window of 10 s.

The low implantation energy of $Z=104$ EVR's in the PSD (see Sec. II) prevented us from distinguishing SF events of 1.2-s ^{262}Rf and determining its yield directly from the EVR-SF correlation data. To estimate the yield of ^{262}Rf we used experimental ratios of $5n$- and $4n$- evaporation cross sections measured at the ^{22}Ne bombarding energies close to the $4n$-evaporation maximum in the reactions ^{236,238}U $+^{22}$Ne [26,20], ^{242}Pu+^{22}Ne (present work), and ^{248}Cm $+^{22}$Ne [2]. From these data, we assumed the $4n$-evaporation cross section to be half of that of the $5n$ channel at the ^{22}Ne energy of 114 MeV. Using these assumptions and estimating that 113 SF events from ^{262}Rf occurred, we set the 68% confidence level upper limit at 3% for the α-decay branch of ^{262}Rf. Both the low α branching and the predominance of SF in the decay of ^{262}104 (and all other even-even Rf isotopes) is a very definite prediction of theory [3–5].

The group of LBNL, Berkeley, studied the reaction ^{244}Pu+^{22}Ne by using the gas-jet technique coupled with a rotating wheel system [27]. No α decays correlated with SF from ^{258}No were observed and an upper limit of 0.8% for the α-decay branch of ^{262}Rf was set. It should be noted that the authors of Ref. [27] assigned all the 200 observed SF events with an apparent half-life $T_{1/2}=2.1\pm0.2$ s and maximum production cross section of $\sim$0.7 nb at $E(^{22}\text{Ne})=114.4$ MeV to the ground-state decay of ^{262}Rf on the basis of excitation function measurements. Mass and kinetic-energy distributions of coincident fission fragments were measured. The possible interfering SF background, which would raise the reported α-decay branch limit, was considered to be low enough to ascribe all the observed fission activity to ^{262}Rf.

C. Production of Rf isotopes in the ^{242}Pu+^{22}Ne and ^{238}U+^{26}Mg reactions

We also studied the production of the Rf isotopes in the reactions ^{242}Pu+^{22}Ne and ^{238}U+^{26}Mg, resulting from the same compound nucleus ^{264}Rf. One of the aims of these experiments was to compare production and detection rates of the Rf nuclides in these two reactions in order to choose between the ^{22}Ne- and ^{26}Mg-induced reactions for the synthesis of new Sg isotopes. The ^{22}Ne beam energy was chosen to correspond to the maximum of the $4n$ evaporation channel excitation function, and the two bombarding energies of ^{26}Mg ions were chosen to be close to the expected maxima of the $4n$ and $5n$ evaporation reactions. Correlation data and fission activities from these experiments are summarized in Table II.

The well-known isotope ^{259}Rf [12], the product of the $5n$-evaporation channel, was identified by detecting 17 out-of-beam α-α correlations with ^{255}No decays in the reaction ^{242}Pu+^{22}Ne and 5 α-α correlations in the ^{238}U+^{26}Mg reaction. In the last case, due to higher EVR detection efficiency, we also observed 22 EVR-α correlations of ^{259}Rf. Measured cross sections of the $5n$-evaporation channel for both these reactions are listed in Table I. Even at the lower bombarding energy, corresponding to the maximum of the $4n$-evaporation channel, the cross section of the ^{242}Pu(^{22}Ne,$5n$)^{259}Rf reaction is larger than that of the ^{238}U(^{26}Mg,$5n$)^{259}Rf reaction at its expected peak by a factor of approximately 1.5.

In both reactions we observed SF activities. We could single out 22 fission events correlated with preceding EVR's in the ^{242}Pu+^{22}Ne experiment and 4 such events in the ^{238}U+^{26}Mg reaction which show an identical half-life of about 20–30 ms. After the end of the ^{242}Pu+^{22}Ne experiment, off-line measurements were performed to look for long-lived α activities implanted in the detectors. From the upper limit for ^{242}Cm production, we estimated the possible contribution from the 14-ms isomer ^{242mf}Am to be three SF events or less. Thus the observed 20–30-ms SF activity could be tentatively assigned, based on literature data [12], to the spontaneous fission of ^{260}Rf, the $4n$-evaporation product. In the reaction ^{242}Pu+^{22}Ne, no correlations were found between out-of-beam α particles with $E_\alpha=8.25-8.60$ MeV characteristic of ^{256}No [12,28] and preceding α events with $E_\alpha\geqslant7.6$ MeV within a time interval of 100 s. An upper limit of 20% follows for the α-decay branch of ^{260}Rf.

In both reactions a few SF events can be attributed to a shorter-lived activity, with a half-life of $\sim$0.1 ms. The 19 SF events observed in the reaction ^{238}U+140-MeV ^{26}Mg show a half-life of 12^{+4}_{-2} ms and a production cross section of 0.3 nb. Their most probable source is the product of the $6n$-evaporation channel, ^{258}Rf [12], although a contribution from ^{260}Rf cannot be excluded.

TABLE II. Summary of observations from $^{242}Pu+^{22}Ne$ and $^{238}U+^{26}Mg$ reactions.

Reaction	Projectile energy (MeV)	Number of correlated events[a]	Measured half-life	Assignment
$^{242}Pu+^{22}Ne$	114[b]	17 α-α	$3.2^{+1.6}_{-0.9}$ min	$^{259}Rf \rightarrow ^{255}No \rightarrow ^{251}Fm$
	114[b]	22 EVR-SF	21^{+17}_{-9} ms	^{260}Rf[c]$\rightarrow$
$^{238}U+^{26}Mg$	134[b]	4 EVR-SF	28^{+39}_{-13} ms	^{260}Rf[c] $\rightarrow$
	140	22 EVR-α	$2.5^{+1.1}_{-0.7}$ s	$^{259}Rf \rightarrow$
	140	5 α-α	$5.1^{+3.8}_{-1.6}$ min	$^{259}Rf \rightarrow ^{255}No \rightarrow ^{251}Fm$
	140	19 EVR-SF	12^{+4}_{-2} ms	^{258}Rf[c] $\rightarrow$

[a]The detection efficiencies of the Rf EVR's were about 30% in the reaction $^{242}Pu+^{22}Ne$ and 60% in the $^{238}U+^{26}Mg$ reaction (due to higher EVR initial and, consequently, implantation energy).
[b]A detector array of six Si(Au) detectors of 3-cm high $\times$ 2-cm wide without position sensitivity was employed for these irradiations.
[c]Tentative assignments of EVR-SF sequences are based on literature data [12].

IV. CONCLUSION

In bombarding ^{244}Pu targets with 114- and 120-MeV ^{22}Ne projectiles, we detected 69 α-α correlations linking α decays of ^{261}Rf and ^{257}No. The correlation technique used in our work made it possible to measure the α-energy spectrum of ^{261}Rf without any contribution from the α decays of its daughter. We observed a single α peak with $E_\alpha = 8.30 \pm 0.06$ MeV for ^{261}Rf. The half-life of ^{257}No was measured to be 25 ± 3 s. The cross section of the $^{244}Pu(^{22}Ne,5n)^{261}Rf$ reaction was estimated to be about 4 nb at both ^{22}Ne energies. The α-particle energies of 8.19 to 8.40 MeV that we measured for ^{257}No are in agreement with the pattern observed for this nuclide in previous experiments. One α-decay event of ^{257}No was detected with $E_\alpha = 8.07$ MeV; this implies the existence of a weak α group of ^{257}No. No correlations were found between α decays with $E_\alpha \geqslant 7.6$ MeV and subsequent spontaneous fission events, from which we calculated an upper limit of 1.5% for the spontaneous-fission branch of ^{257}No and estimated an upper limit of 3% for the α-decay branch of ^{262}Rf.

In all three reactions studied we observed several SF activities that we could not assign to specific nuclides. Our earlier experiments on the synthesis of ^{266}Sg established unambiguous assignment of the ground-state SF decay of ^{262}Rf following the α decay of the mother nucleus. Similar experiments can produce lighter even-even Rf isotopes that may be the origin of some of the unidentified SF activities reported here. Since both ^{262}Sg and ^{264}Sg are expected to have significant α-decay branches [3–5], their α decay should lead to the even-even daughters ^{258}Rf and ^{260}Rf, which are assigned to short-lived SF activities. The new Sg isotopes could be produced, e.g., in complete fusion reactions $^{242,244}Pu+^{25,26}Mg$ or $^{246,248}Cm+^{22}Ne$ with evaporation of four, five, and probably six neutrons. These experiments will allow the first identification of the ^{258}Rf and ^{260}Rf using decay sequences that are correlated in position and time. The decay properties of these nuclides are important for investigating trends in properties of the $Z=104$ and 106 isotopes with neutron numbers increasing towards $N=162$.

ACKNOWLEDGMENTS

We wish to thank the U400 cyclotron staff for providing the intense ^{22}Ne and ^{26}Mg beams. Our thanks are due to V. I. Krashonkin, V. I. Tomin, and A. M. Zubareva for their essential help. The ^{244}Pu target material was provided by the U.S. DOE through ORNL. Much of the support for the LLNL authors was provided through the U.S. DOE under Contract No. W-7405-Eng-48. The work was supported by Grant No. 96-02-17377 from the Russian Foundation for Basic Research. These studies were performed in the framework of the Russian Federation/U.S. Joint Coordinating Committee for Research on Fundamental Properties of Matter.

[1] A. Ghiorso, M. Nurmia, K. Eskola, and P. Eskola, Phys. Lett. **32B**, 95 (1970).
[2] Yu. A. Lazarev, Yu. V. Lobanov, Yu. Ts. Oganessian, V. K. Utyonkov, F. Sh. Abdullin, G. V. Buklanov, B. N. Gikal, S. Iliev, A. N. Mezentsev, A. N. Polyakov, I. M. Sedykh, I. V. Shirokovsky, V. G. Subbotin, A. M. Sukhov, Yu. S. Tsyganov, V. E. Zhuchko, R. W. Lougheed, K. J. Moody, J. F. Wild, E. K. Hulet, and J. H. McQuaid, Phys. Rev. Lett. **73**, 624 (1994).
[3] R. Smolańczuk, J. Skalski, and A. Sobiczewski, in *Proceedings of the International Workshop XXIV on Gross Properties of Nuclei and Nuclear Excitations ''Extremes of Nuclear Structure,''* Hirschegg, Austria, 1996, edited by H. Feldmeier, J. Knoll, and W. Nörenberg (GSI, Darmstadt, 1996), p. 35.
[4] Z. Patyk and A. Sobiczewski, Nucl. Phys. **A533**, 132 (1991).

[5] R. Smolańczuk, J. Skalski, and A. Sobiczewski, Phys. Rev. C **52**, 1871 (1995).

[6] P. Möller and J. R. Nix, J. Phys. G **20**, 1681 (1994).

[7] Yu. A. Lazarev, Yu. V. Lobanov, Yu. Ts. Oganessian, Yu. S. Tsyganov, V. K. Utyonkov, F. Sh. Abdullin, S. Iliev, A. N. Polyakov, J. Rigol, I. V. Shirokovsky, V. G. Subbotin, A. M. Sukhov, G. V. Buklanov, B. N. Gikal, V. B. Kutner, A. N. Mezentsev, I. M. Sedykh, D. V. Vakatov, R. W. Lougheed, J. F. Wild, K. J. Moody, and E. K. Hulet, Phys. Rev. Lett. **75**, 1903 (1995).

[8] Yu. A. Lazarev, Yu. V. Lobanov, Yu. Ts. Oganessian, V. K. Utyonkov, F. Sh. Abdullin, A. N. Polyakov, J. Rigol, I. V. Shirokovsky, Yu. S. Tsyganov, S. Iliev, V. G. Subbotin, A. M. Sukhov, G. V. Buklanov, B. N. Gikal, V. B. Kutner, A. N. Mezentsev, K. Subotic, J. F. Wild, R. W. Lougheed, and K. J. Moody, Phys. Rev. C **54**, 620 (1996).

[9] Yu. A. Lazarev, in *Proceedings of the International Workshop XXIV on Gross Properties of Nuclei and Nuclear Excitations ''Extremes of Nuclear Structure,''* Hirschegg, Austria, 1996, [3], p. 11.

[10] A. Ghiorso, T. Sikkeland, and M. J. Nurmia, Phys. Rev. Lett. **18**, 401 (1967).

[11] P. Eskola, K. Eskola, M. Nurmia, and A. Ghiorso, Phys. Rev. C **2**, 1058 (1970).

[12] *Table of Isotopes* 8th ed., edited by R. B. Firestone and V. S. Shirley (Wiley, New York, 1996).

[13] R. Silva, J. Harris, M. Nurmia, K. Eskola, and A. Ghiorso, Inorg. Nucl. Chem. Lett. **6**, 871 (1970).

[14] E. K. Hulet, R. W. Lougheed, J. F. Wild, J. H. Landrum, J. M. Nitschke, and A. Ghiorso, J. Inorg. Nucl. Chem. **42**, 79 (1980).

[15] B. Kadkhodayan, A. Türler, K. E. Gregorich, P. A. Baisden, K. R. Czerwinski, B. Eichler, H. W. Gäggeler, T. M. Hamilton, D. T. Jost, C. D. Kacher, A. Kovacs, S. A. Kreek, M. R. Lane, M. F. Mohar, M. P. Neu, N. J. Stoyer, E. R. Sylwester, D. M. Lee, M. J. Nurmia, G. T. Seaborg, and D. C. Hoffman, Radiochim. Acta **72**, 169 (1996).

[16] L. P. Somerville, M. J. Nurmia, J. M. Nitschke, A. Ghiorso, E. K. Hulet, and R. W. Lougheed, Phys. Rev. C **31**, 1801 (1985).

[17] A. Türler, R. Dressler, B. Eichler, H. W. Gäggeler, D. T. Jost, M. Schädel, W. Brüchle, K. E. Gregorich, N. Trautmann, S. Taut, Phys. Rev. C **57**, 1648 (1998).

[18] E. K. Hulet, J. F. Wild, R. J. Dougan, R. W. Lougheed, J. H. Landrum, A. D. Dougan, P. A. Baisden, C. M. Henderson, R. J. Dupzyk, R. L. Hahn, M. Schädel, K. Sümmerer, and G. R. Bethune, Phys. Rev. C **40**, 770 (1989).

[19] Yu. A. Lazarev, Yu. V. Lobanov, Yu. Ts. Oganessian, Yu. S. Tsyganov, V. K. Utyonkov, F. Sh. Abdullin, G. V. Buklanov, B. N. Gikal, S. Iliev, A. N. Mezentsev, A. N. Polyakov, I. M. Sedykh, I. V. Shirokovsky, V. G. Subbotin, and A. M. Sukhov, Heavy Ion Physics, Scientific Report 1991–1992 (JINR Report No. E7-93-57, Dubna, 1993), p. 39.

[20] E. D. Donets, V. A. Schegolev, and V. A. Ermakov, Yad. Fiz. **2**, 1015 (1965) [Sov. J. Nucl. Phys. **2**, 723 (1965)].

[21] Yu. A. Lazarev, Yu. V. Lobanov, A. N. Mezentsev, Yu. Ts. Oganessian, V. G. Subbotin, V. K. Utyonkov, F. Sh. Abdullin, V. V. Bekhterev, S. Iliev, I. V. Kolesov, A. N. Polyakov, I. M. Sedykh, I. V. Shirokovsky, A. M. Sukhov, Yu. S. Tsyganov, and V. E. Zhuchko, Heavy Ion Physics, Scientific Report 1991-1992 (JINR Report No. E7-93-57, Dubna, 1993), p. 203; *Proceedings of the International School-Seminar on Heavy Ion Physics*, Dubna, 1993 (JINR Report No. E7-93-274, Dubna, 1993), Vol. 2, p. 497.

[22] V. B. Zlokazov, Nucl. Instrum. Methods **151**, 303 (1978); K.-H. Schmidt, C.-C. Sahm, K. Pielenz, and H.-G. Clerc, Z. Phys. A **316**, 19 (1984).

[23] S. Hofmann, V. Ninov, F. P. Hessberger, P. Armbruster, H. Folger, G. Münzenberg, H. J. Schött, A. G. Popeko, A. V. Yeremin, S. Saro, R. Yanik, and M. Leino, Z. Phys. A **354**, 229 (1996).

[24] H. W. Gäggeler, Paul Scherer Institut Preprint PSI-PR-97-19; M. Schädel, W. Brüchle, R. Dressler, B. Eichler, H. W. Gäggeler, R. Günther, K. E. Gregorich, D. C. Hoffman, S. Hübener, D. T. Jost, J. V. Kratz, W. Paulus, D. Schumann, S. Timokhin, N. Trautmann, A. Türler, G. Wirth, and A. Yakuschev, Nature (London) **388**, 63 (1997).

[25] R. Dressler, Ph.D. thesis, University of Bern, 1999.

[26] A. N. Andreyev, D. D. Bogdanov, V. I. Chepigin, A. P. Kabachenko, O. N. Malyshev, R. N. Sagaidak, L. I. Salamatin, G. M. Ter-Akopian, and A. V. Yeremin, Z. Phys. A **345**, 389 (1993).

[27] M. R. Lane, K. E. Gregorich, D. M. Lee, M. F. Mohar, M. Hsu, C. D. Kacher, B. Kadkhodayan, M. P. Neu, N. J. Stoyer, E. R. Sylwester, J. C. Yang, and D. C. Hoffman, Phys. Rev. C **53**, 2893 (1996).

[28] D. C. Hoffman, D. M. Lee, K. E. Gregorich, M. J. Nurmia, R. B. Chadwick, K. B. Chen, K. R. Czerwinski, C. M. Gannett, H. L. Hall, R. A. Henderson, B. Kadkhodayan, S. A. Kreek, and J. D. Leyba, Phys. Rev. C **41**, 631 (1990).