New Nuclide $^{267}108$ Produced by the $^{238}U + {}^{34}S$ Reaction

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(Received 14 March 1995)

In bombardments of ^{238}U targets with 186-MeV ^{34}S projectiles we discovered the α-decaying nuclide $^{267}108$ with a half-life of 19^{+29}_{-10} ms, $E_\alpha = 9.74$ to 9.87 MeV, and a production cross section of about 2.5 pb. The new nuclide was identified by measuring correlations in energy, time, and position to establish genetic links between its implantation in a position-sensitive silicon detector and subsequent α decay followed by α decays of known descendant nuclides.

PACS numbers: 25.70.Gh, 23.60.+e, 25.85.Ca, 27.90.+b

The ground-state decay properties of the heaviest $Z = 106$ isotopes, $^{265}106$ ($T_{1/2} \approx 2-30$ s) and $^{266}106$ ($T_{1/2} \approx 10$–30 s, $b_{sf} \lesssim 50\%$) [1], revealed a large enhancement in their stability as compared to that of nuclides with *lower* Z and N values. For example, at $N = 160$ the transition from $^{262}102$ ($T_{1/2} \approx T_{sf} = 5$ ms [2]) to $^{266}106$, an increase of four protons, increases the stability against spontaneous fission (SF) by a factor of $\gtrsim 4 \times 10^3$. Similarly, in going from $^{260}106$ ($T_{1/2} \approx 3.6$ ms, $b_{sf} = b_\alpha = 50\%$ [3,4]) to $^{266}106$, the stability increases by factors of $\gtrsim 3 \times 10^3$ for SF decay and $\sim 3 \times 10^3$ for α decay. Thus the ground-state decay properties of $^{266}106$ provide a strong indication of the existence of deformed shell closures near $N = 162$ and $Z = 108$ that were predicted by macroscopic-microscopic calculations (see, e.g., Refs. [5–10]).

We report here on experiments designed to explore further the nuclear stability near $N = 162$ and $Z = 108$ by producing new heavy isotopes of element 108 with $N = 159$ and $N = 160$ in the complete fusion reaction $^{238}U + {}^{34}S$. Another goal of our experiments was to probe cross section values for the actinide-target-based fusion-evaporation reactions leading to the $Z = 108$ nuclides. Preliminary results from these experiments carried out in March–April 1994 were included in Ref. [11]. Prior to our work, two isotopes of element 108 had been produced in "cold fusion" reactions [12,13]. These are the even-even isotope $^{264}108$ with $T_{1/2} \approx 0.08$ ms, identified by one decay sequence [12], and the odd-A α emitter $^{265}108$ with $T_{1/2} = 1.8$ ms, for which three correlated α-decay sequences were observed [12].

The even-even nuclide $^{268}108$ is predicted to be an α emitter with $T_{1/2} \sim 0.1$ s [7–9]. Its partial SF half-life was predicted to be of the order of 10^2 s [9]. Thus a signature for $^{268}108$ would be the observation of α decay with the α-particle energy $E_\alpha \sim 9.7$ MeV [7] followed by SF decay of the unknown daughter $^{264}106$, or eventually by SF decay of the short-lived granddaughter $^{260}104$, if $^{264}106$ undergoes α decay. Considering decay properties observed for $^{264,265}108$ [12], as well as those predicted for $^{266,268}108$ [7–9], we expect a signature for $^{267}108$ to be the observation of α decay with $E_\alpha \sim 10$ MeV, followed by α decays of the known nuclides $^{263}106$ ($T_{1/2} = 0.9$ s, $E_\alpha = 8.95$ to 9.10, 9.25 MeV [14,15]), $^{259}104$ ($T_{1/2} = 3.1$ s, $b_\alpha = 93\%$, $E_\alpha = 8.77$, 8.87 MeV, $b_{sf} = 7\%$ [16]), and $^{255}102$ ($T_{1/2} = 3.1$ min, $b_\alpha = 61.4\%$, $E_\alpha = 7.62$ to 8.31 MeV, $b_{EC} = 38.6\%$ [16]).

We used a ^{34}S bombarding energy of 186 MeV, 5 MeV above the Bass fusion barrier [17], to produce $Z = 108$ compound nuclei with an excitation energy of ≈ 50 MeV, sufficient for the evaporation of four or five neutrons. Beams of ^{34}S projectiles were delivered by the Dubna U400 cyclotron. The time structure of the pulsed ^{34}S beam was determined by the cyclotron modulating frequency of 150 Hz and a duty factor of $\approx 40\%$, which corresponds to a beam cycle of 6.7 ms and a beam pulse duration of ≈ 2.7 ms. Six uranium targets electrodeposited on 0.70 mg cm^{-2} Ti substrates with average areal densities of 0.54 mg cm^{-2} of ^{238}U (99.9%) and a total area of 36 cm^2 were arranged on a wheel rotating at 3000 rpm. In an 860 h bombardment, with an average intensity of 6×10^{12} particles/s of ^{34}S, the targets received a total beam dose of 1.7×10^{19} particles.

Evaporation residues (EVR's) recoiling out of the ^{238}U targets were separated in flight from beam particles and various transfer-reaction products by the Dubna Gas-filled Recoil Separator described in Ref. [18]. The separator was filled with hydrogen at a pressure of 1.0 Torr. The field B of the separator's dipole magnet was adjusted to center the quasi-Gaussian distribution of EVR's on the focal-plane detector in the horizontal direction. To set the B value for the $Z = 108$ EVR's with the expected average velocity $\langle v/v_0 \rangle \approx 1.7$ ($v_0 = 2.2 \times 10^6$ m s^{-1} is

0031-9007/95/75(10)/1903(4)$06.00

the Bohr velocity), we used prior measurements [1,18] of the average charge states $\langle q \rangle$ for the EVR's with $Z = 89$ through 106, as well as new $\langle q \rangle$ data from various calibration bombardments performed in the present work (see Fig. 1). The separated EVR's passed through a time-of-flight (TOF) measurement system composed of two multiwire proportional chambers in a 1.5 Torr pentane-filled module and were finally implanted in a 120×40 mm^2 position-sensitive detector (PSD) array.

The PSD array consisted of three 40×40 mm^2 passivated boron implanted silicon detectors produced by Canberra Semiconductor NV, with each detector having 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 fully depleted detectors. Top and bottom or y-position signals from each strip were each divided into a signal for α/implant events ($\sim$2–12 MeV) and a signal for SF events ($\sim$20–250 MeV). We also recorded the energy sum of the α/implant signals; we determined the total energy of SF events by an 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/implant or fission events, and the time since the beginning of the data acquisition cycle in 0.1 ms intervals. The data were acquired in list mode.

α-energy calibrations were periodically performed for each strip using the α peaks from nuclides produced in the $^{\mathrm{nat}}$W + ^{34}S reaction. Most of the strips had α-energy peak FWHM's of about 70 keV (an average value for the 36 day bombardment during which there was degradation of the detector performance due to radiation damage). An approximate fission-energy calibration was obtained by extrapolating the α-energy calibration, as well as by detecting the known SF activities 2.3 s ^{252}102 [16] and 21 ms ^{238}Cf [19] produced in the ^{206}Pb + ^{48}Ca and ^{207}Pb + ^{34}S reactions, respectively. These reactions were also used to estimate the collection efficiency of the $Z = 108$ EVR's and the range of their energies measured by the PSD. By using known α-decay sequences from the calibration reactions, we estimated the FWHM y-position deviation Δpos to be 1.2 mm (3% of the strip height) for α-α correlations. For EVR-α and EVR-SF correlations, Δpos depends significantly on EVR energies measured by the PSD. Our calibration EVR-α data for ^{217}Th produced in the $^{\mathrm{nat}}$W + ^{34}S and $^{\mathrm{nat}}$Pt + ^{26}Mg reactions show an increase in Δpos from 1.2 to 3.5 mm with a decrease in the average measured EVR energy from $\approx$8 to $\approx$3 MeV, which spans the expected average of 6 MeV and a FWHM of 3 MeV for the Gaussian-like distribution of measured $Z = 108$ EVR energies.

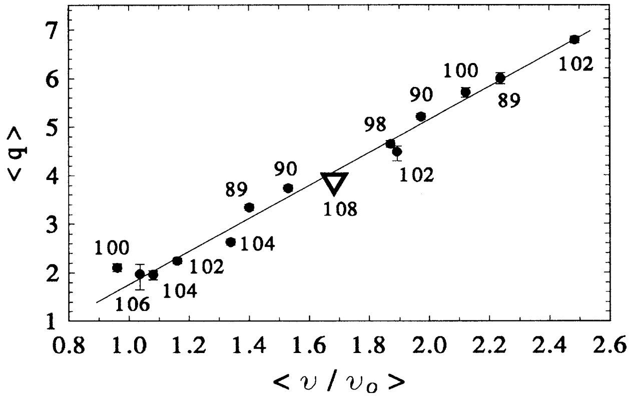

FIG. 1. The velocity dependence of the average charge states $\langle q \rangle$ of very heavy atoms traversing 1 Torr of hydrogen. Z values for atoms are given near the data points. The systematics is based on both previous data [1,18] and $\langle q \rangle$ measurements performed in this work. The triangle shows the $\langle q \rangle$ value for $Z = 108$. The line is included to guide the eye.

We searched off-line for event sequences in which $Z = 108$ EVR's were correlated in time and (x, y) position with subsequent α and/or SF decays. Table I lists the correlated event sequences that we attribute to $Z = 108$ implantation events followed by the detected α decays of ^{267}108. The out-of-beam α-particle energy spectrum is shown in Fig. 2 for all strips for the entire bombardment; in Fig. 3 we show, for each strip, the number of out-of-beam α decays in the energy range of interest, 7.6 to 10.4 MeV, an average rate of about four events per day per strip (the in-beam rate is about three events per minute per strip), resulting in a high statistical significance for event sequences with one or two α particles detected between cyclotron beam pulses. We examined the statistical significance of each observed

TABLE I. The measured parameters of the correlated ^{267}108 event sequences observed in the ^{238}U + ^{34}S reaction. All α-decay events are out of beam except for those two indicated. The accuracy of the measured α-particle energies is estimated to be ±60 keV.

Particle	Particle energy (MeV)	Strip no.	Time interval	Δpos^{c} (mm)
EVR^{a}	4.1	11		
α	9.74	11	29 ms	−1.0
α	7.75	11	9.1 min	−2.1
EVR	6.2	6		
α	9.86	6	32 ms	−0.6
α^{b}	8.80	6	3.6 s	−0.6
EVR	4.4	9		
α^{b}	9.87	9	20 ms	+0.2
α	8.80	9	2.0 s	+0.4

[a] The quoted EVR energies are measured values. No estimate was included for the pulse-height defect.

[b] Event occurred during the beam pulse.

[c] The y-position deviations are given with respect to the EVR implantation site.

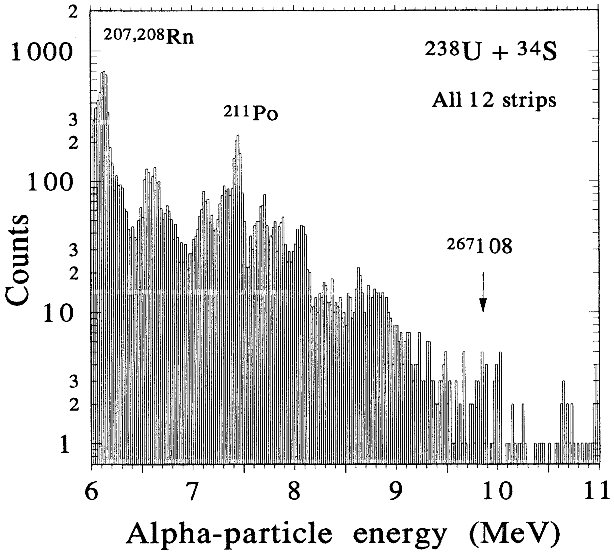

FIG. 2. Energy spectrum of out-of-beam α particles detected in all 12 strips in the 36-day bombardment of ^{238}U with 1.7×10^{19} particles of ^{34}S. Most α activities seen in the spectrum are due to $Z \geq 84$ nuclides produced in deep inelastic collisions between ^{238}U and ^{34}S, with possible traces of some α emitters from $^{\mathrm{nat}}$W + ^{34}S calibrations.

correlation by calculating from our data the number of random EVR-α-α sequences with similar properties N_b that are expected to occur for the whole PSD array during the entire measurement time $T = 860$ h. For example, in considering the event sequence with $E_{\alpha 1} = 9.87$ MeV, we calculated N_b for implantation events with measured EVR energies of 3.5 to 8.5 MeV that are followed within the time interval $\Delta t_1 = 20$ ms by an in-beam α decay with $E_{\alpha 1} = 9.4$ to 10.4 MeV and then, within the time interval $\Delta t_2 = 2.0$ s, by an out-of-beam α decay with $E_{\alpha 2} = 8.6$ to 9.0 MeV. Using the y-position window $|\Delta \mathrm{pos}| = 0.4$ mm, we determined $N_b < 0.0002$ for this case. Similar N_b calculations, with corresponding changes in energy, time, and position windows, were also performed for the EVR-α-α sequences with $E_{\alpha 1} = 9.74$ and 9.86 MeV, which resulted in $N_b < 0.02$ and < 0.003, respectively.

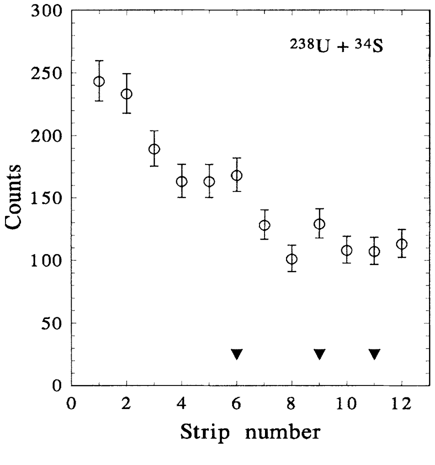

FIG. 3. The distribution across detector strips for all out-of-beam α-decay events with $E_\alpha = 7.6$ to 10.4 MeV detected in the ^{238}U + ^{34}S bombardment. The triangles show the location of the three $Z = 108$ event sequences.

We interpret the three correlated event sequences with $E_{\alpha 1} = 9.74, 9.86$, and 9.87 MeV as the decay of ^{267}108 followed by α decay of one of the descendant nuclides, ^{255}102 in the first sequence, and ^{259}104 in the two other sequences. We also observed two out-of-beam α-α correlations with $N_b < 0.03$ linking α decays of ^{263}106, ^{259}104, and ^{255}102. It is possible that these correlations originate from the decays of ^{267}108 where the ^{267}108 α particles escaped the PSD array.

The measured parameters of the correlated EVR-α-α sequences listed in Table I fully support the assignment of these sequences to the implantation in the PSD array and subsequent α decay of ^{267}108 EVR's. The 9.74 to 9.87 MeV α-particle energies are in good agreement with both predictions [7] and systematics (see, e.g., Fig. 5 in Ref. [12]). The measured $E_{\alpha 2}$ values, as well as the measured time intervals between the correlated α decays, agree with the α-particle energy and half-life values for ^{255}102 and ^{259}104 [16]. The measured EVR energies of 4.1 to 6.2 MeV are in the range of 6.0 ± 2.5 (2σ) MeV that is expected for $Z = 108$ EVR's on the basis of calibration measurements; the detection of the EVR's by the PSD was accompanied by TOF signals consistent with expectations for a compound nucleus. From our data, we estimated the average charge state $\langle q \rangle$ of $Z = 108$ EVR's in hydrogen to be 3.8 ± 0.2 at $\langle v/v_0 \rangle = 1.7$. This value is in good agreement with the $\langle q \rangle$ systematics shown in Fig. 1.

The above observations and arguments provide strong evidence for the identification of ^{267}108. From measured time intervals between implantation and α decay events of the ^{267}108 nuclides, we calculate a maximum likelihood half-life value of 19^{+29}_{-10} ms (see also Ref. [20]). The production cross section of the new isotope at the ^{34}S bombarding energy of 186 MeV is 2.5 pb, with an estimated accuracy of a factor of ~ 3. We note, however, that the *upper* confidence limit of the reported cross section value could be higher if we include an uncertainty in the collection efficiency due to the small number of observed $Z = 108$ events which are used to estimate how precisely the $Z = 108$ EVR distribution is centered in the horizontal direction at the focal plane.

We detected nine correlated EVR-SF event pairs, with time intervals of 0.08 to 5.6 ms between the pair members (we note that the dead time of the electronics system was about 55 μs in this experiment). We found no time and position correlations of both out-of-beam and in-beam SF events to preceding $Z = 108$ α decays. The probability density distribution of the time intervals in the observed EVR-SF correlations does not contradict the assumption of a single SF activity with a maximum likelihood half-life of $1.0^{+0.8}_{-0.3}$ ms, although a $\approx 20\%$ contribution from longer-lived SF species cannot be excluded. The apparent cross section corresponding to the 1 ms SF activity is of the order of 1 pb. We consider the most probable origin of the observed SF events to be the 0.9 ms spontaneously fissioning isomer ^{240mf}Am, with possible smaller contributions from 14 ms ^{242mf}Am and 1.0 ms ^{244mf}Am, which are produced by transfer reactions with cross sections of the order of 10 to 100 nb (see Ref. [21]). Such transfer products are expected to be suppressed by the gas-filled separator by a factor of $\sim 10^4 - 10^5$ [18].

From the absence of α-SF correlations that we can attribute to the production of ^{268}108, we calculated that the 68% confidence level upper limit for its production cross section is 1.3 to 1.9 pb, assuming for the unknown ^{264}106 b_{sf} values between 0 and 100%, respectively. The non-observation of ^{268}108 is consistent with measured cross-section ratios of $\sim 0.2 - 0.3$ for the 4n to 5n evaporation channels in the fusion-evaporation reactions of ^{18}O, ^{22}Ne, and heavier projectiles with actinide targets (see, e.g., Refs. [1,18,22]). Finally, the nonobservation of any SF events following the three detected α decays of ^{267}108 allows us to calculate straighforwardly an upper limit for the SF branch of ^{263}106. Taking into account the 100% probability of detecting SF events, we obtain $b_{sf} <$ 30% at the 68% confidence level, in contrast to $b_{sf} \approx$ 70% suggested for ^{263}106 in Ref. [23] on the basis of indirect arguments.

An important result of the present work is the measurement of the 2.5 pb cross section for the ^{238}U(^{34}S,5n) reaction, which is 10^5 times lower than that of the reaction ^{238}U(^{22}Ne,5n) [22]. As will be discussed in a future publication, this dramatic cross section decrease reveals a fusion limitation mechanism different from that associated with the overcritical Coulomb-to-nuclear force ratio in the entrance reaction channel.

To conclude, we have produced and positively identified the new $N = 159$ isotope of element 108. The decay properties that we established for ^{267}108 offer further evidence for significantly increased nuclear stability near the predicted deformed shells $N = 162$ and $Z = 108$.

We wish to thank Ts. Vylov and V.G. Kadyshevsky for the effective support of the present work. We thank the U400 cyclotron staff for providing the intense ^{34}S beams. Our thanks are due to Yu. D. Shipilov and coworkers for producing the highly enriched ^{34}S material, as well as to V. Ya. Lebedev and A.B. Yakushev for preparing the ^{34}SO$_2$ used in the U400 ion sources. We are grateful to P. Burger and M. Keters for the fruitful cooperation. We thank V.I. Chepigin, V.I. Krashonkin, V.I. Tomin, V.E. Zhuchko, and A.M. Zubareva for their essential help. The Livermore collaborators thank their Russian hosts for their hospitality during the experiment. Much of the support for the LLNL authors was provided through the U.S. DOE under Contract No. W-7405-Eng-48. Some of the Dubna authors acknowledge the individual financial support by Grant No. RFN000 from the International Science Foundation. These studies were performed in the framework of the Russian Federation/U.S. Joint Coordinating Committee for Research on Fundamental Properties of Matter.

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