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NEW NUCLIDE ²⁶⁷108 PRODUCED BY THE ²³⁸U+³⁴S REACTION

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*University of California, Lawrence Livermore National Laboratory, Livermore, California 94551, USA The ground-state decay properties of two recently discovered Z=106 nuclides, ²⁶⁵106 (T_{1/2}≈2-30 s) and ²⁶⁶106 (T_{1/2}≈10-30 s, $b_{sf} \leq 50\%$) [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 ²⁶²102 (T_{1/2}≈T_{sf}=5 ms [2]) to ²⁶⁶106, an increase of four protons, increases the stability against spontaneous fission (SF) by a factor of $\geq 4 \times 10^3$. Similarly, in going from ²⁶⁰106 (T_{1/2}=3.6 ms, $b_{sf}=b_{\alpha}=50\%$ [3,4]) to ²⁶⁶106, the stability increases by factors of $\geq 3 \times 10^3$ for SF decay and $\sim 3 \times 10^3$ for α decay. Thus, the ground-state decay properties of ²⁶⁶106 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]). The discovery of significantly increased nuclear stability near N=162 and Z=108 creates new opportunities for extending the nuclear domain at its upper edge.

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

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isotope ²⁶⁴108 with $T_{1/2}\approx 0.08$ ms, identified by one correlated decay sequence [12], and the odd-A α emitter ²⁶⁵108 with $T_{1/2}=1.8$ ms, for which three correlated α -decay sequences were observed [12].

According to the predictions made in Refs. [7–9], the even-even nuclide ²⁶⁸108 is expected to be an α emitter with $T_{1/2}\sim0.1$ s. Its partial SF half-life was predicted to be of the order of 10^2 s [9]. Thus, a signature for ²⁶⁸108 would be the observation of α decay with the α -particle energy $E_{\alpha}\sim9.7$ MeV [7] followed by SF decay of the unknown daughter ²⁶⁴106, or eventually by SF decay of the short-lived grand-daughter ²⁶⁰104, if ²⁶⁴106 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 ²⁶⁷108 to be the observation of the α decay with $E_{\alpha}\sim10$ MeV, followed by α decays of the known nuclides ²⁶³106 ($T_{1/2}=0.9$ s, $E_{\alpha}=8.95$ to 9.10, 9.25 MeV [14,15]), ²⁵⁹104 ($T_{1/2}=3.1$ s, $b_{\alpha}=93\%$, $E_{\alpha}=8.77$, 8.87 MeV, $b_{sf}=7\%$ [16]), and ²⁵⁵102 ($T_{1/2}=3.1$ min, $b_{\alpha}=61.4\%$, $E_{\alpha}=7.62$ to 8.31 MeV, $b_{EC}=38.6\%$ [16]).

In the present work we used a ³⁴S bombarding energy of 186 MeV to produce Z=108 compound nuclei with an excitation energy of ≈ 50 MeV, sufficient for the evaporation of 4 or 5 neutrons. The laboratory fusion barrier energy for the ²³⁸U+³⁴S system is 181 MeV as calculated by the Bass prescription [17]. Beams of ³⁴S projectiles were delivered by the U400 cyclotron of the Joint Institute for Nuclear Research. The SO₂ gas, with sulfur enriched in ³⁴S to $\approx 80\%$, was used to feed the discharge chamber of a PIG ion source with an indirectly heated cathode, which made it possible to obtain intense ³⁴S beams at a reasonably low consumption of the ³⁴S (≈ 30 mg h⁻¹ p μ A⁻¹). The time structure of the pulsed ³⁴S 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 0.70mg cm⁻² Ti substrates with average areal densities of 0.54 mg cm⁻² of ²³⁸U (99.9%) and a total area of 36 cm² 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 ³⁴S, the targets received a total beam dose of 1.7×10^{19} particles. For calibration purposes, we performed bombardments of ^{nat}W and ²⁰⁷Pb with ³⁴S, as well as bombardments of ²⁰⁶Pb with ⁴⁸Ca and ^{nat}Pt with ²⁶Mg.

Evaporation residues (EVRs) recoiling out of the ²³⁸U targets were separated in flight from beam particles and various transferreaction 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 magnetic rigidity of the separator's dipole magnet, $B\rho$, was adjusted to center the quasi-Gaussian distribution of EVRs on the focal-plane detector in the horizontal direction. To set the $B\rho$ value for the Z=108 EVRs with the expected average velocity $\langle v/v_0 \rangle \approx 1.7$ ($v_0 = 2.2 \times 10^6$ m s⁻¹ is the Bohr velocity), we used prior measurements [1,18] of the average charge states $\langle q \rangle$ for the EVRs 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 EVRs 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 $120 \times 40 \text{ mm}^2$ position-sensitive detector (PSD) array.

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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.

The PSD array consisted of three $40 \times 40 \text{ mm}^2$ passivated boron implanted silicon detectors produced by *Canberra Semiconductor NV*, with each detector having four 40-mm high \times 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 signals from each strip on the resistive detector back were split into three channels to provide y-position signals for α /implant events and SF events, as well as an energy sum signal for α /implant events. We determined total fission energies by summing the y-position signals. With each detected energy event, we also recorded the strip number, TOF information, the time in μ s 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 in a PC AT/386 and periodically transferred to a SUN SPARCstation IPX for storage and analysis.

Alpha-energy calibrations were periodically performed for each strip using the α peaks from nuclides produced in the ^{nat}W+³⁴S reaction. Most of the strips had α -energy peak FWHMs 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 ²⁵²102 [16] and 21-ms ²³⁸Cf [19] produced in the ²⁰⁶Pb+⁴⁸Ca and ²⁰⁷Pb+³⁴S reactions, respectively. These reactions were also used to estimate the collection efficiency of the Z=108 EVRs 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. A comparison of the EVR- α data for $^{217}\mathrm{Th}$ produced in the $^{nat}W+^{34}S$ and $^{nat}Pt+^{26}Mg$ reactions shows an increase in Δpos from 1.2 to 3.5 mm with a decrease in the average measured EVR energy from ≈ 8 to ≈ 3 MeV. For the $^{238}U+^{34}S$ reaction, the Gaussian-like distribution of measured Z=108 EVR energies is expected to have an average of 6 MeV and a FWHM of 3 MeV.

In 'the off-line analyses we searched for event sequences in which EVRs within the expected energy range 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 4 events per day 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 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 $|\Delta pos|=0.4$ 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.

Table I. The measured parameters of the correlated ${}^{267}108$ event sequences observed in the ${}^{238}\text{U}+{}^{34}\text{S}$ 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 | $\Delta \mathrm{pos}^{\mathrm{c}}$ mm |
|--------------------|---------------------------|--------------|-------------------|---|
| EVR^{a} | 4.1 | 11 | | |
| lpha | 9.74 | 11 | 29 ms | -1.0 |
| α | 7:75 | 11 | $9.1 \min$ | -2.1 |
| EVR | 6.2 | 6 | | |
| α | 9.86 | 6 | $32 \mathrm{ms}$ | -0.6 |
| $lpha^b$ | 8.80 | 6 | 3.6 s | -0.6 |
| EVR | 4.4 | 9 | | |
| $lpha^b$ | 9.87 | 9 | $20 \mathrm{~ms}$ | +0.2 |
| α | 8.80 | 9 | 2.0 s | +0.4 |

^aThe quoted EVR energies are measured values. No estimate was included for the pulse-height defect.

^bEvent occurred during the beam pulse.

^cThe y-position deviations are given with respect to the EVR implantation site.

We interpret the three correlated event sequences with $E_{\alpha 1}=9.74$, 9.86, and 9.87 MeV as the α decay of the new isotope ²⁶⁷108 followed by α decay of one of the descendant nuclides, ²⁵⁵102 in the first sequence, and ²⁵⁹104 in the two other sequences. We also observed two out-of-beam α - α correlations with N_b<0.03 linking α decays of ²⁶³106, ²⁵⁹104, and ²⁵⁵102. It is possible that these correlations orig-

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

inate from the decays of $^{267}108$ where the $^{267}108 \alpha$ 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 the ²⁶⁷108 EVRs. 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



Fig. 3. The distribution across detector strips for all outof-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.

intervals between the correlated α decays, agree with the α -particle energy and half-life values for ²⁵⁵102 and ²⁵⁹104 [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 EVRs on the basis of calibration measurements. For all three correlations, the detection of the EVRs by the PSD array was accompanied by TOF signals consistent with expectations for a compound nucleus. From our data, we estimated the average charge state <q> of the Z=108 EVRs 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 μ s 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 ²⁶⁸108, 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 ²⁶⁴106 b_{sf} values between 0 and 100%, respectively. The non-observation of ²⁶⁸108 can be readily explained, since for fusion-evaporation reactions induced by ¹⁸O, ²²Ne, and heavier projectiles on actinide targets, cross sections for the 4n-evaporation channel are 3 to 5 times smaller than for the 5n channel (see, e.g., Refs. [1,18,22]). Finally, the non-observation of any SF events following the three detected α decays of ²⁶⁷108 allows us to calculate straightforwardly an upper limit for the SF branch of ²⁶³106. 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 ²⁶³106 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.

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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.

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²⁶⁷108 determined at GSI ($E_{\alpha}=9.88$ MeV, $T_{1/2}=33^{+19}_{-9}$ ms) fully corroborate the results of the present work.

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