Search for the production of element 112 in the ^{48}Ca+ ^{238}U reaction

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We have searched for the production of element 112 in the reaction of 231 MeV ^{48}Ca with ^{238}U. We have not observed any events with a ''one-event'' upper limit cross section of 1.6 pb for evaporation residue- (EVR-) fission events and 1.8 pb for EVR-alpha events.

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I. INTRODUCTION

The heaviest elements are a laboratory to study nuclear structure and nuclear dynamics under the influence of large Coulomb forces. The results of heavy element research deal with fundamental issues in both chemistry and physics. During the past six years, there have been spectacular advances in this field, i.e., the discovery of elements 110–112, the synthesis of elements 114 [1,2] and element 116 [3] by ''hot fusion'' reactions, the first chemical studies of elements 104–108, and the spectroscopy of the transfermium nuclei.

As an aside, we note the two different traditional paths to the heavy elements: (a) ''cold fusion,'' involving the reaction of massive projectiles with Pb and Bi target nuclei, leading to low excitation energies in the completely fused species (with resulting high survival probabilities) and reduced fusion cross sections, and (b) ''hot fusion,'' the reaction of lighter projectiles with actinide target nuclei, leading to larger fusion cross sections but reduced survival probabilities (due to the higher excitation energies of the completely fused species.) At present, it appears that hot fusion reactions are the preferred path to synthesize new heavy elements (Fig. 1) although the large cross sections associated with the production of elements 112–116 are poorly understood [4]. In any case, it is imperative to confirm these reported hot fusion cross sections in laboratories not connected to the original work.

In 1999, a Dubna-GSI-RIKEN collaboration [5] reported the successful synthesis of ^{283}112 using the reaction 231 MeV ^{48}Ca+ ^{238}U$\rightarrow$ ^{286}112$\rightarrow$ ^{283}112+3n with the observation of two events. The nuclide ^{283}112 ($t_{1/2}=81^{+147}_{-32}$ s) was reported to decay by spontaneous fission (SF) and was produced with a cross section of $5.0^{+6.3}_{-3.2}$ pb. The decay mode of ^{283}112 is somewhat unexpected as all the other isotopes of element 112 ($A=277$, 284, and 285) decay by alpha emission. The Dubna-GSI-RIKEN Collaboration searched for alpha decay in ^{283}112 but could not see any events. Subsequently, in the reaction of ^{48}Ca with ^{242}Pu, two events were found in which an evaporation residue (EVR) emitted an alpha particle, producing a daughter nucleus that decayed by SF [6]. These latter SF decays were attributed to the decay of ^{283}112 and, if taken with the previous work, imply a half-life of $\sim$3 min for ^{283}112.

In Fig. 2(a), we show the predicted [7–10] and observed Q_α values for the well-characterized alpha decay of ^{277}112 and its daughters (^{273}110, ^{269}Hs, ^{265}Sg, ^{261}Rf, and ^{257}No). The semiempirical predictions of Liran *et al.* [8] apparently do not include the nuclear structure effects near the $N=162$ subshell. The theoretical predictions of Smolańczuk [9] seem to do the best job of predicting the observed values of Q_α ($\chi^2_{Möller}=960$, $\chi^2_{Liran}=400$, $\chi^2_{Smolańczuk}=160$,$\chi^2_{Royer}=400$). In Fig. 2(b), we show a similar plot of the predicted and observed values of Q_α for the α decay of various isotopes of element 112. The predictions of Liran *et al.* deviate significantly from the observed values with the predictions of Royer and Möller *et al.* being similar. The theoretical predictions of Möller *et al.* and Smolańczuk are approximately equal in their ability to predict Q_α with a slight preference being given to the predictions of Möller *et al.* ($\chi^2_{Möller}=240$, $\chi^2_{Liran}=1080$, $\chi^2_{Smolańczuk}=400$, $\chi^2_{Royer}=240$) Using these comparisons of predicted and observed values of Q_α as a guide, we favor the predictions of Smolańczuk as being the most reliable guide to the expected decay properties of element 112. However, some caution must be exercised as none of the predictions provide a statistically significant fit to the data. In the only calculation [9] to address the spontaneous fission and alpha decay of the isotopes of 112, alpha decay is predicted to be the dominant mode of decay for all isotopes although the differences in predicted half-lives are only an order of magnitude for the nuclei of interest.

We show in Fig. 3, the expected alpha-decay sequence for ^{283}112 based upon the predictions of Smolańczuk for the

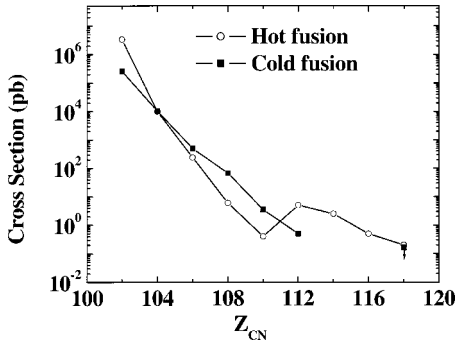

FIG. 1. The predicted and observed cross sections for the synthesis of heavy nuclei using ''hot'' and ''cold'' fusion reactions. The value shown for element 118 is an upper limit.

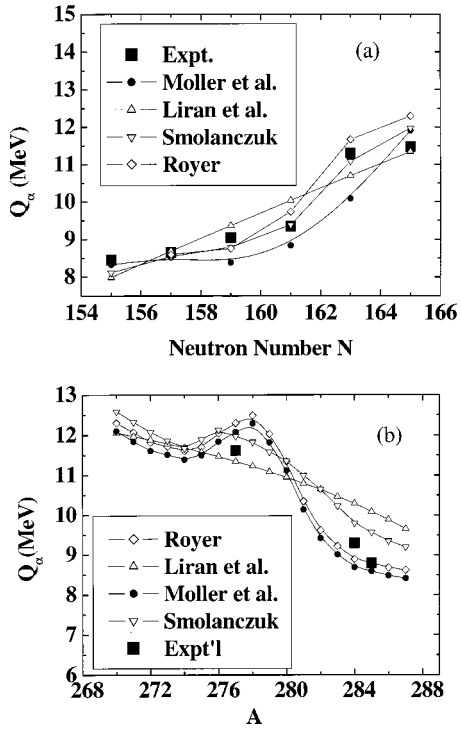

FIG. 2. (a) Predicted and measured Q_α values for the decay of $^{277}112$. (b) Predicted and measured Q_α values for the decay of various isotopes of element 112.

masses of the heaviest elements and the Hatsukawa-Nakahara-Hoffman rules for the alpha-decay lifetimes of the heavy nuclei [11]. As indicated earlier, in searching for these predicted alpha-decay sequences, one must be sensitive over a wide range of nuclear lifetimes.

The nucleus $^{283}112$ and its synthesis play an important role in our understanding of the recent syntheses of elements 114 and 116 by hot fusion reactions [1–3]. $^{283}112$ is directly populated in the deexcitation of $^{287}114$ synthesized using the $^{48}Ca+^{242}Pu$ reaction [6]. The long half-life is typical of elements 112 and 114 nuclei produced in the synthesis of elements 114 and 116 [1–3]. The relatively large reported production cross section, 5 pb, is typical of the higher cross sections associated with hot fusion reactions compared to cold fusion reactions (Fig. 1) for the synthesis of $Z>112$. It is these same cross sections which challenge our understanding because current theoretical predictions of the survival probabilities in these reactions [12] would not give cross sections of this magnitude. For example, Armbruster [13], using the best available data on the capture cross sections, the probability of evolving from the contact configuration to the completely fused system, and the survival probabilities, estimated an evaporation residue production cross section for the reaction of 231 MeV $^{48}Ca+^{238}U\rightarrow^{286}112\rightarrow^{283}112+3n$ of 50 fb.

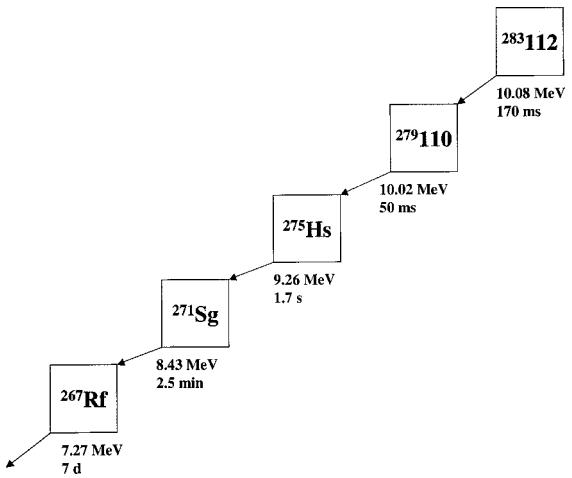

FIG. 3. Predicted alpha-decay sequences for the decay of $^{283}112$.

II. EXPERIMENT

The reaction $^{238}U(^{48}Ca,3n)$ was studied at the 88-Inch Cyclotron of the Lawrence Berkeley National Laboratory, using the Berkeley Gas-filled Separator (BGS) [14]. The experimental apparatus was a modified, improved version of the apparatus used in [14], including improved detectors and data acquisition system, continuous monitoring of the separator gas purity, and better monitoring of the ^{48}Ca beam intensity and energy. A $^{48}Ca^{10+}$ beam was accelerated to 243.5 MeV with an average current of $\sim 3\times10^{12}$ ions/s (480 particle nA). The beam went through the 45-μg/cm^2 carbon entrance window of the separator before passing through the ^{238}U target placed 0.5 cm downstream from the window. The targets were UF_4 deposits (U thickness=463 μg/cm^2) with an 0.54 mg/cm^2 Al backing on the upstream side. Nine of the arc-shaped targets were mounted on a 35-cm wheel that was rotated at 300 rpm. The beam energy in the target was 228–234 MeV [15], encompassing the projectile energy range used in [5]. The beam intensity was monitored by two silicon p-i-n detectors (mounted at $\pm27°$ with respect to the incident beam), which detected elastically scattered beam particles from the target. Attenuating screens were installed in front of these detectors to reduce the number of particles reaching them (and any subsequent radiation damage to the detector). The run lasted approximately 5.5 days.

The EVRs ($E\sim39$ MeV) were separated spatially in flight from beam particles and transfer reaction products by their differing magnetic rigidities in the gas-filled separator. The separator was filled with helium gas at a pressure of 96 Pa. The expected magnetic rigidities of 39-MeV $^{283}112$ EVRs were estimated using the data of Ghiorso *et al.* [16] This estimate was 2.25 Tm from extrapolation of the data in their Fig. 3. The optimum $B\rho$ values determined experimentally with the BGS for the EVRs from the reaction of 202 MeV ^{48}Ca with ^{176}Yb, 215 MeV ^{48} Ca with ^{208}Pb and 309 MeV ^{64}Ni with ^{208}Pb corresponded to the ''graphical value'' of $B\rho$, and thus we chose a $B\rho$ of 2.25 Tm for the separator magnetic field.

To determine the transport efficiency of the BGS, we used a combination of measurements and Monte Carlo simulations. We measured the transport efficiency of the separator, the efficiency of transporting an EVR produced in the target

and implanting it in the focal plane detector, to be 57% for the reaction of 202 MeV ^{48}Ca with ^{176}Yb, assuming a cross section for this reaction of $\sim$790 μb. (This latter value was extrapolated from the measured data of Sahm *et al.* [17].) A Monte Carlo simulation of the separator efficiency for this reaction [18] predicted an efficiency of 53%. We measured a transport efficiency of 45% for the reaction of 215 MeV ^{48}Ca+ ^{208}Pb$\rightarrow$ ^{254}No+2n. [This efficiency is based on a cross section for the ^{208}Pb(^{48}Ca,2n) reaction of 3.0 μb [19].) The Monte Carlo simulation program predicted 51%. Having thus ''validated'' the Monte Carlo simulation code, we used it to estimate a transport efficiency for the ^{283}112 EVRs of 49% for the reaction of 231 MeV ^{48}Ca with ^{238}U under the conditions described above. This value is similar to efficiencies reported for similar reactions using the Dubna gas-filled separator [20].

As a further demonstration of our ability to measure events similar to those being sought in the ^{48}Ca+ ^{238}U experiment, we measured the cross section for the 215.5 MeV ^{48}Ca+ ^{206}Pb$\rightarrow$ ^{252}No+2n reaction by detecting the SF decay (SF branching ratio 0.269) of ^{252}No. We measured a cross section of 585$\pm$90 μb for this reaction in agreement with the known value of 500 μb [21].

In the focal plane region of the separator, the EVRs passed through a 10 cm$\times$10 cm parallel-plate avalanche counter (PPAC) [22] that registered the time, ΔE, and x,y position of the particles. This PPAC has an approximate thickness equivalent to 0.6 mg/cm^2 of carbon. The PPAC was $\sim$29 cm from the focal plane detector. The time of flight of the EVRs between the PPAC and the focal plane detector was measured. The PPAC was used to distinguish between beam-related particles hitting the focal plane detector and events due to the decay of previously implanted atoms. During these experiments, the PPAC efficiency for detecting beam-related particles depositing between 8 and 14 MeV in the focal plane detector was 97.5%–99.5%.

After passing through the PPAC, the recoils were implanted in a 32-strip, 300-μm-thick passivated ion-implanted silicon detector at the focal plane that had an active area of 116 mm$\times$58 mm. The strips were position sensitive in the vertical (58 mm) direction. The energy resolution of the focal plane detector was measured to be $\sim$70 keV [full width at half maximum (FWHM). The differences in measured positions for the ^{252}No-^{248}Fm full energy α-α correlations in a study of the 215.5 MeV ^{48}Ca+ ^{206}Pb reaction had a Gaussian distribution with a FWHM of 0.52 mm (σ=0.22 mm). The measured position resolution for full energy alpha particles correlated to ''escape'' alpha particles (which deposited only 0.5–3.0 MeV in the detector) was $\sim$1.2 mm (FWHM). A second silicon strip ''punchthrough'' detector was installed behind this detector to reject particles passing through the primary detector. A ''top'' and a ''bottom'' detector were installed in front of the focal plane detector to detect escaping alpha particles and fission fragments. The focal plane detector combined with these ''top'' and ''bottom'' detectors had an estimated efficiency of 75% for the detection of full energy 10 MeV α particles following implantation of a ^{283}112 nucleus.

Any event with $E>$0.5 MeV in the focal plane Si-strip

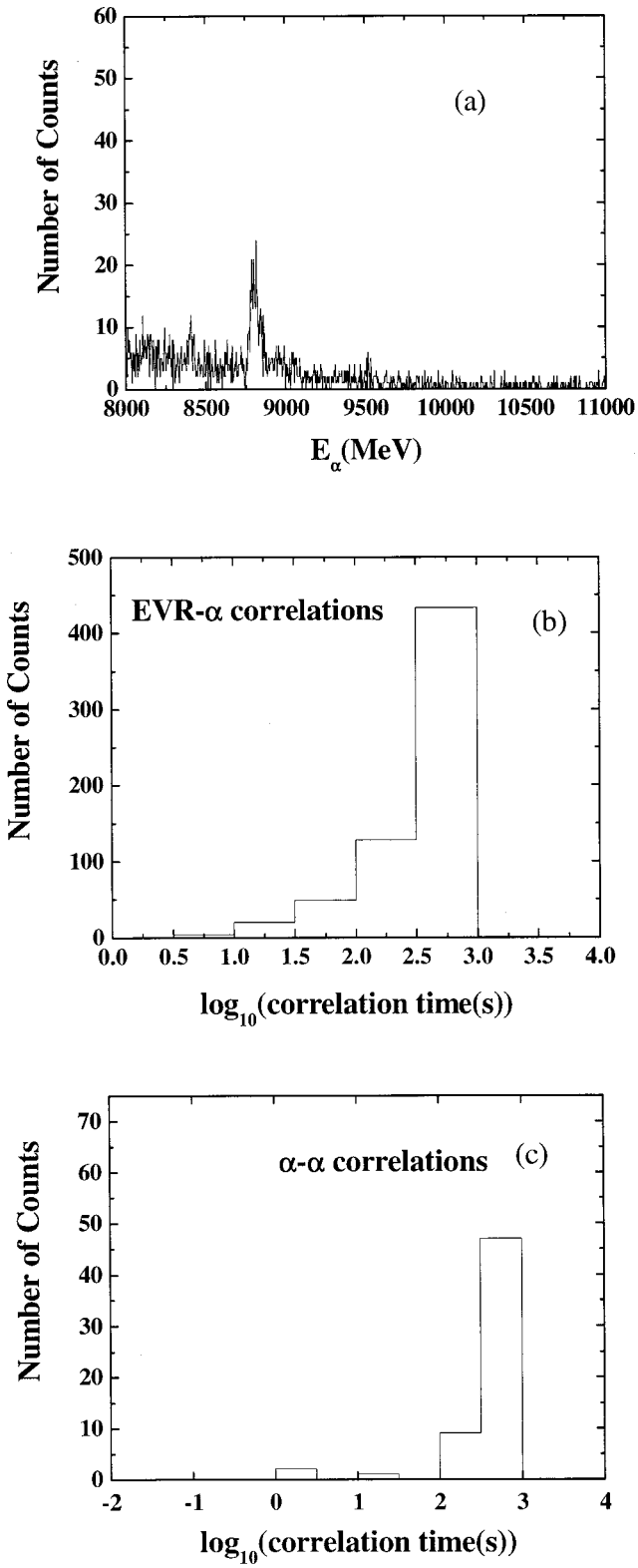

FIG. 4. Representative spectra of (a) the implanted α-particle decays detected in the focal plane detector in the energy range 8 $\leqslant E_\alpha \leqslant$11 MeV, (b) the EVR-$\alpha$ correlation time distribution, and (c) the α-α correlation time distribution for the reaction of 231 MeV ^{48}Ca with ^{238}U. These data were collected using a maximum correlation time of 1000 s.

detector triggered the data acquisition. Data were recorded in list mode and included the time of the trigger, the position and energy signals from the PPAC and the Si-strip detectors, and energy signals from the ''top,'' ''bottom'' and ''punch-through'' detectors. With the use of buffering analog-to-digital converters (ADCs) and scalers, the minimum time between successive events was 15 μs.

In a study of the 215 MeV ^{48}Ca+ ^{206}Pb reaction, the pulse height defect for the $\sim$17 MeV ^{252}No recoils was determined to be $\sim$10 MeV. This correction was used to determine the expected range of energies associated with the $\sim$15 MeV ^{283}112 recoils as they struck the focal plane detector.

With a beam current of 3×10^{12} ^{48}Ca ions striking the target, the average total counting rates ($E>0.5$ MeV) in the focal plane detector were $\sim$0.84/s. The average rate of ''alpha particles'' (7–14 MeV with no PPAC signal) was <1.7/m. No SF events were observed. In Fig. 4(a), we show the singles spectrum (in anticoincidence with the PPAC) measured with the focal plane detector during a single run in which a dose of 3.9×10^{17} ions was delivered to the target. The peak in the spectrum at 8.78 MeV is due to the decay of ^{212}Po^{m} which in turn is the result of the decay of transfer products from the ^{228}Th decay series.

III. RESULTS AND DISCUSSION

Two search strategies were used to look for events corresponding to the implantation and decay of ^{283}112 nuclei. The first strategy assumed the decay of ^{283}112 would occur as outlined in Fig. 3, in accordance with the predictions of Smolańczuk. We searched for EVR-α, α-α, and EVR-fission events occurring within 6 s, restricting the range of α-particle energies to be from 8 to 11 MeV and the single-fragment fission energies to be $\geqslant$90 MeV. (This latter limit was chosen to include 96% of the expected single-fragment kinetic energy distribution assuming the SF single fragment kinetic energy distributions have similar shapes for ^{252}No and ^{283}112). No events were observed with a total dose of 1.1×10^{18} ions. This corresponds to a one-event upper limit cross section of 1.8 pb for ^{283}112 nuclei decaying by alpha-particle emission and 1.6 pb for spontaneously fissioning ^{283}112 nuclei when one takes into account the differing efficiencies of detecting fission fragments and alpha-particle decay chains. (A one-event upper limit cross section is the cross section that would result if we observed one event in the experiment).

A second strategy involved searching for events similar to those reported by the Dubna-GSI-RIKEN group. [5]. We searched for EVR-α, α-α, and EVR-fission events occurring within 1000 s, using the same energy restrictions as in the first search. No EVR-fission events were found, leading to a one-event upper limit cross section of 1.6 pb for the type of event reported by the Dubna-GSI-RIKEN group or any chain terminating in an SF decay. As a result of a significant number of accidental EVR-α and α-α events, no meaningful upper limit could be set for EVR-α events with these longer correlation times, as in the Dubna experiment. [In Figs. 4(b) and 4(c), we show the EVR-α and α-α time correlation distributions for the search window of $\Delta t=1000$ s for the run associated with Fig. 4(a). The correlation distributions indicate accidental correlations as do the observed decay sequences.]

The one event upper limit cross section for the production of spontaneously fissioning ^{283}112 nuclei of 1.6 pb is just below that reported by the Dubna-GSI-RIKEN group of $5.0^{+6.3}_{-3.2}$ pb. Another relevant observation is that of Yakushev *et al.* [23], who reported the failure to observe any spontaneously fissioning ^{283}112 nuclei in the reaction of 234 MeV ^{48}Ca with ^{238}U using the assumption that element 112 behaves like Hg, a volatile liquid, in its chemistry. If element 112 behaves chemically like Hg, then this observation would suggest an upper limit cross section of 1.5 pb for this reaction. An alternative explanation [23,24] is that element 112 behaves chemically like a noble gas (Rn). Recent theoretical predictions [25] using the dinuclear system approach have suggested a cross section for the ^{238}U(^{48}Ca,3n) ^{283}112 reaction of 1.7 pb.

Further work is needed to establish the cross section for the production of ^{283}112 in the ^{238}U(^{48}Ca,3n) reaction. Because the reported spontaneous fission decay is not definitive to determine the Z and A of this nucleus, it seems especially important to detect the α-decay branch for this nuclide. The apparently small cross sections and/or weaker α-decay branching ratios make this worthwhile effort difficult. If, as indicated in this work, the production cross section for ^{283}112 in the ^{238}U(^{48}Ca,3n) reaction is $\sim$2 pb or less, then it becomes more difficult to understand the reported cross sections of $\sim$1 pb for the production of elements 114 and 116 in similar reactions.

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[1] Y.T. Oganessian *et al.*, Phys. Rev. Lett. **83**, 3154 (1999).
[2] Y.T. Oganessian *et al.*, Phys. Rev. C **63**, 011301(R) (2001).
[3] Y.T. Oganessian *et al.*, Phys. Rev. C **62**, 041604(R) (2000).
[4] V.I. Zagrebaev, Y. Aritomo, M.G. Itkis, and Yu.Ts. Oganessian, Phys. Rev. C **65**, 014607 (2001).
[5] Y.T. Oganessian *et al.*, Eur. Phys. J. A **5**, 63 (1999).
[6] Y.T. Oganessian *et al.*, Nature (London) **400**, 242 (1999).
[7] G. Royer, J. Phys. G **26**, 1149 (2000).

[8] S. Liran, A. Marinov, and N. Zeldes, Phys. Rev. C **62**, 047301 (2000); **63**, 017302 (2001).
[9] R. Smolańczuk, Phys. Rev. C **56**, 812 (1997).
[10] P. Möller *et al.*, At. Data Nucl. Data Tables **59**, 185 (1995).
[11] Y. Hatsukawa, H. Nakahara, and D.C. Hoffman, Phys. Rev. C **42**, 674 (1990).
[12] G.G. Adamian, N.V. Antonenko, and S.P. Ivanova, Phys. Rev. C **62**, 064303 (2000).
[13] P. Armbruster, Rep. Prog. Phys. **62**, 465 (1999).
[14] V. Ninov and K.E. Gregorich, in *ENAM 98, Exotic Nuclei and Atomic Masses*, edited by B. M. Sherrill, D. J. Morrissey, and C. N. Davids, AIP Conf. Proc. No. 455 (AIP, Woodbury, 1998), p. 704.
[15] F. Hubert, R. Bimbot, and H. Gauvin, At. Data Nucl. Data Tables **46**, 1 (1990).
[16] A. Ghiorso, S. Yashita, M.E. Leino, L. Frank, J. Kalnins, P. Armbruster, J.-P. Dufour, and P.K. Lemmertz, Nucl. Instrum. Methods Phys. Res. A **269**, 192 (1988).
[17] C.C. Sahm *et al.*, Nucl. Phys. **A441**, 316 (1985).
[18] K. E. Gregorich *et al.*, Eur. Phys. J. A (submitted).
[19] H.W. Gäggeler *et al.*, Nucl. Phys. **A502**, 561c (1989).
[20] K. Subotic, Y.T. Oganessian, V.K. Utyonkov, Y.V. Lobanov, F.S. Abdullin, A.N. Polyakov, Y.S. Tsyganov, and O.V. Ivanov, Nucl. Instrum. Methods Phys. Res. A **481**, 71 (2002).
[21] Y.T. Oganessian *et al.* Phys. Rev. C **64**, 054606 (2001).
[22] D. Swan, J. Yurkon, and D.J. Morrissey, Nucl. Instrum. Methods Phys. Res. A **348**, 314 (1994).
[23] A.B. Yakushev *et al.*, Radiochim. Acta **89**, 743 (2001).
[24] K.S. Pitzer, J. Chem. Phys. **63**, 1032 (1975).
[25] N.V. Antonenko, G.G. Adamian, A. Diaz-Torres, and W. Scheid, in *Proceedings of the International Nuclear Physics Conference, INPC 2001*, edited by E. Norman *et al.*, AIP Conf. Proc. No. 610 (AIP, Woodbury, 2002), pp. 633–637.