Measurement of the ^{208}Pb(^{52}Cr, n)^{259}Sg excitation function

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The excitation function for the ^{208}Pb(^{52}Cr, n)^{259}Sg reaction has been measured using the Berkeley Gas-filled Separator at the Lawrence Berkeley National Laboratory 88-Inch Cyclotron. The maximum cross section of 320^{+110}_{-100} pb is observed at a center-of-target laboratory-frame energy of 253.0 MeV. In total, 25 decay chains originating from ^{259}Sg were observed and the measured decay properties are in good agreement with previous reports. In addition, a partial excitation function for the ^{208}Pb(^{52}Cr, $2n$)^{258}Sg reaction was obtained, and an improved ^{258}Sg half-life of $2.6^{+0.6}_{-0.4}$ ms was calculated by combining all available experimental data.

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Introduction. "Cold" nuclear fusion reactions, using Pb or Bi targets with projectiles ranging from Ca to Zn, have been successfully employed in the production of elements 102–113 (see [1,2] for reviews and [3–5] for more information). Most of these experiments used the most neutron-rich stable projectile available, but the lack of information on production cross section systematics for reactions using projectiles with fewer neutrons has stimulated recent research to study this effect. For example, the compound nucleus (CN) in the ^{52}Cr + ^{208}Pb reaction is more neutron-deficient than in the ^{54}Cr + ^{208}Pb reaction, so the evaporation residue (EVR) decay energies provide a probe of the ground state mass surface (and hence the shell correction) in this region. Additionally, the measured EVR cross sections provide theorists with information for evaluating systematic trends in the projectile capture cross section σ_{cap}, the probability of CN formation P_{CN}, and the ratio of neutron-emission to fission Γ_n/Γ_f, which are vital for the planning of future experiments.

For these reasons, we conducted an experiment to study the effect of using a less neutron-rich projectile using the ^{208}Pb(^{52}Cr, n)^{259}Sg reaction. The analogous reaction ^{208}Pb(^{54}Cr, n)^{261}Sg has been studied by Antalic *et al.* [6], so a comparison can be made. We also measured a partial excitation function for the ^{208}Pb(^{52}Cr, $2n$)^{258}Sg reaction and compared it to the ^{209}Bi(^{51}V, $2n$)^{258}Sg reaction [7,8], which has the same compound nucleus.

Experiments. Experiments were conducted at the Lawrence Berkeley National Laboratory (LBNL) 88-Inch Cyclotron using the Berkeley Gas-filled Separator (BGS), and the setup was identical to that described in [9] except for the beam shutoff parameters described below. The ^{52}Cr^{12+} beam (with average intensity 120 particle nA and energy error ≈1% [10]) passed from the evacuated beamline to the 0.5-torr He chamber of the BGS by passing through a 45-μg/cm^2 $^{\text{nat}}$C entrance window. Approximately 0.5 cm downstream the beam passed through a 35-μg/cm^2 $^{\text{nat}}$C target backing which supported (470 ± 60)-μg/cm^2-thick Pb targets (isotopic composition 98.4% ^{208}Pb, 1.1% ^{207}Pb, 0.5% ^{206}Pb). A 5-μg/cm^2 $^{\text{nat}}$C layer covered the target material to prevent sputtering and enhance cooling. The nine arc-shaped targets were mounted on the periphery of a 14-inch-diameter wheel that rotated at ≈7 Hz to prevent excessive target heating. Energy losses of the beam in these materials were calculated using the SRIM-2003 program [11] and used to determine the lab-frame center-of-target (cot) energy E_{cot}. Luminosity and primary beam energy were monitored via Rutherford scattering using two *p-i-n* detectors mounted at ≈27° to the beam axis. The experimental conditions are summarized in Table I.

The focal plane detectors were calibrated using external alpha-particle sources of ^{148}Gd, ^{239}Pu, ^{241}Am, and ^{244}Cm. A correction for the energy of the daughter recoils was calculated using the implanted α-decaying products of the reaction of the ^{52}Cr beam with stationary targets of ^{112,114}Sn. The energy resolution (1σ) for α particles fully stopped in the strip detectors was ≈0.023 MeV; it was ≈0.052 MeV for "reconstructed" α particles which escaped from the front of a strip and implanted in an upstream detector so that the energies could be summed. The efficiencies for detecting fully stopped and reconstructed alphas were ≈55% and ≈30%, respectively. In this work uncertainties are quoted at the 1σ(68%) confidence level using methods described in [12] unless specified otherwise.

The average charge state of Sg EVRs was estimated to be approximately +7.6 using Eq. (1) in [10], resulting in an estimated magnetic rigidity of 2.13 T m. The first six decay chains were observed with higher rigidities, so the central rigidity was increased to 2.17 T m for the remainder of the experiments.

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TABLE I. Summary of experimental results. E_{cot} is the beam energy at the center-of-target. Upper limit cross sections are reported at the 84% (1.84-event) confidence limit.

Energy from cyclotron (MeV)	E_{cot} (MeV)	E^*_{cot} (MeV)	Target thickness (μg/cm^2)	Dose (10^{16})	^{259}Sg		^{258}Sg	
					Events	Cross section (pb)	Events	Cross section (pb)
250.7	246.3	13.3	470 ± 60	4.9	1	33^{+75}_{-27}	0	<55
254.0	249.6	15.9	470 ± 60	4.3	6	230^{+140}_{-100}	0	<63
257.4[a]	253.0	18.6	470 ± 60	8.0	16	320^{+110}_{-100}	0	<34
260.0	255.6	20.7	470 ± 60	3.4	0	<88	1	44^{+100}_{-37}
263.1	258.7	23.2	470 ± 60	1.9	2	170^{+220}_{-110}	0	<140
266.2	261.8	25.7	470 ± 60	7.9	0[b]	<38	8	150^{+74}_{-57}

[a]Combines data from runs with beam energies of 257.8 MeV and 256.9 MeV.
[b]A chain beginning with ^{255}Rf was observed at this energy.

An online program continuously searched for evidence of a heavy element decay chain and could interrupt the primary beam within ≈140 μs if an implantation event with energy 7–25 MeV was followed within 10 s by an α particle with energy 8–11 MeV. These events were required to occur in the same strip and to have vertical positions within 3 standard deviations, calculated according to Eqs. (1) and (2) in [9]. A correlation meeting these conditions switched the beam off for 180 s, and the total fraction of time with the beam off was 1.8%.

The assignment of decay chains to ^{259}Sg or ^{258}Sg is straightforward based on the significantly differing decay properties of these two nuclides. ^{259}Sg decays with an α branch of (90 ± 10)%, spontaneous fission (SF) branch <20%, and half-life of $0.48^{+0.28}_{-0.13}$ s [13]. ^{258}Sg decays by SF with a ≈100% branch and half-life of $2.9^{+1.3}_{-0.7}$ ms [14]. The probabilities for detecting α particles (either fully stopped or reconstructed) and SF events are ≈85% and ≈100%, respectively. Based on the known decay properties, the probability of observing a ^{259}Sg or ^{258}Sg decay chain is estimated to be (91 ± 5)% and (100 ± 2)%, respectively. The transmission of the BGS for products of the ^{208}Pb(^{52}Cr, n)^{259}Sg and ^{208}Pb(^{52}Cr, $2n$)^{258}Sg reactions is estimated to be (51 ± 5)% using the Monte Carlo simulation described in Ref. [15] with a small correction for the fact that the vertical distribution of implantation events was off-center by a small amount. Overall cross section systematic error is comparable to that reported in [10] (12%).

Results. A total of 25 decay chains originating from ^{259}Sg was detected and in all cases α decay was observed. The measured half-life (see [16]) is 320^{+80}_{-60} ms, and agrees with the previously reported value of 480^{+280}_{-130} ms [13] within the reported errors. The 14 events with α energies greater than 9.5 MeV form a single group with an energy of 9.593 ± 0.046 MeV, in good agreement with the reported literature value of 9.62 ± 0.03 MeV [13]. The remaining α energies were distributed in the range 9.00–9.47 MeV, except for two escape alphas. A spectrum of all α-like events is available online [17]. The upper limit branching ratio for fission of ^{259}Sg is ⩽8.6% at the 84% confidence level, corresponding to a lower limit of ⩾3.0 s for the partial fission half-life. In order to estimate the electron capture (EC) branch of ^{259}Sg it was necessary to estimate the alpha branch in ^{259}Db, the ^{259}Sg EC daughter. Combining experimental [18] and theoretical [19,20] decay properties of ^{259}Db, we estimate an α branch of ≈96%. This leads to a ^{259}Sg upper limit EC branch of ⩽13%, and a lower limit partial EC half-life of ⩾2.1 s.

Nine events were assigned to ^{258}Sg, and all nine decayed via SF. The measured half-life is $2.1^{+1.0}_{-0.6}$ ms, in good agreement with $2.9^{+1.3}_{-0.7}$ ms reported in [8] and $2.7^{+0.9}_{-0.7}$ ms reported in [7]. Combining data from [7,8] with the current work, the overall half-life of 28 ^{258}Sg fission events is $2.6^{+0.6}_{-0.4}$ ms, in good agreement with all three experiments. Although small α branches in even-even Rf isotopes have recently been observed [8,21], α decay of even-even ^{258}Sg was not observed, corresponding to an upper limit for the α branch of ⩽38%. (An upper limit of ⩽20% was reported in [8].) The corresponding lower limit for the partial α decay half-life is ⩾5.8 ms, compared to ⩾182 ms calculated using Parkhomenko and Sobiczewski α-decay systematics [22] with shell-corrected mass excesses from [23]. Although, our experimental setup was not suitable for measuring the total kinetic energy (TKE) of fission fragments, the data for three pairs of coincident fragments are consistent with known TKE systematics as a function of $Z^2/A^{1/3}$ (see Fig. 9 in [24] and references therein).

The average magnetic rigidity of all ^{258,259}Sg EVRs in He is 2.16 ± 0.03 T m (statistical uncertainty only), corresponding to an average charge state of +7.4. A detailed listing of all decay chains is available online in the EPAPS repository [17].

Decay of ^{255}Rf was observed after all 25 decays of ^{259}Sg. In addition, a decay chain was observed in the E_{cot} = 261.8 MeV run that was consistent with an implantation event followed by the α decay of ^{255}Rf and ^{251}No. Including this EVR-^{255}Rf-^{251}No chain, there were 26 ^{255}Rf decays, 13 by α particle emission and 13 by fission. The combined half-life of all 26 observed decays is $2.3^{+0.8}_{-0.5}$ s, compared to 1.68 ± 0.09 s [25] reported previously. The α and SF branching ratios are (52^{+13}_{-17})% and (48^{+17}_{-13})%, respectively. The SF branching ratio is consistent with two earlier reports: (52 ± 7)% [26] and (45 ± 6)% [8]. The data from six coincident pairs of fission fragments are consistent with known TKE systematics.

The 13 α decays of ^{255}Rf led to ^{251}No, and 11 subsequent α decays are attributed to the decay of the latter isotope. In a twelfth chain the ^{255}Rf α was followed by a reconstructed α with energy 7.509 MeV and lifetime 158.52 s, consistent with EC decay of ^{251}No to ^{251}Md, which then underwent α decay. In the thirteenth chain, no additional decays were observed after α decay of ^{255}Rf. Our experiment did not have sufficient energy resolution to distinguish decay of the ground state ($E_\alpha = 8.612 \pm 0.004$ MeV, $t_{1/2} = 0.80 \pm 0.01$ s) from decay of the first isomeric state ($E_\alpha = 8.668 \pm 0.004$ MeV, $t_{1/2} = 1.02 \pm 0.03$ s) of ^{251}No [25], but the measured half-life of all 11 ^{251}No α decays combined is $0.78^{+0.38}_{-0.22}$ s, in excellent agreement with the ground state half-life.

Two isomers of ^{247}Fm are known: ^{247}Fm^g ($E_\alpha =$ 7.824 MeV, $t_{1/2} = 31 \pm 1$ s) and ^{247}Fm^m ($E_\alpha = 8.172$ MeV, $t_{1/2} = 5.1 \pm 0.2$ s) [24]. Five α decays following the decay of ^{251}No were observed and are attributed to ^{247}Fm^g based on the decay energies. A sixth escape α could not be assigned to a specific isomer but its lifetime was 78.4 s, consistent with ^{247}Fm^g. The combined half-life of all six decays is 57^{+30}_{-17} s, which is in fair agreement with the known ground-state half-life. Heßberger *et al.* [25] observed that ^{251}No^g decays primarily to ^{247}Fm^g and ^{251}No^m decays primarily to ^{247}Fm^m; our observations are consistent with these results.

An analysis of possible random correlations was conducted using methods described in the Appendix of [9]. The average rates of α particle events, both fully stopped and reconstructed, across the entire focal plane with energies from 7–11 MeV was 4.47×10^{-3}s^{-1}. The total number of implantation events with energies from 7–25 MeV was 1.56×10^5. During data analysis, a minimum of two α particles or one valid fission event correlated to an implantation event was considered necessary for establishing a decay chain. The expected number of random EVR-α-α correlations within a 5.0-mm vertical pixel and 180 s of implantation is 0.16, while 12 were observed. The overall rate of fission-like events with energies from 100–300 MeV was 4.8×10^{-5}s^{-1}. Using a similar analysis, the expected number of random EVR-SF correlations in a 5.0-mm pixel and within 20 ms of implantation is 2.7×10^{-4}, while the actual number of observed ^{258}Sg decay chains was nine. The expected number of random EVR-α-SF correlations (corresponding to ^{259}Sg α decay followed by SF of ^{255}Rf) within 10 s is $\approx 10^{-5}$, compared to 13 observed decay chains.

^{208}Pb(^{52}Cr; $n, 2n$)^{259,258}Sg *excitation functions.* The measured ^{208}Pb(^{52}Cr, n)^{259}Sg excitation function is shown in Fig. 1, and is symmetrical in shape, consistent with the majority of other measured cold fusion excitation functions (see Fig. 4 in [27]). The maximum observed cross section is 320^{+110}_{-100} pb at $E_{\text{cot}} = 253.0$ MeV. The upper limit measured at $E_{\text{cot}} = 255.6$ MeV is unexpected since much larger cross sections were observed at energies slightly above and below this energy. The 84% confidence limit for the expected cross section at this energy is $\geqslant$200 pb, so $\geqslant$4.3 decay chains would have been expected given the measured dose. The Poisson probability of observing zero decay chains when $\geqslant$4.3 are expected is $\leqslant$1.6%. Although this probability is small, we believe that the smooth variation in the other observed cross sections and the good agreement of the observed decay properties with previous reports suggests that the remaining data should not be discredited, and form a complete excitation function.

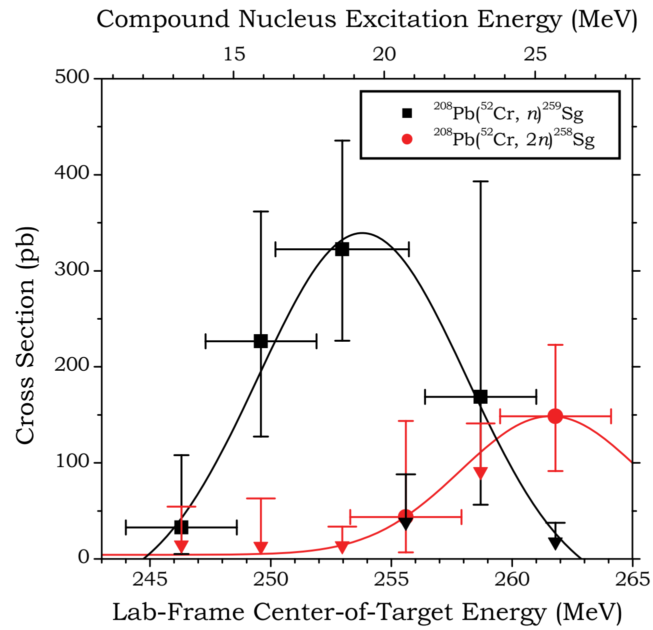

FIG. 1. (Color online) Excitation function for the production of ^{259}Sg (squares) and ^{258}Sg (circles) in the reaction of ^{52}Cr with ^{208}Pb. The abscissa shows the energy of the projectile in the laboratory frame at the center of the targets. Upper limits are indicated with arrows at the 84% (1.84-event) confidence limit. The vertical error bars represent statistical and systematic errors, and the horizontal error bars represent the range of projectile energies subtended by the targets.

A partial excitation function for the ^{208}Pb(^{52}Cr, $2n$)^{258}Sg reaction was measured but additional measurements are needed to complete the $2n$ excitation function. The maximum observed cross section is 150^{+74}_{-57} pb at $E_{\text{cot}} = 261.8$ MeV. For comparison, previous measurements of the maximum cross section of the analogous ^{209}Bi(^{51}V, $2n$)^{258}Sg reaction are 38 $\pm$ 13 pb [8] and 50^{+30}_{-20} pb [7]. Using laboratory-frame Coulomb barriers calculated from Eq. 5 in [28], the maximum of the ^{208}Pb(^{52}Cr, $2n$)^{258}Sg reaction is $\approx$2.4 MeV below the barrier while that for the ^{209}Bi(^{51}V, $2n$)^{258}Sg reaction is peaked $\approx$7 MeV below the barrier, possibly explaining the difference in their cross sections. Since these two reactions produce the same CN, this difference may also be due to a hindrance in the ^{51}V + ^{209}Bi reaction resulting from the need to pair the odd proton in the projectile and target when forming the CN.

Recently, the ^{208}Pb(^{54}Cr, xn)$^{262-x}$Sg reaction has been studied by Antalic *et al.* [6]. They observed maximum $1n$ and $2n$ cross sections of approximately 1900 $\pm$ 190 pb and 600 $\pm$ 120 pb (see Fig. 4 in [6]), respectively, while our current study finds that the corresponding cross sections with ^{52}Cr beams are 320^{+110}_{-100} pb and 150^{+74}_{-57} pb, respectively. These results indicate a ratio of ^{54}Cr/^{52}Cr $1n$ cross sections of $5.9^{+2.0}_{-2.1}$, while Dragojević *et al.* measured a ratio of 101^{+34}_{-22} in the case of ^{50}Ti/^{48}Ti [29]. The fusion by diffusion theory [28] with updated parameters [30] predicts these ratios to be $\approx$18 for ^{54}Cr/^{52}Cr and $\approx$37 for ^{50}Ti/^{48}Ti, which can be considered fair agreement with the experimental data given the large error bars reported.

The differences in cross section ratios with different projectile pairs can be attributed to the interplay of the Q-value for CN formation, Q_{CN}, and the energies relative to the Coulomb barriers. In all four cases (projectiles of ^{48,50}Ti and ^{52,54}Cr reacting with ^{208}Pb), the maxima of the excitation functions are observed to occur at CN excitation energies in the narrow range of 16.0–18.6 MeV. Thus, the energy required to produce the initial CN excited state is the dominant factor in determining the most favorable $1n$ energy, as suggested by the "Optimum Energy Rule" [28]. At the same time, the maxima for the more neutron-rich ^{54}Cr and ^{50}Ti $1n$ reactions are 8.2–8.6 MeV below the laboratory-frame Coulomb barrier, while those for the ^{52}Cr and ^{48}Ti $1n$ reactions are 14.6–15.3 MeV below. The fusion cross section is larger closer to the barrier, resulting in larger EVR cross sections for the neutron-rich projectiles. Comparing projectiles with the same Z, Q_{CN} is 4.0 MeV more negative for ^{54}Cr than ^{52}Cr, while it is 5.5 MeV more negative for ^{50}Ti than ^{48}Ti. When Q_{CN} is more negative, higher projectile energies are needed to reach the optimum excitation energy, leading to larger EVR cross sections for neutron-rich projectiles. Thus, the difference in Q_{CN} explains the difference in the ^{54}Cr/^{52}Cr and ^{50}Ti/^{48}Ti ratios.

Summary and conclusions. The ^{208}Pb(^{52}Cr, n)^{259}Sg excitation function has been measured using the BGS at the LBNL 88-Inch Cyclotron. It is symmetric in shape, in agreement with other cold fusion excitation functions, and the maximum cross section is 320^{+110}_{-100} pb at $E_{\mathrm{cot}} = 253.0$ MeV. In total, 25 decay chains from ^{259}Sg and nine decay chains from ^{258}Sg were observed. The observed decay properties are in good agreement with previous reports.

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