Development of an Odd-*Z*-Projectile Reaction for Heavy Element Synthesis: ^{208}Pb(^{64}Ni, *n*)^{271}Ds and ^{208}Pb(^{65}Cu, *n*)^{272}111

C. M. Folden III,[1,2] K. E. Gregorich,[1] Ch. E. Düllmann,[1,2] H. Mahmud,[1] G. K. Pang,[1,2] J. M. Schwantes,[1,2] R. Sudowe,[3] P. M. Zielinski,[1,2] H. Nitsche,[1,2] and D. C. Hoffman[1,2]

[1] Nuclear Science Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720

[2] Department of Chemistry, University of California, Berkeley, California 94720

[3] Chemical Sciences Division, Lawrence Berkeley National Laboratory, Berkeley California 94720

Seven ^{271}Ds decay chains were identified in the bombardment of ^{208}Pb targets with 311.5- and 314.3-MeV ^{64}Ni projectiles using the Berkeley Gas-filled Separator. These data, combined with previous results, provide an excitation function for this reaction. From these results, an optimum energy of 321 MeV was estimated for the production of ^{272}111 in the new reaction ^{208}Pb(^{65}Cu, *n*). One decay chain was observed, resulting in a cross section of $1.7^{+3.9}_{-1.4}$ pb. This experiment confirms the discovery of element 111 by the Darmstadt group who used the ^{209}Bi(^{64}Ni, *n*)^{272}111 reaction.

PACS numbers: 25.70.-z, 25.70.Gh, 23.60.+e, 27.90.+b

Cold nuclear fusion reactions have been successfully used in the production of superheavy elements, most notably in experiments leading to the discovery of elements 107-111 and production of element 112 (see summary in [1]). Other laboratories have successfully used these reactions for the production of transactinide elements; e.g., Ref. [2-4]. These reactions use shell-stabilized ^{207,208}Pb and ^{209}Bi targets with projectile energies near the Coulomb barrier to produce compound nuclei with low excitation energies (≈10-16 MeV [1]). Only one neutron evaporates from the compound nucleus and the interfering exit channels 2*n*, *pxn*, αxn, etc. are nearly completely eliminated. In the case of elements 110 (darmstadtium, Ds) and 111, a change of target from ^{208}Pb to ^{209}Bi allows for the production of both using ^{64}Ni projectiles. This additional proton in the target results in a decrease of the maximum 1*n* cross section from 15^{+9}_{-6} pb [1] to $2.9^{+1.9}_{-1.3}$ pb [5].

Among the goals of our experiments were production of ^{272}111 in the odd-*Z* projectile reaction $^{65}_{29}\mathrm{Cu} + {}^{208}_{82}\mathrm{Pb}$, and comparison of the resulting cross section to that obtained using the $^{64}_{28}\mathrm{Ni} + {}^{209}_{83}\mathrm{Bi}$ reaction. The ^{208}Pb(^{64}Ni, *n*)^{271}Ds excitation function was measured first to aid in the estimation of the optimum ^{65}Cu beam energy for the production of ^{272}111. The current work serves as an independent confirmation of the discovery of element 111 [6] using a different reaction. It also demonstrates a rather simple method for estimating optimum projectile energies for production of heavy elements in cold fusion reactions which have very narrow excitation functions. Experiments were carried out at the Lawrence Berkeley National Laboratory (LBNL) 88-Inch Cyclotron using the Berkeley Gas-filled Separator (BGS). The BGS has been described previously [7], although several improvements have been made. The focal plane strip detector now contains 48 vertical strips with an active area 179 mm wide by 58 mm high. Horizontal positions are determined by the 3.625-mm wide strips (numbered 0-47 from high to low magnetic rigidity) and vertical positions (from -29 mm to +29 mm) are determined by resistive charge division. Thirty-two non-position-sensitive “upstream” detectors are mounted perpendicular to the face of the main strip detectors. An event depositing more than ≈300 keV in a strip detector or an upstream detector triggers the list-mode data acquisition system. Twelve "punchthrough" detectors mounted directly downstream of the strip detectors provide a veto for light, low-ionizing particles which pass through the strip detectors. All detectors consist of 300-μm-thick Si. Detectors were calibrated using external α-particle sources and products of the ^{40}Ar + ^{208}Pb and ^{64}Ni, ^{65}Cu + ^{116,120}Sn, ^{nat}BaBr$_2$ reactions. Energy calibrations for high-energy events were extrapolated from these data. The energy uncertainty σ_E was ≈20 keV for α particles fully stopped in the focal plane and ≈50 keV for α particle events "reconstructed" from the sum of focal plane and upstream detector energies (all uncertainties in this paper are at the 1σ [68%] confidence level). In general, the position resolution for a single event σ_{pos} was ≈$2800E^{-1}$ keV mm. A multi-wire avalanche counter upstream of the focal plane discriminated implantation events from radioactive decays. An online analysis program using a preliminary calibration initiated a fast beam shutoff (≈140 μs) upon detecting a heavy element decay chain. Targets were 470 ± 60-μg/cm^2 (98.4% ^{208}Pb, 1.1% ^{207}Pb, 0.5% ^{206}Pb) deposited on 35-μg/cm^2 ^{nat}C backing foils and covered with 5-μg/cm^2 ^{nat}C. Nine arc-shaped target segments were mounted on the periphery of a 35.6-cm diameter wheel rotating at 450 rpm. The beam passed through a 45-μg/cm^2 ^{nat}C entrance window (separating the evacuated beamline from the gas-filled separator) and a negligible amount of He gas before striking the target backing. Beam energy losses were calculated using the SRIM-2003 program [8].

Ginter *et al*. [4] observed two ^{271}Ds decay chains in the ^{208}Pb(^{64}Ni, *n*) reaction at a center-of-target energy E_{cot} of 309.2 MeV with a cross section of $8.3^{+11}_{-5.3}$ pb. (All energies in this paper are reported in the laboratory frame unless otherwise specified). In the current experiments, two additional energies were run. In the first, a dose of 2.9×10^{17} ^{64}Ni^{14+} projectiles at 319.8 MeV was collected during a five-day experiment. At E_{cot} = 314.3 MeV the calculated center-of-target excitation energy E^{*}_{cot} was 16.2 MeV using masses from Ref. [9]. The BGS magnets were set to a magnetic rigidity $B\rho$ of 2.04 T m, based on results from Ref. [4]. The BGS was filled with He at 0.5-0.9 torr. In the second, a dose of 2.3×10^{17} $^{64}\text{Ni}^{14+}$ projectiles at 317.0 MeV was collected in five days of beam time, with E_{cot} = 311.5 MeV and E^{*}_{cot} = 14.1 MeV. Based on the results of the first run, the BGS magnets were readjusted to $B\rho$ = 2.06 T m.

Correlated EVR-α and α-α searches were conducted offline using two independent analysis codes. Decay chains attributed to ^{271}Ds are shown in Fig. 1(a). Chains 1-2 were observed at E_{cot} = 314.3 MeV and chains 3-7 were observed at E_{cot} = 311.5 MeV. The average observed EVR implantation pulse height corresponded to 25.9 ± 1.6 MeV. The observed energies have not been corrected for pulse-height defect. In all cases except chain 4 [10], the decay sequence could be followed to the decay of ^{255}No ($t_{½}$ = 3.1 min).

The data are in excellent agreement with previously reported ^{271}Ds decay data [1-2,4]. The α events assigned to ^{271}Ds in chains 5-7 have an average energy of 10753 keV and are assigned to the known transition at 10738 keV. The 10688-keV event in chain 1 can be assigned to either the transition at 10681 keV or the one at 10738 keV within reconstructed α event error. The half-life of the six ^{271}Ds events (chains 1-3, 5-7) was calculated to be $1.6^{+0.9}_{-0.5}$ ms. (The MLDS code [12] was used to calculate half-lives and uncertainties). This result is in excellent agreement with the reported half-life of $1.63^{+0.44}_{-0.29}$ ms [2].

Only online analysis results are available [10] for chain 4. An implantation event was followed by two high-energy α events (9880 keV and 9227 keV) that can be assigned to the decays of ^{267}Hs and $^{263\text{m}}$Sg, respectively. The remaining α particles in this chain may have been emitted out of the front of the focal plane (≈45% probability each) and were not recorded by the online analysis. The 22-ms lifetime reported with ^{267}Hs in chain 4 is the sum of the ^{271}Ds and ^{267}Hs lifetimes as reported by the online analysis.

The ^{267}Hs events from chains 1, 4, and 5 with an average energy of 9877 keV are consistent with the known 9882-keV decay. The other full-energy α event in chain 2 (9830 keV) can be assigned to the 9829-keV transition. The observation of two unusually short ^{267}Hs lifetimes of 0.482 ms and 2.45 ms in chains 2 and 7, respectively, suggests the presence of a short-lived isomer with half-life $0.94^{+12}_{-4.5}$ ms. The other ^{267}Hs lifetimes (chains 1, 3, 5, and 6) are consistent with a half-life of 55^{+32}_{-18} ms, in good agreement with the previously reported ^{267}Hs half-lives of 59^{+30}_{-15} ms [1] and 53 ms [2].

The six full-energy ^{263}Sg events in chains 1-6 have an average energy of 9245 keV and are assigned to ^{263}Sg^{m} (E_α = 9248 keV) [11]. The half-life of these six events is 290^{+170}_{-90} ms, in excellent agreement with the known ^{263}Sg^{m} half-life (310^{+160}_{-80} ms). The lifetime of the escape event in chain 7 (270 ms) is consistent with both the ^{263}Sg^{m} and ^{263}Sg^{g} ($t_{½}$ = 800 ± 200 ms) half-lives, so no definitive assignment is possible.

The ^{259}Rf events from chains 5 (8898 keV) and 7 (8908 keV) clearly belong to the known ^{259}Rf transition at 8895 keV. The chain 2 event (8863 keV) can be assigned to the transition at 8861 keV. Within uncertainties, the reconstructed α event in chain 3 (8792 keV) can be assigned to the transition at 8756 keV, and the completely stopped α event from chain 1 (8878 keV) can be assigned to either of the 8861- and 8895-keV transitions. The measured half-life of all six ^{259}Rf events is $2.2^{+1.7}_{-0.8}$ s, compared to the previously reported values 2.6 s [2] and $2.6^{+1.4}_{-0.7}$ s [1].

The overall count rates of α particles (fully stopped and reconstructed) at full beam intensity in a 3.0-mm pixel from 8500-12000 keV in the E_{cot} = 314.3 MeV and E_{cot} = 311.5 MeV runs were 7.8×10^{-6} Hz and 1.7×10^{-5} Hz, respectively. These rates were used to estimate the Poisson probability of observing exactly two α events within 15 s of an implantation event, since all ^{271}Ds decay chains had at least two fully-stopped α particles among the first four members of the chain. Multiplying the number of implantation events and the Poisson probabilities, the expected numbers of random EVR-α-α correlations were 6.2×10^{-4} and 5.6×10^{-3}, respectively. These calculations give upper limits because there are no requirements that the observed events agree with previous data or that additional escape α particles be present to complete the decay chain.

Six events observed in these decay chains were assigned to ^{255}No based on its known α decay properties. The observed half-life of all six events was 200^{+140}_{-70} s, compared to 190 ± 10 s measured previously [11]. An analysis similar to the one above indicates that each ^{255}No event has a probability of ≈5% of resulting from a random correlation.

The observed cross sections at E_{cot} = 314.3 MeV (E^{*}_{cot} = 16.2 MeV) and E_{cot} = 311.5 MeV (E^{*}_{cot} = 14.1 MeV) are $7.7^{+10}_{-5.2}$ pb and 20^{+15}_{-11} pb, respectively. Uncertainties in cross sections are computed according to methods described in Ref. [13]. Chain 3 was obtained with a set of targets that was too thick for an accurate cross section measurement and is not included in cross section calculations. Its observed lifetimes and decay energies have been used in the appropriate discussions. Combined with previous results [4], the current work establishes an excitation function for the ^{208}Pb(^{64}Ni, n)^{271}Ds reaction at the LBNL 88-Inch Cyclotron. The average magnetic rigidity of all seven ^{271}Ds EVRs was $B\rho$ = 2.09 ± 0.03 T m.

Using Ref. [14-15] an estimate was made of the optimum ^{65}Cu + ^{208}Pb bombarding energy from the optimum ^{64}Ni + ^{208}Pb bombarding energy. These references suggest that the maximum 1*n* cross section is obtained ≈0.3 MeV above the threshold for second-chance fission. It was estimated [15] that the maximum cross section of the ^{208}Pb(^{65}Cu, *n*)^{272}111 reaction should be obtained at E_{cot} 9.6 MeV higher than in the ^{208}Pb(^{64}Ni, *n*)^{271}Ds reaction. Thus, a new experiment was performed with a ^{65}Cu beam at E_{cot} = 321.1 MeV (E^{*}_{cot} = 13.2 MeV). The beam energy out of the cyclotron was 326.9 MeV. The same targets and experimental conditions were used in this experiment as in the two ^{271}Ds runs. The magnetic rigidity of ^{272}111 in this reaction was estimated to be 2.08 T m. During two five-day runs, a total dose of 6.6×10^{17} ^{65}Cu^{15+} particles was collected.

One decay chain of interest was observed in the experiment and is shown in Fig. 1(b). An implantation event with a pulse height corresponding to 28.58 MeV was followed 0.263 ms later by an α event with energy 11042 keV. This energy agrees well with the 11027-keV ^{272}111 α energy group reported in Ref. [5] and the main α group at ≈11000 keV in Fig. 4(b) of Ref. [2]. The observed lifetime was 0.263 ms, compared to the reported half-lives of $1.6^{+1.1}_{-0.5}$ ms [5] and $3.8^{+1.4}_{-0.8}$ ms [2]. A subsequent 10114-keV α decay is consistent with the 10097-keV ^{268}Mt event observed in chain 2 of Ref. [6] within uncertainty and the observed 12.6-ms lifetime agrees with the reported ^{268}Mt half-lives of 42^{+29}_{-12} ms [5] and 21^{+8}_{-5} ms [2]. These three signals, correlated in energy, time, and position, provide evidence of the presence of a high-*Z* element, which we assign to ^{272}111 based on the similarity of the observed decay properties to those observed previously by other groups.

Additionally, three more α-like signals were observed following the ^{268}Mt event. An escape α is assigned to ^{264}Bh, as the observed lifetime of 1.16 s agrees well with the reported ^{264}Bh half-lives of $1.0^{+0.7}_{-0.3}$ s [5] and $0.9^{+0.3}_{-0.2}$ s [2]. A reconstructed α event, with sum energy 9416 keV, is assigned to the decay of ^{260}Db as its relatively high decay energy is consistent with the α group at ≈9300 keV shown in Fig. 4(b) of Ref. [2]. The observed lifetime (1.45 s) is consistent with the previously reported ^{260}Db half-life (1.52 ± 0.13 s [11]). Within the detector resolution, a second reconstructed α with energy 8613 keV and lifetime 3.16 s can be assigned to the known ^{256}Lr ($t_{½}$ = 28 ± 3 s) transition at 8635 keV [11]. No other α events with energy greater than 1000 keV or fission events were observed at any location in the strip within 150 s of the implantation event.

The observed ^{272}111 EVR magnetic rigidity in He was $B\rho$ = 2.10 T m. The same random correlation analysis was performed as for the ^{271}Ds experiments. The beam did not shut off during this event (see Fig. 1) because of a bias supply problem in the beam "chopper," so the calculation must be done with count rates at maximum beam intensity. The average count rate of α particles (fully stopped and reconstructed) in both runs with energies 8200-12000 keV in a 3.0-mm pixel was 4.8×10^{-6} Hz. Based on the number of implantation events and the Poisson probability of observing two or four α events within 180 s of an implantation event at the above count rates, the expected numbers of random EVR-α-α and EVR-α-α-α-α correlations due to unrelated events are 0.14 and 8.5×10^{-9}, respectively

Based on the observed decay chain, a cross section of $1.7^{+3.9}_{-1.4}$ pb was calculated for the ^{208}Pb(^{65}Cu, n)^{272}111 reaction. In agreement with predictions [15], it appears to be somewhat lower than the cross section maxima of $2.9^{+1.9}_{-1.3}$ pb [5] and $2.6^{+2.3}_{-1.5}$ pb [2] reported for the ^{209}Bi(^{64}Ni, n)^{272}111 reaction.

In conclusion, the cross section of the ^{208}Pb(^{65}Cu, n)^{272}111 reaction has been measured for the first time and is comparable to that for the ^{209}Bi(^{64}Ni, n)^{272}111 reaction. Our work provides an independent confirmation of the discovery of element 111 [6] in a new reaction. Such confirmation experiments are critical in establishing the validity of claims for the discovery of new elements.

We thank D. Leitner and the staff of the LBNL 88-Inch Cyclotron for developing and providing intense, stable beams of ^{64}Ni and ^{65}Cu. The authors wish to express their appreciation to W. J. Swiatecki for many stimulating and informative discussions. This work was supported in part by the Office of High Energy and Nuclear Physics, Nuclear Physics Division, United States Department of Energy under contract number DE-AC03-76SF00098.

REFERENCES AND FOOTNOTES

[1] S. Hofmann, Rep. Prog. Phys. **61**, 639 (1998).

[2] K. Morita *et al.*, Nucl. Phys. **A734**, 101 (2004).

[3] S. Grévy *et al.*, J. Nucl. Radiochem. Sci. **3**, 9 (2002).

[4] T. N. Ginter *et al*., Phys. Rev. C **67**, 064609 (2003).

[5] S. Hofmann *et al*., Eur. Phys. J. A **14**, 147 (2002).

[6] S. Hofmann *et al*., Z. Phys. A **350**, 281 (1995).

[7] K. E. Gregorich and V. Ninov, J. Nucl. Radiochem. Sci. **1**, 1 (2000).

[8] J. F. Ziegler, computer code SRIM-2003, available from http://www.srim.org.

[9] W. D. Myers and W. J. Swiatecki, Nucl. Phys. **A601**, 141 (1996).

[10] As intended, our data acquisition system periodically closed the current data file and opened a new one. A previously undetected design flaw allowed the data from chain 4 to be discarded because it could not be written to disk while the system was changing files. The manufacturer has supplied us with corrected software.

[11] R. B. Firestone, *Table of Isotopes*, (John Wiley & Sons, New York, 1996), 8th ed.

[12] K. E. Gregorich, Nucl. Instrum. Methods Phys. Res. **A302**, 135 (1991).

[13] K.-H. Schmidt, C.-C. Sahm, K. Pielenz, and H.-G. Clerc, Z. Physik A **316**, 19 (1984).

[14] W. J. Swiatecki, K. Siwek-Wilczynska, and J. Wilczynski, Acta Phys. Pol. B, **34**, 2049 (2003).

[15] W. J. Swiatecki (private communication); W. J. Swiatecki, K. Siwek-Wilczynska, and J. Wilczynski, Phys. Rev. C (to be submitted).

FIGURE CAPTION

FIG. 1. Decay chains observed in the (a) ^{208}Pb(^{64}Ni, *n*) ^{271}Ds and (b) ^{208}Pb(^{65}Cu, *n*)^{272}111 reactions. Implantation event is shown above each chain. Notation $x + y = z$ indicates that strip and upstream detector signals were observed simultaneously with energies x and y, respectively, and sum z (all in keV). Black triangle in lower right corner of box indicates beam was shut off. Events with energies and positions estimated from one available strip signal only are denoted by (¶). Lifetime marked with diamond (♦) is sum of ^{271}Ds and ^{267}Hs lifetimes.

FIGURE

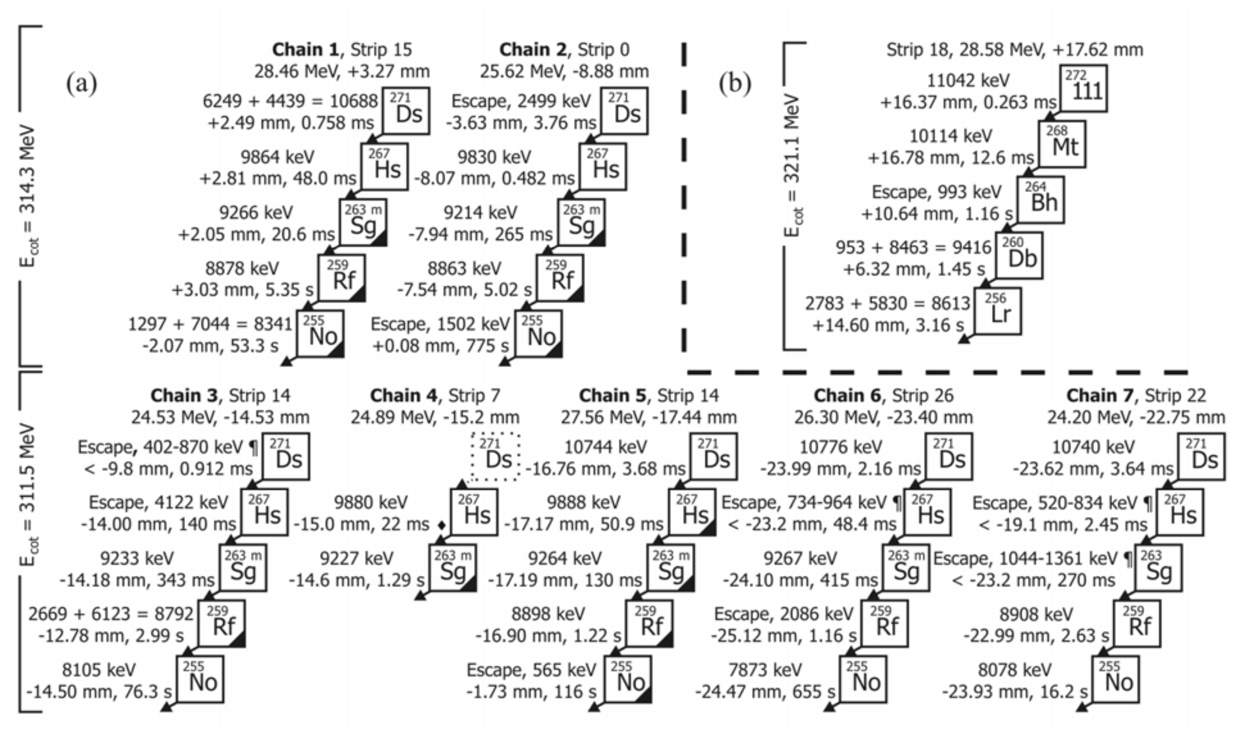
(a)
E_cot = 314.3 MeV
Chain 1, Strip 15
28.46 MeV, +3.27 mm
6249 + 4439 = 10688
+2.49 mm, 0.758 ms
271 Ds
9864 keV
+2.81 mm, 48.0 ms
267 Hs
9266 keV
+2.05 mm, 20.6 ms
263 m Sg
8878 keV
+3.03 mm, 5.35 s
259 Rf
1297 + 7044 = 8341
-2.07 mm, 53.3 s
255 No
Chain 2, Strip 0
25.62 MeV, -8.88 mm
Escape, 2499 keV
-3.63 mm, 3.76 ms
271 Ds
9830 keV
-8.07 mm, 0.482 ms
267 Hs
9214 keV
-7.94 mm, 265 ms
263 m Sg
8863 keV
-7.54 mm, 5.02 s
259 Rf
Escape, 1502 keV
+0.08 mm, 775 s
255 No
(b)
E_cot = 321.1 MeV
Strip 18, 28.58 MeV, +17.62 mm
11042 keV
+16.37 mm, 0.263 ms
272 111
10114 keV
+16.78 mm, 12.6 ms
268 Mt
Escape, 993 keV
+10.64 mm, 1.16 s
264 Bh
953 + 8463 = 9416
+6.32 mm, 1.45 s
260 Db
2783 + 5830 = 8613
+14.60 mm, 3.16 s
256 Lr
E_cot = 311.5 MeV
Chain 3, Strip 14
24.53 MeV, -14.53 mm
Escape, 402-870 keV ¶
< -9.8 mm, 0.912 ms
271 Ds
Escape, 4122 keV
-14.00 mm, 140 ms
267 Hs
9233 keV
-14.18 mm, 343 ms
263 m Sg
2669 + 6123 = 8792
-12.78 mm, 2.99 s
259 Rf
8105 keV
-14.50 mm, 76.3 s
255 No
Chain 4, Strip 7
24.89 MeV, -15.2 mm
271 Ds
9880 keV
-15.0 mm, 22 ms ◆
267 Hs
9227 keV
-14.6 mm, 1.29 s
263 m Sg
Chain 5, Strip 14
27.56 MeV, -17.44 mm
10744 keV
-16.76 mm, 3.68 ms
271 Ds
9888 keV
-17.17 mm, 50.9 ms
267 Hs
9264 keV
-17.19 mm, 130 ms
263 m Sg
8898 keV
-16.90 mm, 1.22 s
259 Rf
Escape, 565 keV
-1.73 mm, 116 s
255 No
Chain 6, Strip 26
26.30 MeV, -23.40 mm
10776 keV
-23.99 mm, 2.16 ms
271 Ds
Escape, 734-964 keV ¶
< -23.2 mm, 48.4 ms
267 Hs
9267 keV
-24.10 mm, 415 ms
263 m Sg
Escape, 2086 keV
-25.12 mm, 1.16 s
259 Rf
7873 keV
-24.47 mm, 655 s
255 No
Chain 7, Strip 22
24.20 MeV, -22.75 mm
10740 keV
-23.62 mm, 3.64 ms
271 Ds
Escape, 520-834 keV ¶
< -19.1 mm, 2.45 ms
267 Hs
Escape, 1044-1361 keV ¶
< -23.2 mm, 270 ms
263 Sg
8908 keV
-22.99 mm, 2.63 s
259 Rf
8078 keV
-23.93 mm, 16.2 s
255 No

FIG. 1.