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Brief Reports

Brief Reports are short papers which report on completed research or are addenda to papers previously published in the **Physical Review**. *A Brief Report may be no longer than 3½ printed pages and must be accompanied by an abstract.*

Search for superheavy elements using the $^{48}Ca + ^{254}Es^{g}$ reaction

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We measured upper limits for the production of superheavy elements from the compound nucleus reaction $^{48}Ca + ^{254}Es$. This combination permits the closest approach to the predicted island of stability at the 184 closed-neutron shell of any practical fusion reaction. We used aqueous and gas-phase separations to isolate superheavy element fractions within an hour from the end of the bombardments. In these experiments we did not observe superheavy elements above a limit of 3×10^{-31} cm^2 for spontaneous-fission half-lives from fractions of a day to a few months.

Theoretical calculations by Randrup *et al.*[1] and Fiset and Nix[2] predict the maximum stability for the superheavy elements (SHE's) against spontaneous fission (sf) to be at the 184-neutron closed shell. However, sf half-lives differ by 10^9 between these predictions. The calculations of Randrup *et al.* show a more rapid decrease in fission barrier height below 184 neutrons, becoming only 3–4 MeV at 178 neutrons with corresponding sf half-lives of 10^{-16} years, as compared with years or more at 184 neutrons. The half-lives at or near 184 neutrons are thus dominated by alpha or beta decay. Recent calculations by Leander *et al.* and Möller predict $Z = 114$ and 115 to be the most stable atomic numbers and the most stable neutron numbers to be shifted nearer 176 to 178, with shell corrections of 8 to 9 MeV vs 6 to 7 MeV at 184 neutrons.[3]

The sensitivity of sf half-life to neutron number for the SHE's dictates a very small choice of nuclear reactions. The prediction of maximum stability in the 176 to 178 neutron region indicates that ^{48}Ca plus ^{248}Cm, which would produce element 116 with ~ 178 neutrons, should be the best available fusion reaction, while if maximum stability is nearer 184 neutrons, as predicted in Refs. 1 and 2, the ^{48}Ca plus ^{254}Es reaction is best. However, extensive experiments using the Ca plus Cm reaction have failed to produce SHE's.[4,5] Upper limits for the production cross section of 10^{-34} to 10^{-35} cm^2 were measured for SHE's with half-lives as short as 10^{-6} s produced by this reaction.[5] These low limits indicate no enhanced stability at 176 to 178 neutrons, which results in either half-lives being below the limits of detection and/or drastically low production cross sections.

If the predictions of Randrup *et al.* are realistic, the fusion products from the Ca plus Cm reaction would be expected to have shorter half-lives ($\sim 10^{-9}$ s) than the sensitivity of any of the experiments that used the Ca plus Cm reaction. Furthermore, a fission barrier below the neutron binding energy could result in a reduction of more than 10^9 in the reaction cross section due to fission during the deexcitation of the compound nucleus. The consequences of such short half-lives or low yields make it imperative to choose a reaction that lands as close as possible to the most stable neutron shell, where the fission barriers are predicted to be highest. For these reasons, we believe the $^{48}Ca + ^{254}Es$ reaction is the most promising untested fusion reaction for the production of SHE's. For example, Randrup *et al.* predict fission barriers of greater than 6 MeV for the 2n out product of the complete fusion reaction with Es and sf half-lives comparable to the alpha half-lives. A large additional gain in half-life should be made because the 2n out product from this reaction is an odd-odd nucleus. Bjørnholm and Swiatecki noted the merits of the Es reaction in a detailed comparison of several reactions, including the ^{48}Ca reaction with ^{248}Cm.[6]

We have briefly discussed the importance of choosing a reaction capable of producing a SHE with a sufficiently long half-life and a large enough production cross section to be detectable. In addition, the survivability of fusion products requires a bombardment energy as low as possible while still permitting fusion. However, some "extra-push" energy may also be necessary for fusion[6,7] for this and several other fusion reactions capable of producing SHE's with ^{48}Ca projectiles. For these reasons, we chose two bombardment energies: one just at the estimated fusion barrier and one several MeV above to compensate for any extra-push energy that might be required for fusion in such a heavy system. The resultant excitation energies in the compound nucleus were 26 and 34 MeV, respectively.

We bombarded a target containing 1.14×10^{16} atoms/cm^2 ^{254}Es with ^{48}Ca at the SuperHILAC accelerator. At this time

the target also contained 0.7×10^{16} atoms/cm^2 of ^{250}Cf from decay of the Es. We performed four separate bombardments, each lasting several hours, for a total of 8.6×10^{15} ^{48}Ca particles. The ^{48}Ca bombarding energies were 308 and 317 MeV, as measured with a surface-barrier detector. The calculated laboratory energies entering the Es deposit were then 240 (the calculated laboratory fusion barrier) and 249 MeV, respectively, using the energy losses calculated from Hubert *et al.*[8] The Es was electroplated in a 3.0-mm-diam spot on a 2.2 mg/cm^2 Be foil that had been coated with a 0.2 mg/cm^2 Pd layer to prevent attack of the Be by the electroplating solution. A 0.02 mg/cm^2 Pd layer was then vaporized over the Es oxide deposit to reduce transfer of Es to the recoil collecting foils during bombardment. The foil arrangement in the target chamber is shown in Fig. 1.

Recoils from the Es target were collected on either beryllium or copper foils that were chemically processed to yield SHE fractions mounted on thin foils for coincident sf counting. Based on the predicted properties of SHE's,[9-11] both solution and gas-phase separation chemistries were performed. We designed these separations to recover elements 110 through 118. The primary fusion product, element 119, is predicted to decay rapidly[1,2] by alpha emission into this range of elements and thus would be detected by the sf or, possibly, α decay of its descendants.

An aqueous separation procedure was performed during the one hour from the end of bombardment (EOB) to the beginning of counting. The procedure consisted of a sulfide precipitation from a HCl solution of the dissolved Be foil followed by cation-exchange chromatography from HCl solution and a final sulfide precipitation. All of the chemical separation steps were designed to remove unwanted actinide sf emitting isotopes. The counting samples were prepared by transferring the final sulfide precipitate to a 45 μg/cm^2 carbon foil using a mixture of water and ethyl alcohol and subsequently evaporating the solution. Chemical yields of the naturally occurring elements Pt through Po were used as substitutes for elements 110 (eka-Pt) through element 118 (eka-Rn). The final sample contained about 80 μg of Au, Bi, and Pb carriers in the sulfide form. The recoil products from two of the bombardments, one at each energy, were processed using this method. Chemical yields were fairly consistent in bombardments A and B except for Pb, as shown in Table I. The yield of Pb was apparently low because of slow elution from the cation-exchange column. Because eka-Pb is predicted to be more strongly complexed in HCl, its chemical yield should have been higher.[12]

Gas-phase and a subsequent aqueous separation were performed on the recoil products from the remaining two bombardments. This separation is based on the predicted increase in volatility and noble character of elements 112 through 116 in their elemental states in comparison to their naturally occurring homologs.[10] The copper recoil catcher foil was heated to 1050 °C in a quartz apparatus using H_2 carrier gas. During the heating procedure the noble gases were condensed in a liquid nitrogen cooled cryogenic chamber and counted *in situ* for sf and α decay within 30 min after EOB. Two fractions were condensed on Pd-covered Ni foils. These fractions included elements that condense in the range of +5 to 400 °C and 400 to 1000 °C, representing Hg and Pb-like SHE's respectively. Counting for sf and α decay in these fractions was begun 2 to $2\frac{1}{2}$ hours after EOB. After collecting these volatile fractions, we dissolved the copper foil and, finally, an aqueous SHE fraction was obtained from separations based on the predicted bromide complexation of SHE's.[12] Spontaneous-fission counting was begun on this fraction about four hours after EOB. Chemical yields for these fractions are presented in Table I (bombardments C and D).

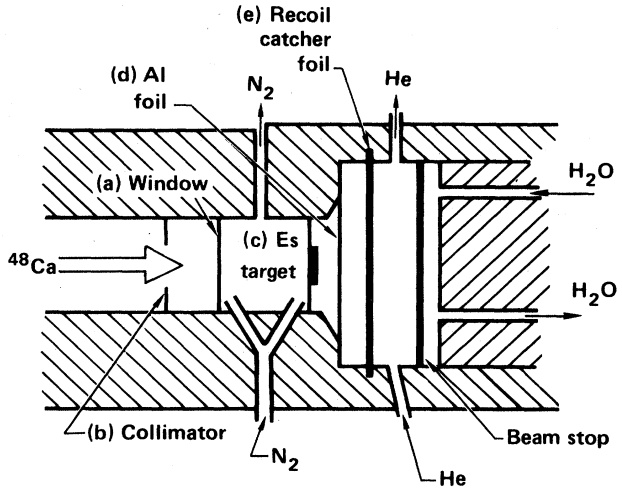

FIG. 1. A schematic diagram of the foil arrangement in the target chamber. (a) 1.8 mg/cm^2 HAVAR window. HAVAR is a trademark of the Hamilton Precision Metal Company, Lancaster, PA, and contains mostly Co, Cr, Ni, and Fe. (b) Ta collimator. (c) The Es target electroplated on a Be foil. Nitrogen gas (0.2 mg/cm^2) is directed against the HAVAR and Be for cooling. (d) A 30 μg/cm^2 Al foil to prevent transfer of the target isotope to the recoil foil. (e) A 6.3 mg/cm^2 Cu or 2.2 mg/cm^2 Be recoil-catcher foil. The 17.4-mm-diam catcher foil was 5 mm from the target.

TABLE I. Bombardment parameters and chemical yields for SHE homologs. Some yields were determined by the addition of tracers prior to the chemical separations for these bombardments. Others were estimated based on separate experiments.

	Bombardment Time (min)	Fluence ($\times10^{-15}$)	^{48}Ca^{14+} energy entering the Es (MeV)	Z: eka:	Chemical yield (percent) 110 Pt	111 Au	112 Hg	113 Tl	114 Pb	115 Bi	116 Po	117 At	118 Rn
A	773	2.22	240		40	44	48	47	18	54	43	a	a
B	383	2.14	249		47	53	55	53	21	61	69	a	a
C	338	2.02	240		85	85	75	85	80	85	85	85	10
D	393	2.19	249		85	85	75	85	80	85	85	58	10

[a]Not collected.

TABLE II. Cross sections for At and Pb isotopes from the gas-phase separations.

Bombardment energy (MeV)	Isotope	Cross section (nb)
240	^{211}At	290 ± 120
240	^{212}Pb	140 ± 30
249	^{209}At	1500 ± 400
249	^{211}At	1100 ± 200
249	^{212}Pb	590 ± 120

Spontaneous-fission counting was performed in a fragment-fragment coincidence mode with opposing surface-barrier detectors. The counting efficiency for coincident fission events was about 70%, except for the noble gas (eka-Rn) fraction, which had a counting efficiency of 40% because of the use of a single annular surface-barrier detector. In more than two months of counting, no sf coincident events with total energies greater than 40 MeV were observed in any of the counting samples. A single event with 74-MeV energy was observed after 37 days in the sulfide fraction from the 240-MeV bombardment. A total of five low-energy (< 40-MeV) coincident events were observed in the eka-Hg and eka-Pb fractions from the gas-phase separations. These events occurred between 10 h and 4 d after EOB, with total kinetic energies of 22 to 36 MeV. The low energies and lack of later events led us to conclude that these events may have been caused by electronic noise, although we observed no such events in any of the calibrations with ^{252}Cf or an alpha source. Measurements for some At and Pb isotopes were made in the SHE fractions from the gas-phase separations. Production cross sections for these isotopes (Table II) show an expected increase with energy above the fusion barrier.

Cross-section limits for SHE's were calculated on the basis of three events. This is the 95% confidence level for the observation of zero events.[13] We present two curves in Fig. 2 for the higher-energy bombardments (B and D in Table I). Curve (a) presents the results for the experiments based on sulfide precipitations that collected all of the SHE's (except elements 117 and 118 and possibly eka-Pb) with similar chemical yields at the same time after EOB. Curve (b) shows cross-section limits for the eka-Pb and eka-Hg fractions from the gas-phase separations. The eka-Rn fraction would have lower cross-section limits for shorter half-lives because of faster processing of the sample, but would have considerably higher limits at late times because of poor chemical yield. Similarly, the SHE fraction obtained with bromide complexation would have higher cross-section limits for shorter half-lives because of longer preparation times, but for longer half-lives the limits would be similar to the eka-Hg and eka-Pb curves in Fig. 2. We think the eka-Hg and eka-Pb fractions are more representative because most theoretical estimates predict that the longer-lived

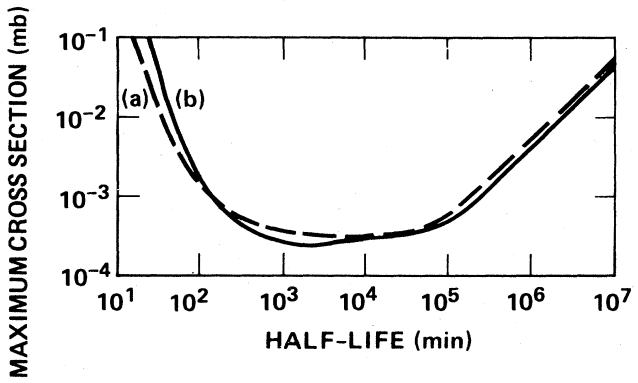

FIG. 2. Cross-section limits for the production of SHE's as a function of half-life for the two bombardments at 249 MeV. (a) Cross-section limits from the sulfide precipitations of bombardment B. A chemical yield of 55% was used to calculate the limits. (b) Cross-section limits for eka-Pb and eka-Hg from the gas-phase separations of bombardment D.

SHE's will be in these fractions. The lowest cross-section limit for SHE production is 2.5×10^{-31} cm^2 for half-lives of a few days from these experiments. If both types of separations collected SHE's the lowest limit would be 1.5×10^{-31} cm^2. The limits for the bombardments performed at the fusion barrier (A and C in Table I) would be nearly identical except for slightly higher limits at the shorter half-lives.

We believe that the ^{48}Ca + ^{254}Es reaction deserves further attention as a pathway to SHE's, particularly in view of the failure of the ^{48}Ca + ^{248}Cm reaction to produce SHE's. The LEAP[14] proposal for the production of 40 μg of ^{254}Es would make possible a target 100 times larger than that used in our experiments. With a larger target and increased bombardment times, cross sections of nearly 10^{-35} cm^2 should be achievable.

It is also important to increase the sensitivity for short lifetimes. Randrup's calculations suggest that we cannot expect a long-lived SHE to result from alpha and beta decay of element 119, the primary fusion product. Instead, one or two alpha decays, several tens of milliseconds apart, would be followed by sf decay. These millisecond half-lives are accessible by other experimental techniques such as the velocity filter SHIP[15] or a fast He-jet system.

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[1]J. Randrup, S. E. Larsson, P. Möller, A. Sobiczewski, and A. Lukasiak, Phys. Scr. **10A**, 60 (1974).
[2]E. O. Fiset and J. R. Nix, Nucl. Phys. **A193**, 647 (1972).
[3]G. A. Leander, P. Möller, J. R. Nix, and W. M. Howard, in *Proceedings of the Seventh International Conference on Atomic Masses and Fundamental Constants, Darmstadt, Seeheim, 1984,* edited by O. Klepper (GSI, Darmstadt, 1984), p. 466; P. Möller (private communication).

[4]E. K. Hulet *et al.*, Phys. Rev. Lett. **39**, 385 (1977); J. D. Illige *et al.*, Phys. Lett. **78B**, 209 (1978); R. J. Otto *et al.*, J. Inorg. Nucl. Chem. **40**, 589 (1978); A. Ghiorso *et al.*, Lawrence Berkeley Laboratory Report No. LBL 6575, 1977 (unpublished); Yu. Ts. Oganessian *et al.*, Nucl. Phys. **A294**, 213 (1978).
[5]P. Armbruster *et al.*, Phys. Rev. Lett. **54**, 406 (1985).
[6]S. Bjørnholm and W. J. Swiatecki, Nucl. Phys. **A391**, 471 (1982).
[7]H. Gäggeler, T. Sikkeland, G. Wirth, W. Brüchle, W. Bögl, G. Franz, G. Herrmann, J. V. Kratz, M. Schädel, K. Sümmerer, and W. Weber, Z. Phys. A **316**, 291 (1984).
[8]F. Hubert, A. Fleury, R. Bimbot, and D. Gardes, Ann. Phys. (Paris) **5** (1980), Suppl.
[9]B. Fricke, Struc. Bonding (Berlin) **21**, 89 (1975).
[10]B. Eichler, Kernenergie **19**, 307 (1976).
[11]K. S. Pitzer, J. Chem. Phys. **63**, 1032 (1975).
[12]J. V. Kratz, J. O. Liljenzin, and G. T. Seaborg, Inorg. Nucl. Chem. Lett. **10**, 951 (1974).
[13]D. B. Owen, *Handbook of Statistical Tables* (Addison-Wesley, Reading, MA, 1962).
[14]A. Ghiorso, D. Hoffman, E. K. Hulet, O. L. Keller, and G. T. Seaborg, LEAP (Large Einsteinium Accelerator Program), Lawrence Berkeley Laboratory Report No. PUB-5118, 1984 (unpublished).
[15]G. Münzenberg *et al.*, Nucl. Instrum. Methods **161**, 65 (1979).