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SEARCH FOR SUPERHEAVY ELEMENTS PRODUCED IN THE 136se + 238U REACTION AND AN UPPER LIMIT CROSS SECTION FOR THE natGd(136Xe, X)212pb REACTION

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Author
Otto, R.J.

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SEARCH FOR SUPERHEAVY ELEMENTS PRODUCED IN THE $^{136}$Xe + $^{238}$U REACTION AND AN UPPER LIMIT CROSS SECTION FOR THE

$^{\text{nat}}_{\text{Gd}}(^{136}\text{Xe},X)^{212}\text{Pb}$ REACTION*


Lawrence Berkeley Laboratory
University of California
Berkeley, California 94720

Summary

A search has been made for spontaneous fission activity in four different chemical fractions following irradiations of thick $^{238}$U targets with 1150 MeV $^{136}$Xe beams. The chemically separated fractions were a sulfide precipitate, a complexed metal bromide fraction, a volatile metal bromide fraction, and a noble gas fraction. No evidence was found for superheavy elements, which would correspond to the transfer of $\sim$50 nucleons. In addition an analogous experiment has been done to look for alpha particle activity from elements near doubly magic $^{208}$Pb produced in the $^{136}$Xe + $^{\text{nat}}_{\text{Gd}}$ reaction also corresponding to the transfer of $\sim$50 nucleons. Based on an upper limit cross section for $^{212}\text{Pb}$ the usefulness of the $^{136}$Xe + $^{238}$U reaction as a test for the existence of the island of stability is discussed.

I. Introduction

There are only two known reaction mechanisms for the $^{136}$Xe + $^{238}$U reaction, the deep inelastic transfer reaction (DIT) and the quasi-elastic

*Work supported in part by the Division of Physical Research, U. S. Department of Energy.
transfer reaction (QET) (1). The probability of producing superheavy elements (SHE) in the region of $^{298}_{114}\text{(SHE)}$ will depend on, first, the probability of transferring about 22 protons and $38 + n$ neutrons from the $^{136}\text{Xe}$ projectile to the $^{238}\text{U}$ target nucleus, and second, on the fission stability of the SHE's in the transition process (emission of $n$ neutrons) leading to a ground state nucleus. Assuming that the SHE's have a reasonable stability against fission at excitation energies of less than $\sim 45$ MeV (2), the second factor depends on the fraction of those events with a large enough transfer of protons and neutrons that also result in SHE fragments with sufficiently low values of excitation energy ($<45$ MeV) and angular momentum to reduce losses by fission. It is the nucleon transfer factor that we have tried to test using the $^{136}\text{Xe} + \text{nat Gd}$ (21.9% $^{160}\text{Gd}$) reaction to produce elements in the closed shell region of $^{208}\text{Pb}$ via a transfer reaction. In order to produce $^{212}\text{Pb}$, for instance, $18 + p$ protons and $34 + n$ neutrons have to be transferred from $^{136}\text{Xe}$ to $^{160}\text{Gd}$. The transfer reaction that could occur would be $^{160}\text{Gd}(^{136}\text{Xe}, 84-n\text{Kr})^{212+n}\text{Pb}$ where an allowance is made of $n$ neutrons for deexcitation leading to $^{212}\text{Pb}$. The most favorable aspect of this reaction is the negligible losses due to fission for the transfer products in the lead region (3). In addition there is evidence (4) that the degree of nucleon diffusion in the deep inelastic transfer process for lighter systems such as Kr + Ho and Xe + Ho are comparable with the heavier Kr and Xe + Bi systems. However, the transfer of $\sim 60$ nucleons is expected to correspond to a cross section as small as a nanobarn (4).
Assuming the production of SHE's and taking into account the large uncertainties in their half-lives (5), there is the remaining (although smaller) uncertainty in their predicted chemical properties. We have considered four possibilities: (a) SHE's will form insoluble sulfide compounds and/or be reduced in the presence of H₂S and will be coprecipitated with CuS (6); (b) the SHE's form anionic metal bromide complexes, or (c) volatile neutral metal bromide compounds (7); and (d) the SHE's behave chemically as noble gases (8).

The existence of spontaneous fission activity in chemical fractions with elements having the above chemical properties, and the assurance of actinide element elimination from these fractions, could be interpreted as evidence for SHE's. The lack of spontaneous fission activity in the same chemical fractions could result from incorrect assumptions ranging from their postulated ground state stability and half-lives to their assumed chemical properties, or it could indicate that elements in the SHE region are not produced in the chosen reaction.

A short review of the search for SHE's was given recently by Bemis and Nix (9). The reaction of heavy ions ranging from ⁴⁰Ar (10) and ⁴⁸Ca (11, 12) to ²³⁸U (13) with various actinide targets were tried. Flerov and Oganessian (14) reported that a spontaneous fission activity with 150-day half-life can be isolated in a sulfide precipitate fraction following the irradiation of ²³⁸U with about 900 MeV ¹³⁶Xe. The reported cross section was given as ~10⁻³³ cm².
II. Search for Superheavy Elements - Experiments

Several experiments have been carried out over the last three years. These experiments are summarized in Table 1. In the most recent experiment (experiment 1) the $^{136}$Xe beam energy was monitored continuously using a gold scatter foil supported on a thin Al backing. The average beam energy was 1150 MeV ± 50 MeV (FWHM). The target was a 19-mm disc of natural uranium, 0.25-mm thick. The range of $^{136}$Xe from the surface of the target to the reaction barrier was taken to be 17 mg/cm$^2$ for all of the Xe + U experiments. Following the experiment the target was dissolved in a 10 ml solution of 2 M HCl containing 1 ml of 30% H$_2$O$_2$. The solution was prefiltered to remove undissolved carbon deposits that formed on the surface of the target during the irradiation. A copper carrier (124 µg) was added to the solution followed by a few ml of saturated H$_2$S solution. The CuS precipitate was filtered onto a 6-mm diameter area, using cellulose nitrate filter paper. The sample was briefly counted for alpha particle activity to check the thickness of the precipitate. The CuS sample was then redissolved using hot concentrated aqua regia, diluted and reprecipitated as CuS. The chemical yield for this sample was estimated to be ~50% based on the $^{211}$At alpha activity. A second CuS scavenging of the solution was carried out and this fraction contained approximately 25% of the $^{211}$At activity. Each sample was placed within 1 mm of a 300-mm$^2$ area gold surface barrier detector. These samples were pulse height analyzed continuously for spontaneous fission and alpha events for approximately 200 days. The detection efficiency for spontaneous fission events for this system is 50% ± 10%.
The noble gas fraction (two runs—experiments 2a and 2b) was obtained following irradiations of 0.25-mm × 16-mm diameter natural uranium metal targets. After the heavy-ion irradiations the targets were placed in a sidearm flask which was connected with Tygon tubing to a calcium sulfate drying tube followed by a cold (−20°C) activated charcoal trap. (The activated charcoal consists of a new coconut charcoal which has been baked under vacuum for several hours.) After the apparatus was purged of air with helium, ~100 mg/cm² was dissolved from the surface of the target using a solution of 250 λ HNO₃, 50 λ HCl and 400 λ H₂O (experiment 2a) or the target was completely dissolved in 9 M HCl (experiment 2b). The helium flow was continued for five minutes following dissolution. Desorption was effected by gradually heating the charcoal to about 325°C. The gases migrated to a mica disc placed at the end of a copper rod immersed in liquid nitrogen. A condensation efficiency of 90% ± 8% was determined by using ²²²Rn (3.82 day) from a natural uranium generator solution in place of the dissolved target. The condensed fraction was positioned 2.5 mm from a gold surface barrier detector. The total spontaneous fission counting efficiency was determined to be 14% ± 1%. The separation factor from all elements other than the noble gas elements was found to be greater than 10⁷. Separate pulse height analysis was done for alpha particle events and spontaneous fission events. Radon alpha activity was observed in the rare gas fractions, and a production cross section of 0.7 ± 0.2 mb for ²¹¹Rn was determined consistent with the mass distribution studies (1). A more detailed report of the heavy-ion noble gas experiments is given elsewhere (15).
Experiments 3 and 4 have been done using the chemical procedures developed by Kratz et al. (16). In these experiments the beam energy was checked intermittently during the irradiation. The targets were fabricated by sputtering 30 mg/cm$^2$ of depleted uranium metal onto a 1-cm diameter area of an aluminum disc. Precautions were taken to trap any superheavy elements that might normally escape during the dissolution of the target. Also, the elemental halide volatilization step was omitted and the superheavy element (SHE) fraction and lead (Pb) fraction were combined. The combined SHE and Pb fractions from experiment 3 were taken to dryness on a glass disc and positioned below a surface barrier detector. The volatile bromide fraction obtained from experiment 4 was precipitated as As$_2$S$_3$ using 200 µg of As$^{+3}$ and a trace amount of Hg$^{+2}$ in solution, and filtered using cellulose nitrate filter paper. These samples were pulse height analyzed for spontaneous fission and alpha particle activity. The spontaneous fission counting efficiency was 15% ± 5%.

III. Search for Superheavy Elements - Results

The results of the spontaneous fission counting for each of the chemical fractions except the noble gas fraction can be seen in Fig. 1c, 1d and 1e. The total number of events observed in the spontaneous fission detection systems for each fraction are also listed in Table 1. Figure 1a shows a $^{248}$Cm spontaneous fission energy spectrum obtained.

*The targets were fabricated by E. K. Hulet and staff at the Lawrence Livermore Laboratory.
from a thin source and counted using the same electronic system, a similar detector and the same geometry conditions as were used in the most recent SHE experiment (experiment 1). Figure 1b shows the spontaneous fission energy distribution from $^{248}\text{Cm}$ dispersed in an $\text{As}_2\text{S}_3$ precipitate so that the geometry and sample thickness were comparable to the SHE precipitate samples (experiments 1 and 4). (The spontaneous fission spectrum shown in Fig. 1b was obtained by precipitating $\text{As}_2\text{S}_3$ at pH 2 from a solution containing $\approx 10^3$ spontaneous fissions per minute of $^{248}\text{Cm}$. Approximately 0.1% coprecipitated under these conditions.)

The events summarized in Table 1 and shown in Fig. 1c, 1d and 1e cannot be spontaneous fission events for two reasons. First, the energy distribution is completely different than the measured energy distribution shown in Fig. 1b for $^{248}\text{Cm}$ coprecipitated with $\text{As}_2\text{S}_3$ and counted under the same geometry conditions. Second, we have recently found a correlation between the frequency of these events and the bias voltage applied to the detectors. We have reduced the background events to less than one low energy count per 40 days by lowering the bias voltage on the surface barrier detectors.

Three events were observed while counting spontaneous fissions in the noble gas fractions from experiments 2a and 2b. The energies were 110 MeV, 68 MeV and 40 MeV and these events were detected 1 hr, 18 hr and 24 hr respectively after the end of the bombardment. The mica detector present during the experiment which produced the 68 MeV count registered a single track. These counts associated with the noble gas fraction cannot be easily explained as spurious events because
they occurred a short time after the irradiation, there were no background counts 24 hours prior to the experiment, and a high degree of pileup rejection was used in the system.

Based on the assumption of an upper limit of one valid spontaneous fission event in each experiment, upper limit production cross sections have been calculated. An upper limit for the noble gas experiments for half-lives between 30 minutes and 100 hours is set at \(4 \times 10^{-34}\) cm\(^2\).

Figure 2 shows the upper limit cross sections for the other fractions based on assumed half-lives and taking into account all factors, such as bombardment history, chemical yield and counting efficiency. These results indicate an upper limit cross section for a sulfide insoluble SHE with a half-life of 150 days to be less than \(1 \times 10^{-34}\) cm\(^2\).

Our results set upper limit cross sections for SHE's with half-lives of 150 days well below the nanobarn value given by Flerov (14). The variety of assumptions about the chemical properties, the use of a similar sulfide precipitation and the precautions taken in the dissolution of the target make the possibility of not observing SHE's at the nanobarn level very unlikely for the half-life region of one day to several hundred days.

### IV. The \(^{136}\)Xe + natGd Reaction

Six pieces of 99.9% pure natural gadolinium metal (21.9% \(^{160}\)Gd) were milled and fitted to make a collimator with a 0.035-mm diameter aperture. An analysis was done for Pb, Bi, and 39 other elements using
emission spectroscopy. Limits for Pb and Bi contamination were set at <0.01% weight. The collimator was used during a $^{136}$Xe irradiation of a uranium target and received a total flux of $\approx 8 \times 10^{15}$ particles. Three of the six gadolinium pieces were placed in a nitric acid solution and the active surface was dissolved. Twenty-five $\mu$g of Cu$^{+2}$ carrier was added to the solution and then precipitated using a saturated H$_2$S solution. (Lead, bismuth, polonium and astatine will precipitate quantitatively under these conditions.) The precipitate was filtered onto cellulose nitrate filter paper and the sample was pulse height analyzed for alpha particles 15 hours after the end of the bombardment in a detector showing no background above 3 MeV measured over a period of 24 hours.

A search was made for 8.78 MeV alpha particles from the $^{212}$Po daughter of 10.2 hr $^{212}$Pb. No alpha particle events were observed with energies above 7.45 MeV in over 16 hours of counting. This corresponds to an upper limit cross section of $2 \times 10^{-34}$ cm$^2$ for the production of $^{212}$Pb in the $^{136}$Xe + nat Gd reaction, and $1 \times 10^{-33}$ cm$^2$ for the $^{136}$Xe + $^{160}$Gd reaction. By making a comparison of the $^{136}$Xe + $^{160}$Gd and $^{136}$Xe + $^{238}$U reaction to make $^{212}$Pb and $^{290}$SHE, respectively, it can be seen that both reactions require the transfer of 18 protons and 34 + n neutrons. Since both irradiations were thick target experiments and include projectile energies from 1150 MeV to the barrier, there would have been transfer product nuclei heavier than the target from both reactions having the same excitation energies and corresponding to the same number of protons and neutrons transferred.
Based on the known values (3) of $\Gamma_n/\Gamma_f$ for a $^{209}$Bi compound nucleus formed by the reaction of protons with $^{208}$Pb, and the observation of less than 10% deep inelastic induced fission of gold-like fragments in the reaction (17) of 1150 MeV $^{136}$Xe + $^{197}$Au, fission competition should be unimportant (<10% fission) for Pb fragments produced in a deep inelastic reaction of $^{136}$Xe + $^{160}$Gd. We conclude that the cross section corresponding to the transfer of $\sim 18$ protons and $\sim 34$ neutrons in the $^{136}$Xe + $^{160}$Gd reaction is less than $1 \times 10^{-33}$ cm$^2$, the upper limit for $^{212}$Pb production. This limit can be applied to a similar or greater number of nucleons transferred from $^{136}$Xe to $^{238}$U if it is assumed that the nucleon diffusion rates and the interaction times are nearly the same for the $^{136}$Xe + $^{160}$Gd and $^{136}$Xe + $^{238}$U reactions. This would correspond to the production of the nuclide $^{298}$SHE, which is predicted to be a SHE in the "island of stability." Proportionately more neutrons must be transferred in the $^{136}$Xe + $^{160}$Gd reaction to produce $^{212}$Pb than in the $^{136}$Xe + $^{238}$U reaction to produce $^{298}$SHE products near the center of the island of stability. However, the larger absolute number of nucleons transferred in the $^{238}$U reaction might compensate for this effect. We have estimated the survival probability for $^{298}$SHE based on calculated values of $\Gamma_n/\Gamma_f$ by Moretto (2) to be $4 \times 10^{-3}$ for approximately 4 neutrons emitted. Thus we would estimate the production cross section of $^{298}$SHE to be less than $4 \times 10^{-36}$ cm$^2$ in the $^{136}$Xe + $^{238}$U reaction and it could be considerably less than this value due to smaller values of $\Gamma_n/\Gamma_f$. 
The transfer of 34 neutrons and 18 protons represents a higher n/p ratio than found in $^{136}$Xe (n/p = 1.52). Perhaps SHE's with adequate half-lives formed as the result of the transfer of neutrons and protons closer to the ratio n/p = 1.52 would be produced with a higher cross section than that for production of $^{290}$SHE. Such nuclides would be more neutron deficient and further removed from the closed shell n = 184. Those with higher Z (e.g., $^{290}$SHE, $^{293}$SHE) might still be within the island of stability and have half-lives suitable for detection. A test for the transfer of ~50 nucleons with n/p ratio of 1.52 would be desirable. The production of $^{210}$Po and $^{211}$At in the $^{136}$Xe + $^{160}$Gd reaction would meet these criteria fairly well.

Alpha activity from $^{211}$At and $^{210}$Po was seen in the spectra of the CuS sample from the $^{136}$Xe + nat Gd experiment. The highest energy peak in the spectrum corresponded to the 7.45 MeV (resolution of 140 KeV FWHM) alpha energy of $^{211}$Po, a daughter of $^{211}$At. However, the high ratio of yields, $^{210}$Po/$^{211}$At, pointed to a quasi-elastic transfer rather than a deep inelastic transfer mechanism as the source of the activity. In order to test whether these products might be due to impurities in the Gd target, a cross bombardment of the same Gd used for the Xe experiment was made with 105 MeV $^{12}$C. Following the irradiation the active surface of the Gd was dissolved and a CuS precipitate was prepared and counted using the same procedures described above. Both $^{210}$Po and $^{211}$At alpha particle activity was again observed in approximately the same high ratio of yields. Based on the relative yields of the $^{210}$Po and $^{211}$At activity and the known cross sections for $^{211}$At
produced in heavy ion reactions (18, 19) of $^{12}$C and $^{84}$Kr with $^{209}$Bi, it was concluded that these products are formed in transfer reactions with Bi (or Pb) contaminants present at the level of 5-10 ppm. Using the yield of $^{210}$Po that we have measured in separate experiments for the reaction of 550 MeV $^{86}$Kr + $^{208}$Pb to estimate an expected yield for $^{211}$At, all of the $^{211}$At observed in the Xe + Gd experiment can be accounted for. The upper limit cross section for $^{211}$At from the $^{136}$Xe + $^{160}$Gd reaction can be conservatively estimated to be a factor of ten less than the observed value and this upper limit corresponds to 60 nanobarns ($6 \times 10^{-32}$ cm$^2$). Again with fission competition we would set the cross sections for the production of SHE's in the more neutron deficient region of the island of stability to be $\leq 2 \times 10^{-34}$ cm$^2$ and, again, they could be much less than this due to smaller values of $\Gamma_n/\Gamma_f$. This larger value for the upper limit to the production cross section of higher atomic number SHE's does not change significantly our conclusion that these elements will be difficult to observe through production by the $^{136}$Xe + $^{238}$U reaction.

V. Conclusions

No evidence has been found for SHE's with half-lives in the range of a few hours to several hundred days produced in the reaction of $\leq$1150 MeV $^{136}$Xe + $^{238}$U. The wide range of chemical properties covered in the various studies makes it very unlikely that a sulfide insoluble SHE with a 150-day half-life would not have been seen at the production level corresponding to a nanobarn. The upper limit cross section for
the production of $^{212}\text{Pb}$ in the reaction of $^{136}\text{Xe}$ with $^{160}\text{Gd}$ is one nanobarn. Based on this result an argument is given that the upper limit cross section for the production of $^{290}\text{HSE}$ in the $^{136}\text{Xe} + ^{238}\text{U}$ reaction is $4 \times 10^{-36}$ cm$^2$. We must therefore conclude that the $^{136}\text{Xe} + ^{238}\text{U}$ reaction does not provide a good test for the existence of the predicted island of stability even for beam intensities $10^3$ (and possibly $100$) times the levels used in these experiments.

Acknowledgments

The authors would like to thank Hermann Grunder and the SuperHilac crew for their efforts in producing and maintaining the $^{136}\text{Xe}$ beams. We would also like to thank Dr. Malcolm Fowler and Dr. Irwin Binder for their participation in early phases of this work and Mr. David Morrissey for his help in the more recent bombardments.
Table 1. Summary of $^{136}\text{Xe} + ^{238}\text{U}$, SHE search experiments.

<table>
<thead>
<tr>
<th>Experiment Number</th>
<th>Fraction and Chemical Formula</th>
<th>Total Beam Particles</th>
<th>Days Counted</th>
<th>Background Events</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>CuS</td>
<td>$2 \times 10^{15}$</td>
<td>200</td>
<td>5</td>
</tr>
<tr>
<td>2</td>
<td>Condensed noble gas fraction on mica (two bombardments)</td>
<td>$5 \times 10^{15}$</td>
<td>0.02-4.2</td>
<td>3</td>
</tr>
<tr>
<td>3</td>
<td>Carrier free metal bromide</td>
<td>$2 \times 10^{15}$</td>
<td>320</td>
<td>16</td>
</tr>
<tr>
<td>4</td>
<td>Volatile metal bromide, As$_2$S$_3$</td>
<td>$1 \times 10^{15}$</td>
<td>160</td>
<td>15</td>
</tr>
</tbody>
</table>
Figure Captions

Fig. 1. Comparison of the energy of the events recorded while counting the SHE fractions with $^{248}\text{Cm}$ spontaneous fission spectrum obtained from the same counting system. The energy calibration is based on an alpha energy calibration obtained using a $^{225}\text{Ra}$ source followed by a linear pulsar calibration.

la) The $^{248}\text{Cm}$ spontaneous fission energy spectrum from a thin source.

lb) The energy spectrum of $^{248}\text{Cm}$ spontaneous fission activity coprecipitated with $\text{As}_2\text{S}_3$. Sample thickness and geometry conditions are nearly the same as those for the SHE sources in experiments 1 and 4.

lc) Energy of recorded events from experiment 1 (CuS precipitate).

ld) Energy of recorded events from experiment 3 (metal bromide fraction).

le) Energy of recorded events from experiment 4 (volatile metal bromide fraction - $\text{As}_2\text{S}_3$ precipitate).

Fig. 2. Upper limit cross sections for SHE production versus assumed half-life.
Figure 1
Figure 2
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