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Search for Superheavy Elements in the Bombardment of ^{248}Cm with ^{48}Ca

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We have searched for superheavy elements 110 to 116 with half-lives between 10^4 and 10^8 s in fractions chemically separated after each of a series of bombardments of ^{248}Cm made with 267-MeV ^{48}Ca ions. After 6 months of α and spontaneous-fission counting, our results provide no persuasive evidence for the presence for the presence of superheavy elements. The most plausible explanation for not finding the superheavy elements is that they have either short half-lives or very small formation cross sections.

Numerous searches have been made for superheavy elements (SHE's) located in the "island of stability" believed to be centered at $Z = 114$, $N = 184$. Although these searches have failed to find any convincing evidence for SHE's,[1] the quest continues. This is the first reporting of experiments in which ^{248}Cm has been bombarded with ^{48}Ca ions in the expectation of producing SHE's.

This combination of target and projectile is believed to be the most promising for the synthesis of nuclides close to the center of the island of stability,[1-4] since the neutron-proton ratio of the compound nucleus ^{296}[116] is higher than for any other practical target-projectile combination. Longer half-lives and larger formation cross sections are favored upon approaching this central region, where the stability derived from a closed neutron shell is greatest. At the same time, the relatively large mass defect of the doubly magic ^{48}Ca nucleus helps to reduce the excitation energy of the compound system. It is important that both excitation energy and angular momentum be minimized in order to retain the fragile stability contributed only by shell effects.

Half-lives and radioactive-decay modes have been estimated for many of the SHE isotopes.[5-8] Although there are large uncertaintites in the half-life estimates, it is explicit that any decay chain will eventually lead to a nuclide that undergoes spontaneous fission (SF). Therefore, we have based our detection of SHE's mainly on the observance of SF events, which is sensitive to the detection of a few atoms. Although natural backgrounds from SF are extremely low, many man-made isotopes of the heavy actinides, including ^{248}Cm, decay by SF; it was essential, therefore, to remove the actinide elements in order to identify those fission events arising from the decay of SHE's. To accomplish this, the elements 110 to 116 were chemically separated in all our experiments.

We have carried out a series of ^{48}Ca-ion bombardments with beam intensities from 3×10^{11} to 6×10^{11} particles/s and lasting from 10 h up to 2 d. The principal energy of the ^{48}Ca ions striking the target was 267 MeV, which provided some excess energy above the Coulomb barrier. This was necessary to drive the system dynamically from the point of contact (two touching spheroids) to inside the fission barrier (single-sphere).[3,9] However, above 20 MeV of excitation energy, the stability derived from shell effects is expected to diminish.[10] The ^{248}Cm target was 6 mm in diameter and contained 20 μg/mm^2 of ^{248}Cm (97% isotopic purity) deposited as CmF_3 onto 0.013-mm Be by vacuum sublimation.[11] Thus, the energy spread within the target was 267 to 243 MeV, corresponding to an average excitation energy of 40 to 50 MeV for the compound nucleus. Recoil products from each bombardment were stopped and collected in a 0.013-mm Be foil that was then chemically processed in one of three separation schemes.

Each separation scheme was used to isolate the SHE's from other reaction products and from any ^{248}Cm that may have reached the recoil collection foil. All of these schemes were based on the chemical properties anticipated for elements 110 to 116[12-16]: Their sulfides are expected to be insoluble; the metal ions are likely to be strongly complexed by bromide and chloride ions; the last three 6d elements (110 to 112) should be highly noble metals; and elements 112 and 114 may be quite volatile. Pitzer[15] and Eichler[16] have suggested that 112 and 114 in the elemental form are either inert gases or very volatile liquids. If so, these elements may not have been fully retained in the hot recoil collector during the bombardments. If, in addition, elements 112 and 114 are relatively inert chemically, it is conceivable that they escaped as gases during the dissolution of the collector foils.

The most rapid separation (Scheme I) depends only on the coprecipitation of the SHE's with CuS. Two CuS precipitations were made, one from acid solution and one from basic solution. The yields for Au, Hg, and Bi tracer elements were typically greater than 80% in identical trial runs. These samples were continuously counted for 60 d using surface-barrier detectors.

A more elaborate separation (Scheme II) was intended to remove completely all actinides and to fractionate the SHE's into as many as five separate samples. In this scheme, the actinides were adsorbed by Dowex 50×12 ion-exchange resin, while the SHE's were expected to be complexed and eluted from the column with (0.3–2)M HCl. Because of the possible noble character expected for eka-Pt, eka-Au, and eka-Hg, and the need for thin samples, controlled-potential electrolysis was subsequently employed to fractionate the SHE's. These electrodepositions were made from a buffered acetate solution at +0.3 and −0.1 V relative to a standard calomel electrode. If a SHE failed to electrodeposit, it then should have coprecipitated as a sulfide with either Pb or Pt carriers (200 μg each) that had been introduced earlier. Beside Pb and Pt carriers, 200 μg of Au, 10 μg each of Hg and Bi, and radioactive tracers of Au, Hg, Tl, Bi, and Po were also added to determine chemical yields. These yields varied from 42% for Hg to 96% for Au, except that we were unable to determine the ^{206}Tl and ^{210}Po yields. Deposits resulting from the above separations were made on muscovite mica coated with 0.3 μg/mm^2 of Cr and 1.7 μg/

mm^2 of Au to provide an electrically conductive surface. These samples have been examined for 6 months for α and fission events by pulse-height analysis and for fissions with ionization chambers. Ultimately, a search will be made for fission tracks in the mica, since this detection method affords nearly zero background over a period of years.

Another separation method (Scheme III, based on the stabilization of SHE's as bromide complexes,[17] was used to isolate the SHE's after a long bombardment. Volatile bromide compounds were distilled from an aqueous solution, trapped, and coprecipitated with As_2S_3. However, results from this sample were nullified by the finding of ^{248}Cm contamination. The nonvolatile products of this bombardment were eluted through a cation-exchange column to remove actinides. The SHE's were expected to be eluted with $0.56M$ HBr/Br_2 and $0.1M$ HBr/Br_2. The eluate from the column was evaporated onto a thin film of Vyns that was placed between two surface-barrier detectors. The yield of Au and Ir tracers was 20 and 60%, respectively. Coincident fission fragments from this sample have been counted for approximately 6 months.

A wide range of nuclides was produced in the bombardment of ^{248}Cm with ^{48}Ca. Separate analyses for actinides showed large and easily detectable quantities of Cf, Es, and Fm isotopes produced by quasielastic and deep-inelastic transfer reactions. The formation cross sections of these observed transcurium nuclides are shown as a function of mass number in Fig. 1.

Interference in the detection of α-emitting SHE's came mainly from the nuclides produced in the Pb to Pa region by deep-inelastic transfer reactions. These products emitted α's within the 7.7- to 9.0-MeV span of α energies expected for longer-lived SHE's; also, isotopes of the elements Pt through Po were present in the samples, since they were unseparated from their SHE homologs by the chemical procedures. Some of the observed nuclides in this region and cumulative formation cross sections are given in Table I. These cross sections were derived from α-pulse analysis of the CuS fractions and a LaF_3 fraction obtained in Scheme I.

We have searched this region of α energies in spectra taken from a thin sample containing Au, Hg, Po, and their SHE homologs (from Scheme II). The α spectra taken in the first 2.5 d after the end of a 25-h bombardment revealed five events belonging to ^{213}Po (8.38 MeV), 37 events belonging to ^{212}Po (8.87 MeV), two events at 7.76 and 8.04 MeV, and five unidentified events grouped between 8.61 and 8.67 MeV. Only seven random events were observed in the 7.7–9.0-MeV range in the subsequent 7 d of α counting. We considered the unidentified group of α's around 8.65 MeV as significant, but later failed to observe any such α events in SHE samples containing Au, Hg, and Po, which were obtained in a similar bombardment by a slightly different chemical treatment. Although α particles with 8.65-MeV energy have not been identified with any known isotope, we have tentatively assigned these to the isotopes of Po.

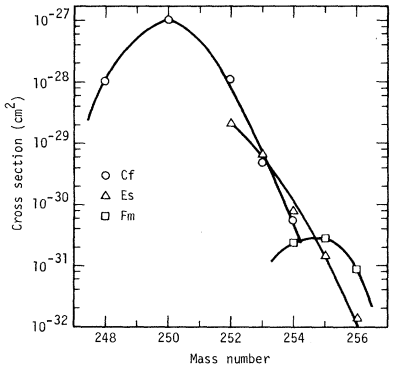

FIG. 1. Approximate yields of transcurium nuclides produced from transfer reactions in the bombardment of ^{248}Cm with ^{48}Ca. Only products recoiling within an included angle of 100 deg (lab) were measured.

TABLE I. Formation cross sections for Po isotopes obtained from separation Scheme I.

Parent isotope (α-energy, MeV, of Po daughter)	Cumulative cross section[a] (μb)
^{213}Bi (8.375)	130 ± 30[b]
^{210}Po (5.305)	740 ± 170
^{211}At (7.450)	130 ± 30
^{223}Ra (7.386)	50 ± 10[c]
^{224}Ra (8.784)	35 ± 10
^{225}Ac (8.375)	160 ± 40[d]

[a]Includes yields of all parent nuclides in α- and β-decay chains unless otherwise specified.

[b]Corrected for growth and decay formation from ^{225}Ac and ^{225}Ra.

[c]Not corrected for growth and decay formation from ^{227}Th.

[d]Not corrected for growth and decay formation of ^{225}Ra.

Spontaneous-fission counting of all of SHE's isolated by the separation methods has given negative or ambiguous results. After the samples were in electronic counters for 6 months, no more than three SF events have been observed in any single sample from Schemes I and II, and some samples show none or only one SF count. These rates approximate the background rates determined for our ionization counters. The fission-fragment energies recorded from five events from samples of eka-Au and eka-Hg were between 24 and 80 MeV; these energies were too low to define the source of these SF events. Of the five SF events observed by coincidence counting of fragments from a nonvolatile SHE fraction (Scheme III), one event was found to have a total kinetic energy (TKE) of 215 MeV. This is an unusually high energy considering that the average TKE for ^{252}Cf is 186 MeV,[18] but it is not too high to be excluded from the TKE distribution for such a nuclide. Thus, all of our results provide no persuasive evidence for the presence of SHE's.

By taking the observed SF events in any sample as the maximum number of SHE atoms that were made and later decayed, we have calculated upper limits for the cross sections to form them. These limits are a function of the assumed half-life, because saturation during the bombardment would occur for short half-lives, and not all atoms would decay for half-lives comparable to, or longer than, the counting period. We give these cross-section limits, corrected for chemical yield, as

$$\bar{\sigma} \leq \begin{cases} 2.0\times 10^{-34}\ \text{cm}^2 \text{ for } t_{1/2} = 10^4\text{–}10^5, \\ 2.5\times 10^{-35}\ \text{cm}^2 \text{ for } t_{1/2} = 10^5\text{–}10^7, \\ 2.0\times 10^{-34}\ \text{cm}^2 \text{ for } t_{1/2} = 10^7\text{–}10^8. \end{cases}$$

These results show that the elements 110 to 116 are not made within the observational range of our experiments: Either their formation cross sections are very meager or their half-lives are too short. Although the reactions ^{208}Pb(^{48}Ca, $2n$)^{254}No[19] and ^{206}Pb(^{48}Ca, $2n$)^{252}No[4] have been demonstrated to proceed with a relatively large cross section of approximately 1 μb, we expect greater losses from prompt fission in forming the compound nucleus ^{296}[116] because of its higher atomic number and the higher excitation energy deposited. However, the extent of these increased losses is difficult to evaluate without more information.

Estimates of the SF half-lives for superheavy nuclei are exceedingly uncertain; for example, those for ^{298}[114] stretch from less than 1 yr[6] to 10^{19} yr.[5] Only the longer, more optimistic estimates[5,7] would have predicted that the SHE nuclei we may have produced could survive to the time of detection. Our failure to find these nuclides then points to the possibility that the SF half-lives of the SHE's made in the reaction ^{248}Cm + ^{48}Ca may be considerably shorter than some predictions. These uncertainties concerning half-lives, cross sections, and chemical properties are not unexpected because of the long theoretical extrapolations necessary to provide information about this completely unknown region. Our search for SHE's has served to narrow the boundaries on some of these uncertainties, and we expect to reduce these bounds further by rapid new physical and chemical methods being devised to isolate and detect SHE's shortly after their formation.

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Core-Excited High-Spin Isomers in ^{212}Rn

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We have observed isomeric states in ^{212}Rn with very high angular momenta (up to $J^{\pi}=30^{+}$). The four isomers of highest spin contain core-excited neutron configurations, the spins of which are strongly aligned with those of the valence protons. The isomerism appears to result from a lowering of the excitation energies by 1 to 2 MeV compared to a sum of the unperturbed values for the constituent configurations.

Recently, mechanisms for producing isomers with very high spins (yrast traps) have been discussed. When such isomers exist, they are particularly useful in the study of nuclear structure at high angular momentum. These isomers might be caused by minima in shell-correction terms[1] overcoming the steeply rising liquid-drop energy, and may involve either triaxial or oblate equilibrium shapes. Additional effects would arise from the large residual interaction between states near the Fermi surface with maximum overlap of the nucleonic wave functions by alignment (MONA) of single-particle angular momenta.[2] Such isomers are expected to be oblate.

In the present work, we have found nine isomeric states in ^{212}Rn, and of those, the higher-lying levels appear to be of MONA origin. The experiments were carried out with the Chalk River Model MP tandem accelerator, and the results are summarized in Fig. 1. The five lower-lying isomers up to $J^{\pi}=17^{-}$ are accounted for[3] by configurations of the four valence protons in ^{212}Rn, but above 6 MeV of excitation, the most appropriate model to explain the isomers appears to be coupling of valence protons to excited states of the ^{208}Pb core. Since the Pauli principle would inhibit excitation of core protons into valence orbitals at spins already occupied by valence protons, it is suggested that mainly the neutron part of the ^{208}Pb excited states contribute to the coupling. We shall discuss the experiments leading to the level assignments, including the magnetic-moment measurements with which we compare the calculated moments for our proposed configurations. Conclusions about core-coupled isomers may be drawn from the proposed configurations.

The heavy-ion reaction ^{204}Hg(^{13}C, $5n$) at $E(^{13}\mathrm{C})=72$–85 MeV was used to produce ^{212}Rn at high angular momentum. The lifetimes of the isomeric levels and their sequence in ^{212}Rn were established by standard pulsed-beam techniques.[4] The data obtained include excitation functions of delayed and prompt γ rays, time distributions of delayed γ rays, and extensive γ-γ coincidence measurements. We have also determined concurrently angular distributions and linear polarizations of delayed γ rays. A Compton polarimeter consisting of three Ge(Li) detectors similar to the one described by Butler *et al.*[5] was used for the polarization measurements. A simultaneous fit of the angular distribution coefficients, A_2 and A_4, and of the linear polarization, P_γ, uniquely determined the angular momentum change, ΔJ, the multipolarity, λ, and the electric or magnetic character of each transition, as well as the mixing ratio, δ, though only the 1047-keV ($18^{-}\rightarrow 17^{-}$) transition was significantly mixed: $\delta(\langle E2\rangle/\langle M1\rangle)$