Attempts to Produce Superheavy Elements by Fusion of ^{48}Ca with ^{248}Cm in the Bombarding Energy Range of 4.5–5.2 MeV/u

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A search for superheavy elements was made in bombardments of ^{248}Cm with ^{48}Ca ions performed at projectile energies close to the interaction barrier in order to keep the excitation energy of the compound nucleus $Z = 116$, $A = 296$ as low as possible. No evidence for superheavy nuclei was obtained in a half-life region from 1 μs to 10 yr with a production cross section greater than 10^{-34} to 10^{-35} cm^2.

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Since all the known heaviest elements have been synthesized first by nuclear fusion, this process was also chosen for the production of superheavy elements, which are expected to occur around the predicted nuclear shell closures at atomic number 114 and neutron number 184. Although a large number of projectile and target combinations have been used in such attempts, no evidence for the formation of superheavy elements has been obtained. It is now obvious[1] that most of these systems are not suitable for this purpose. Either the resulting compound nuclei have an extremely low chance of survival because their fission barriers are too low, or the formation of a compound nucleus is hindered by dynamic effects and cannot occur at projectile energies close to the Coulomb barrier. There are only a few target-projectile combinations which have both favorable fusion and survival probabilities. The reaction of ^{48}Ca projectiles with a ^{248}Cm target is potentially one of these cases.

This system has been studied extensively without positive results.[2] However, in the earlier experiments the interaction energy exceeded the calculated fusion barrier by about 20 MeV. This extra energy was thought to be necessary in order to overcome entrance-channel limitations in the fusion process. As a consequence, the compound nucleus was produced with an excitation energy of at least 45 MeV, which probably reduced its surviving fraction to an undetectable level. More recent work on fusion reactions of heavy systems gives evidence, however, that the dynamic hindrance for the ^{48}Ca + ^{248}Cm system is small,[3] so that it should be possible to form the compound nucleus $^{296}_{116}X$ at a lower excitation energy.

We have carried out a new search for superheavy elements from the reaction of ^{48}Ca projectiles with ^{248}Cm at energies much closer to the Coulomb barrier,

which was calculated to be 235 MeV (0.96× proximity potential), corresponding to a bombarding energy of 4.90 MeV/u. In this series of experiments performed first at the SuperHILAC at the Lawrence Berkeley Laboratory (LBL) and then at the UNILAC at the Gesellschaft für Schwerionenforschung (GSI), Darmstadt, the excitation energy of the compound nucleus varied between 16 and 40 MeV. A variety of improved physical and chemical techniques were used for the isolation and detection of superheavy nuclei to increase the sensitivity relative to the earlier experiments.[2] Recoil-fragment separators, the small-angle separator system (SASSY) at LBL, and the separator for heavy-ion reaction products (SHIP) at GSI, were used to search for short-lived nuclides, and several different radiochemical techniques were applied in the region of longer half-lives. Thus, we are able to cover with high detection sensitivity a range of superheavy nuclide half-lives of 14 orders of magnitude, from 1 μs to 10 yr.

Separation in gas-filled magnetic fields[4] has been adapted at LBL to separate fusion evaporation residues from the bombarding beam. The separation time is the time-of-flight through the system ($\simeq 1$ μs). The many charge-changing collisions experienced by each particle in the gas give the evaporation residues a well-defined magnetic rigidity which is different from that for the projectiles and other reaction products and thus allows a magnetic separation.

The average magnetic deflection does not vary more than 0.2% for an energy change of 10% of the heavy evaporation residues. The LBL gas-filled separator SASSY is described in Ref. 5. Its overall detection efficiency was about 15% for the experiments performed. The suppression of the primary beam was sufficient to allow the use of implantation techniques. An array of position-sensitive surface-barrier detectors registered the implanted recoils and their subsequent disintegration by α decay or fission. ^{248}Cm targets of 400 μg/cm^2 on 250 μg/cm^2 Ti backings were bombarded with ^{48}Ca beams with an average current of 6×10^{11} ions/s. Irradiations were performed at 4.47, 4.83, and 4.93 MeV/u with 1.6×10^{16}, 7×10^{16}, and 0.8×10^{16} particles, respectively. A background fission rate of 4 per hour restricted the usable correlation times between fusion recoils and spontaneous fissions to a few minutes.

In the time range of 5 μs to 1 min no correlated high-energy α particles or out-of-beam fission events were found. The cross section for the production of a fissioning superheavy nucleus in the 4.83-MeV/u irradiation (Fig. 1, curve 1) is less than 3×10^{-34} cm^2.

The experimental technique using SHIP was the same as that which was successfully used in the identification of the heaviest elements; 107, 108, and 109.[6] Heavy evaporation residues from complete-fusion recoil from the target are separated in flight from the

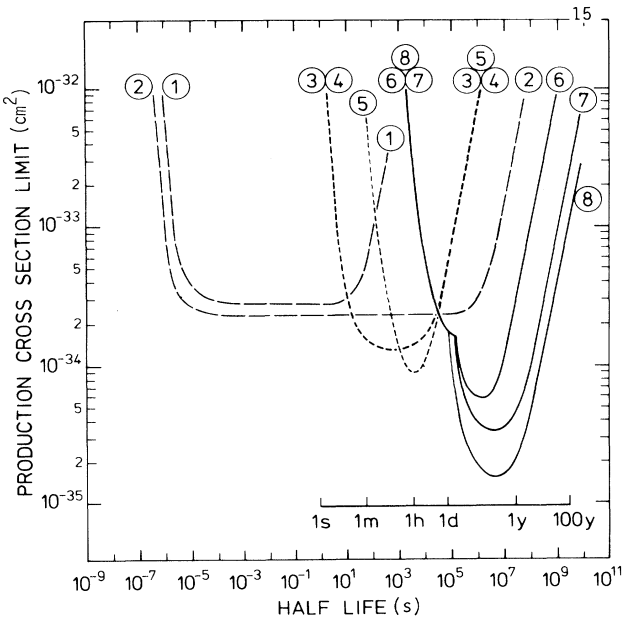

FIG. 1. Upper limits (at 95% confidence level) for the production cross sections of superheavy nuclei in the reaction of ^{48}Ca with ^{248}Cm in the energy range 4.5–5.2 MeV/u as a function of the half-life. The data result from experiments with recoil-fragment separators (curves 1 and 2), with fast on-line chemical separations (3, 4, and 5), and with off-line chemistry (6 and 7), as is pointed out in the text.

projectiles and other nuclear reaction products by the velocity filter SHIP.[7] After a flight time of 2.4 μs, they pass a time-of-flight detector and are finally implanted into an array of position-sensitive surface-barrier detectors. From the implantation energy and time of flight their masses can be determined with a resolution of 10% full width at half maximum. Decay sequences of single implanted atoms can be established by measuring energy, position, and time for each event in the detector array.[8] The calculated detection efficiency for evaporation residues is 12%. Since the ions are implanted close to the detector surface, about 45% of the emitted α particles may escape the detector. Events separated by more than 0.5 μs could be resolved.

The ^{248}Cm target (400 μg/cm^2 on 250 μg/cm^2 Ti) was bombarded with ^{48}Ca beams of 5×10^{11} ions/s average current. Five energies were chosen: 4.69, 4.84, 4.99, 5.08, and 5.24 MeV/u with doses of 4.0×10^{16} and 5.0×10^{16} particles for the lowest and highest and 11×10^{16} for the other three energies. The data were analyzed for α decays above 8 MeV correlated to recoils in the mass-number range from 271 to 321. The upper time limit for true correlations was determined by decays above 8 MeV in the α spectrum and by a background from targetlike recoils in the mass spectrum centered near mass 248 with a tail ex-

tending beyond mass 271. For short-lived nuclei decaying during the 5-ms beam bursts, we found no correlated events within the specified mass and decay-energy range. For decays observed between beam pulses, we could open the time window up to 300 s until we observed the first correlation, with a high probability of its being random. No evidence for decay chains from superheavy atoms was found. In addition, we searched for spontaneous fission decay. For a true fission fragment we required a minimum decay energy of 100 MeV for events in the stop detector and 150 MeV for coincidences between stop and additional detectors where the full total kinetic energy of both fragments is registered. We did not observe such an event during the whole experiment including a background measurement of 23 d after the irradiations. The cross section limits are 2.4×10^{-34} cm^2 for spontaneous fission decay (Fig. 1, curve 2) and 4.4×10^{-34} cm^2 for α decay in the three main irradiations. For the highest and lowest irradiation energies, cross-section limits are larger by a factor of 3.

In the radiochemical search experiments, standard off-line chemical techniques were used for longer-lived species, and fast on-line separations permitted access to nuclides with half-lives down to seconds. Three different approaches were followed, based on predicted chemical properties of superheavy elements: (i) Elements 112 through 116 are believed to be volatile in their elemental states at temperatures up to 1000 °C (Pb like)[9]; (ii) some elements such as 112 or 114 may even be gaseous at room temperature (Rn like)[10]; (iii) elements 108 through 116 are thought to form strong anionic bromide complexes in aqueous solution (Pt like).[11] All radiochemical experiments were carried out with thick curium-oxide targets (1.7 mg/cm^2 ^{248}Cm electroplated on 2.4 mg/cm^2 Be backing), so that the projectiles were slowed down by 0.37 MeV/u in the target.

Three bombardments with an incident energy of 4.98 MeV/u were performed at the SuperHILAC. The reaction products recoiling out of the target were collected in a copper catcher foil which was then chemically processed off-line. First, volatile Pb-like elements were evaporated from the Cu foil by heating it in a closed quartz apparatus in a H_2 stream at about 1000 °C. Provisions were also made to condense Rn-like noble gases into a cryogenic chamber.[12] After this volatilization, the Cu foil was dissolved in a HBr/Br_2 solution to form bromide complexes and the Pt-like elements were passed through a cation exchange column. Final samples from the Pb- and Pt-like chemistry were prepared on thin palladium or graphite foils, respectively, and inspected for α and spontaneous fission activities. In two bombardments with 1.2×10^{16} and 1.1×10^{16} particles, the half-life region between hours and days was inspected, and after a bombardment with 10×10^{16} particles the Pb- and Pt-like samples were measured for nearly two years in a low-background system[13] to extend the covered half-life range to several years. This system registers single and coincident fission fragments, their kinetic energies, and the number of neutrons per fission using surface-barrier detector pairs surrounded by a ^{3}He neutron counter. Neither fragment-fragment-neutron nor fragment-fragment coincidences were observed; three fragment-neutron coincidence signals occurred in the Pt-like fraction.[14] The Rn-like fraction was counted[12] over a period of 79 d for coincident fission fragments without positive results. The upper cross-section limits are shown in Fig. 1 by curve 6 for Rn-like, curve 7 for Pt-like, and curve 8 for Pb-like elements. These values are based on overall efficiencies, chemical plus counting, of 18% for Rn-like and 52% for Pb-like and Pt-like elements.

In the experiments performed at the UNILAC at 5.17 MeV/u incident energy, on-line versions[15] of these procedures were applied with use of a helium-KCl gas jet for transportation of the reaction products within $\simeq 2$ s from the target to the chemistry setups. (i) The on-line gas chromatography apparatus (OLGA)[15] was used for the continuous evaporation of volatile Pb-like species at about 1000 °C and their condensation onto cooled palladium foils mounted on a wheel system. This wheel was rotated stepwise between pairs of surface-barrier detectors in order to count the deposited activity for spontaneously fissioning nuclides. (ii) The cryogenic system[12] coupled to the gas-jet transportation capillary was used to detect short-lived Rn-like species. (iii) The minicomputer-controlled automatic rapid chemistry apparatus (ARCA)[15] was used to search for short-lived Pt-like nuclides. Here, reaction products carried by the gas jet were collected for 30 min on a glass filter frit. In a cyclic mode of operation, after each collection period the deposited activities were dissolved in HBr/Br_2 and treated as described in the preceding paragraph. Again, no evidence for spontaneously fissioning superheavy elements was obtained. The upper cross-section limits are given in Fig. 1 by curves 3 and 4 for experiments with OLGA and the cryogenic system and by curve 5 for experiments with ARCA. These limits are based on overall efficiencies of 9% (curves 3 and 4) and 16% (curve 5), respectively.

In summary, although a broad range of half-lives, 10^{-6} to 10^8 s, and excitation energies, 16 to 40 MeV, has been examined, no evidence for the formation of superheavy elements with cross sections of 10^{-34} to 10^{-35} cm^2 or greater was found. The existence of spherical superheavy nuclei near the compound nucleus ^{296}116 has been predicted by different theoretical approaches. Calculations[16] predict the fission barriers of the heaviest isotopes of elements from fermium to

element 109 to within 0.5 MeV. Similar calculations yield a fission barrier for $^{296}116$ of 5.6 ± 1 MeV.[17] The time range covered in our experiments is equivalent to a 3 MeV uncertainty in the fission barrier, and thus allows one to compensate for the uncertainty in the calculated ground-state shell corrections. The half-lives of isotopes of superheavy elements should fall somewhere in this time range. A rapid disappearance of the shell stabilization with increasing excitation energy has been observed for spherically shell-stabilized nuclei around ^{216}Th.[18] This thermal hindrance may prevent the production of superheavy elements in the reaction chosen. Perhaps the attainment of a compound nucleus with a neutron number closer to 184 would stabilize the evaporation residues to the point where superheavy elements could be synthesized and identified.

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