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RECENT SEARCHES FOR SUPERHEAVY ELEMENTS IN DEEP-INELASTIC REACTIONS

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Abstract - New attempts have been made to synthesize superheavy elements (SHE) by nuclear reactions that may possibly form the products at low excitation energies. Survival of the superheavy elements would then be enhanced because of reduced losses from prompt fission. Classical and diffusion model calculations of deep-inelastic reactions indicate there should be detectable yields of SHE formed with less than 30 MeV of excitation energy. Accordingly, superheavy elements have been sought in such reactions where targets of ^{248}Cm and ^{238}U have been irradiated with ^{136}Xe and ^{238}U ions. In the most recent experiments, targets of ^{248}Cm metal (3.5-7 mg-cm^{-2}) were bombarded with 1.8-GeV ^{238}U ions from the UNILAC accelerator. The longer-lived SHE's and actinides near the target Z were chemically separated and the yields of a number of isotopes of Bk, Cf, Es, and Fm were measured. An upper limit of 30 nb was obtained for the formation of 1-h ^{259}No. In addition, to the off-line chemical recovery and search for SHE's, we performed an on-line experiment to detect volatile SHE's with half lives of a minute or more. All experiments to produce and detect superheavy elements were much less than optimum because of premature failures in the Cm-metal targets. The outcome and status of these experiments, and the implications of the actinide yields in estimating the chances for forming superheavy elements in the ^{248}Cm + ^{238}U reactions are discussed.

The transfer of many nucleons together with little excitation energy in damped collisions seems an extremely attractive method of synthesizing superheavy elements and neutron-rich actinides (Ref. 1 & 2). For the products to survive prompt fission, the net diffusion of energy to the heavy fragment during the collision must be lower than 20 to 30 MeV. Thus, damped collisions accompanied by large mass transfer appears to be the only feasible way to accomplish this, since no compound-nucleus reaction to form elements around Z = 114 leads to anything less than 30-40 MeV of excitation energy.

On the basis of availability and the favorable cross sections predicted (Ref. 3), we selected the reaction of ^{238}U ions with ^{248}Cm target nuclei as the optimum combination that we could currently employ to produce superheavy elements (SHE). Accordingly, in two months of periodic U bombardments at GSI, we have used this reaction for the first time to search for SHE's.

In addition, we have bombarded a target of ^{248}Cm with ^{136}Xe in collaboration with our colleagues at the Lawrence Berkeley Laboratory (LBL) (Ref. 4). One of the major purposes of these bombardments was to determine the extent of nucleon-transfer to the target as the projectiles Z and A increased. We could then compare these results with our previous actinide yields measured for transfer reactions in collisions of ^{48}Ca with ^{248}Cm (Ref. 5) and with the yields from the ^{238}U + ^{248}Cm reaction. Of primary concern was the survival probability of the highly fissionable transcurium isotopes as a function of mass, angular momentum, and energy transfer by the projectile. Because of energy transfer and resultant nuclear excitation, nearly all collisions cause either prompt or sequential fission. Most of the heavy products that survive are formed in the low-energy tails of the energy-loss distribution (Ref. 6 & 7). However, due to the mass and energy balance required in nuclear reactions, it is possible for processes accompanied by large mass and energy transfer to lead to products in relatively low excitation states if Q_{gg} is sufficiently negative (Ref. 8). Among the projectiles studied, ^{48}Ca has the lowest Q_{gg} which, therefore, reduces the energy available for dissipation and excitation.

Thus, by studying the production of highly-fissionable, nearby actinides by transfer reaction, we had hoped to shed some light on forming superheavy elements by the same reaction

process. The superheavy elements are expected to be similar to the heavier actinides with respect to the magnitude of their fission barriers. If so, they would survive their birth to about the same extent as the actinides providing the same risks prevailed, i.e., the same excitation and angular momentum. In turn, the influence on survival probability of varying these parameters could be roughly evaluated by comparing the yields of the same actinide isotopes produced in bombardments with light, medium, and very heavy ions (^{48}Ca, ^{136}Xe, and ^{238}U).

With increasing mass of the projectile, the deep-inelastic or transfer reaction has been found to become the dominant mode of nuclear interactions (Ref. 9). Yields of the primary products before fission should increase with the heavier projectiles like ^{238}U. This was demonstrated by the very strong enhancement in the production of Cm, Cf, and Es isotopes when ^{238}U was used to bombard ^{238}U as compared to using ^{136}Xe (Ref. 6). The actinide yields for these two projectiles are compared in Fig. 1. The factors of 10 to 100 increase in actinide production cross sections with ^{238}U ions were particularly persuasive in leading us to attempt the synthesis of SHE's in the ^{238}U + ^{248}Cm reaction.

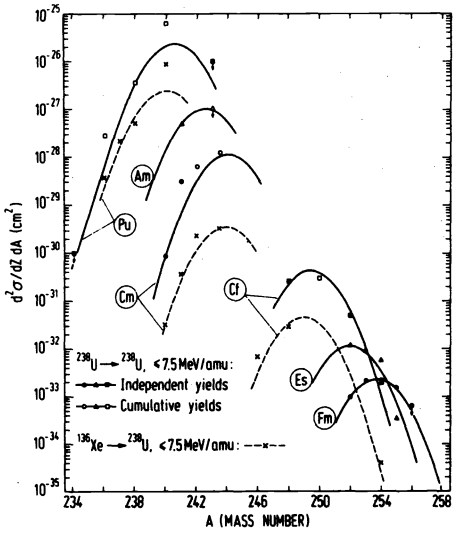

Fig. 1. Cross sections for the formation of heavy actinides in the reactions of 7.5-MeV/u ^{136}Xe and ^{238}U projectiles with ^{238}U targets (Ref. 6).

At the SuperHILAC we bombarded ^{248}Cm with ^{136}Xe at an energy of 1.2 B_C (lab) and at the UNILAC we used nearly the same ^{238}U energy relative to the Coulomb barrier. In the earlier ^{48}Ca bombardments of ^{248}Cm, the comparable ratio E/B_C was 1.1 which is similar enough to allow a meaningful comparison of actinide formation cross sections with those from U + Cm and Xe + Cm reactions. All experiments used ^{248}Cm targets sufficiently thick to reduce the projectile energy to slightly below the Coulomb barrier. Products formed in the bombardment recoiled from the targets and were collected on foils or thick Cu discs placed close behind the target. The actinides (and SHE) were chemically separated into elemental fractions which were then assayed for alpha and spontaneous fission activities over a period of months.

Before the first bombardments of ^{248}Cm with U ions occurred, a large developmental effort was undertaken. Bombardments of actinide targets with intense ^{238}U beams had never been attempted before and it was anticipated that the loss of 660 MeV of energy in foils that were no greater than 20 μm thick would present some exceedingly difficult problems. Foremost is the one of rapid heat removal, since a 1-μA beam of U^{62+} ions deposits about 0.2 joules in the target during each 5-ms beam pulse. The pulse rate is 50 Hz. In addition to their being kept from melting, the target foils must be totally protected from air or oxygen because of the chemical reactivity of Cm. An entirely new target system as illustrated in Fig. 2 was designed, built, and tested, and Cm metal targets were developed and produced for the first time (Ref. 10). Curium metal vaporized onto thin (3 to 4 μm) substrate foils was considered necessary if we were to obtain suitable heat-transfer rates, because Cm in other chemical forms (Cm_2O_3, CmF_3) is a thermal insulator.

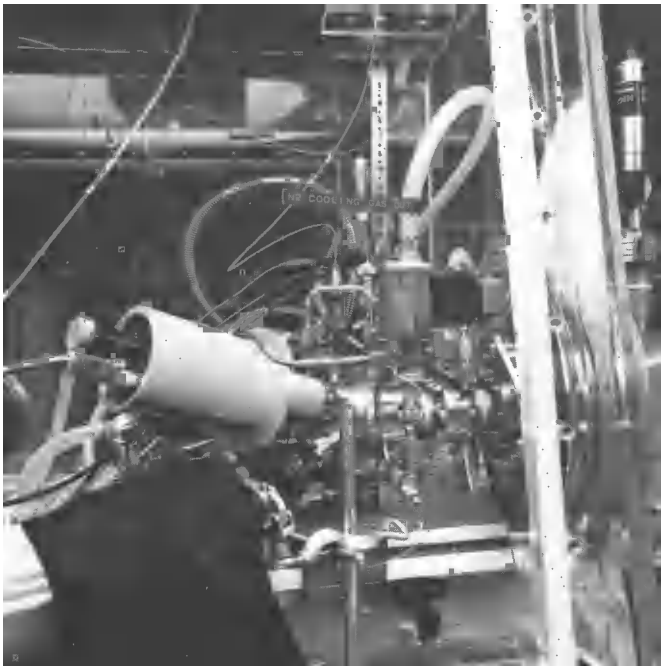

Fig. 2. Target system in an enclosure and attached to the UNILAC beam line at position Y-3. The infrared and television monitors for the target are shown in the left side of the photo.

The major results for these various projectile combinations with ^{248}Cm targets are shown in Fig. 3 where the production cross section for the isotopes of Cf, Es, Fm, and Md can be compared with those from the ^{48}Ca + ^{248}Cm transfer reactions. Yields for isotopes with atomic numbers less than Cf are not shown. A comparison of these results with those obtained from Xe + U and U + U reactions show one striking feature. The factor of 100 enhancement in yields of Cm isotopes found in U + U vs. Xe + U was not observed in the Fm yields when ^{248}Cm was bombarded with these same projectiles. Both cases represent a (4pxn) transfer; yet, the yield enhancement factor was only 5 or less for ^{238}U reactions with ^{248}Cm. In a qualitative sense, we can understand the reduced Fm yields on the basis of the considerably lower fission barriers in the primary Fm products compared to those in the primary Cm products from U + U. The Fm chain is depleted by fission during deexcitation to a much larger extent than the Cm chain formed at the same excitation energy. However, when excitation energies for the mass equilibrated primary products (A_Z) are extracted, we find (Table 1) an inversion in the ratio E*(Xe)/E*(U) between U and Cm targets. These are calculated from Q_{gg}, the actual projectile energies, and the assumptions that all the energy above the Coulomb energy of the outgoing fragments is available for dissipation into collective and nucleon motions and that this energy is equilibrated according to the masses of the fragment. Hence, in the reactions with U targets, U ions favored considerably less average excitation energy in the ^{248}Cm product than did ^{136}Xe ions in forming ^{246}Cm. Furthermore, it is known from a measurement of fragment kinetic energies from U + U reactions that the fragments emerge with kinetic energies greater than the Coulomb energies, which reduces the energy available (Table 1) for dissipation (Ref. 11). The calculated excitation energies for Xe and U interactions with ^{248}Cm targets are nearly the same which implies the same survival probability and could possibly account for the poor enhancement factor in Fm yields from the U bombardments.

TABLE 1. Excitation energy of the most-probable primary fragment (A_Z) formed in (4pxn) transfer reactions with ^{238}U and ^{248}Cm targets. These energies were derived from actual projectile energies, Q_{gg}, and the assumptions noted in the text.

Target nuclide	Projectile	Maximum E/A (lab) MeV/u	Primary (4pxn) product	$-Q_{gg}$ MeV	Excitation energy, E* MeV
^{238}U	^{48}Ca	8.4	^{245}Cm	38	114
	^{136}Xe	7.5	^{246}Cm	18	113
	^{238}U	7.5	^{248}Cm	2	73
^{248}Cm	^{48}Ca	5.6	^{255}Fm	41	10
	^{136}Xe	6.7	^{256}Fm	21	63
	^{238}U	7.4	^{258}Fm	5	62

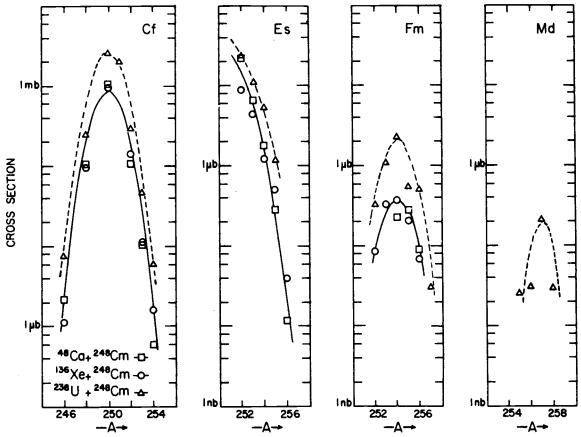

Fig. 3. Cross sections for the production of Cf, Es, Fm, and Md isotopes in the reactions of ^{136}Xe and ^{238}U projectiles with ^{248}Cm targets. Shown for comparison are similar results for ^{48}Ca + ^{248}Cm.

Another important conclusion is that production cross sections from ^{48}Ca reactions with Cm are not markedly improved with Xe or U induced transfer reactions. The fact that there are only small differences in the production of a given isotope made by different projectiles is probably due to a balance between increased mass transfer probability with increasing mass of the projectile and a concurrent decrease in survivability because of an increase in excitation energy. It should be noted from Table 1 that ^{48}Ca, because of a large $-Q_{gg}$, leads to a very low excitation energy in ^{255}Fm. Therefore, while the nucleon transfer probability will be less than with U projectiles, the chance for fission during de-excitation is reduced.

The excitation energies calculated in Table 1 are based on the assumption of attaining an equilibrium during the contact time in the collision, which is equivalent to sharing the kinetic energy of the projectile equally among all nucleons in the resultant light and heavy fragments. A distribution of excitation energies around this average (or a lower average) occurs and only those heavy fragments survive fission that were formed in the low-energy tails of such distributions. We could estimate the region in the distribution where the products we observe were formed if the primary mass distribution for a given atomic number were known. This primary distribution can be approximated by the minimum-potential energy or mass-equilibration model and appropriately depleted by Γ_n/Γ_f to reproduce the measured yield distribution for each element. With the projectiles used to bombard ^{248}Cm to form Cf, Es, and Fm, we find that an average of 3-4 neutrons are emitted from the primary fragment in U + Cm reactions and ~1 neutron in ^{48}Ca + ^{248}Cm collisions. The first value implies excitation energies of 30 to 40 MeV in the heavy fragment that survived birth in U + Cm collisions.

The above result has serious consequences with regard to forming SHE at sufficiently low excitation energies to minimize fission competition during the de-excitation process. To form SHE in U + Cm reactions, many more nucleons and, therefore, more energy, is likely to be transferred than in producing Fm isotopes. This may result in the "tails" of the dissipated energy distributjon moving upward and out of any survival window so that no observable production of SHE would occur. Furthermore, the 30-40 MeV of excitation energy noted for the Fm isotopes formed in U + Cm reactions is not any less than the excitation energy of SHE producible by several complete fusion reactions such as $^{48}Ca + ^{248}Cm$.

Misleading conclusions can be drawn from cross sections measured for a given isotope made by different target-projectile combinations for the reasons that varying amounts of energy are available for dissipation and the A/Z ratio of the projectiles are not constant. Both the widths and centroids of the primary mass distributions depend on the degree of energy damping and the extent of mass-to-charge equilibration (Ref. 12). More meaningful are comparisons of the primary distributions themselves, but reconstruction of these requires detailed measurement of cross sections for the projectile-like fragment as a function of energy loss. To date there exists no such information for heavy-ion/curium systems.

Special efforts had been made in the U and Xe bombardments of ^{248}Cm to detect ^{259}No because it is the product most distant from the Cm target that could feasibly be chemically isolated and identified and, thus, was an excellent test of many-nucleon transfer. None was found with upper limits of 10 nb in Xe + Cm reactions and 30 nb in U + Cm collisions. However, ^{256}Md was detected and measured in the U + Cm, but not in the Xe + Cm bombardments. Because the ^{259}No cross section limits are no greater than the production cross sections known for other nuclear reactions, our hopes of synthesizing heavy, neutron-rich actinides by deep-inelastic reaction have been nearly extinguished.

Our attempts to detect SHE's produced in the bombardments of ^{248}Cm with ^{238}U ions are reported by G. Herrmann in a companion paper of this symposium. Our sensitivity for detecting SHE's was much less than optimum because of the premature failures of the ^{248}Cm-metal targets. Sufficiently intense U beams could not be tolerated by the targets and we were then unable to detect superheavy elements if their formation cross sections were in the range of 10^{-34} to 10^{-35} cm^2, as anticipated from diffusion-model calculations (Ref. 3). Hence, the deep-inelastic transfer mechanism to produce SHE has not yet been disproven, inasmuch as the target limitations in these experiments did not allow a fair test.

The question of why the Cm-metal targets failed is especially important if these experiments to discover superheavy elements are to continue. Accordingly, an extensive metallurgical examination has been made of an unirradiated target and of two irradiated targets of Gd metal (an atomic homolog and stand-in for Cm). The two irradiated Gd targets were from a group of five that were bombarded until they failed with 2.1-GeV U ions from the UNILAC accelerator at GSI. The failure mode was identical to that of the four Cm-metal targets subsequently destroyed during the experiments to synthesize SHE's. Nearly every target failed after a few hours of irradiation despite all attempts to purify the rare gases in contact with the target surface. Valleys of the order of 2-mm long would form and grow deeper until a stress crack would open in the bottom (Fig. 4). An examination of the failed Gd targets by scanning electron microscopy (Fig. 5), microprobe x-ray fluorescence, and microphotography revealed that a strong mechanical bond exists between the Gd metal and the substrate foils. It also appeared that the Gd metal reached a much higher temperature during the U-ion bombardments than the Mo substrate. This information led to the conclusion that target failure occurred because of differential heating and, therefore, expansion of the two metals which produced excessive stresses along the weakest points (rolling lines) of the Mo substrate. The 50-Hz pulse rate of the accelerator induced very many heating and cooling cycles which caused work hardening and stresses that the metals were unable to withstand.

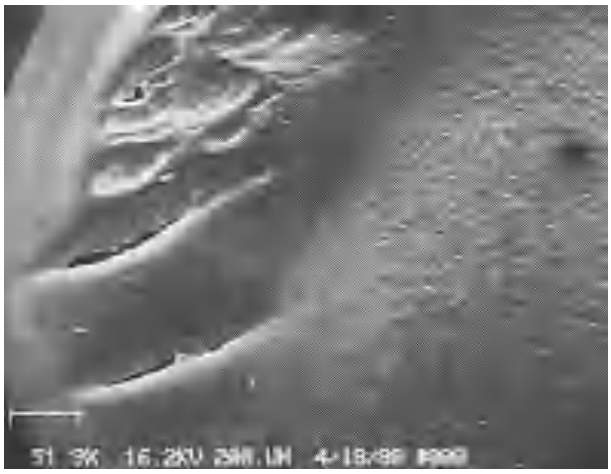

Fig. 4. Scanning electron-microscope (SEM) photo of a Gd target destroyed by 2.1-GeV U^{62+} ions. Photo is from the Gd side of the target and shows deep depressions that opened into cracks with continued irradiation from the U beam.

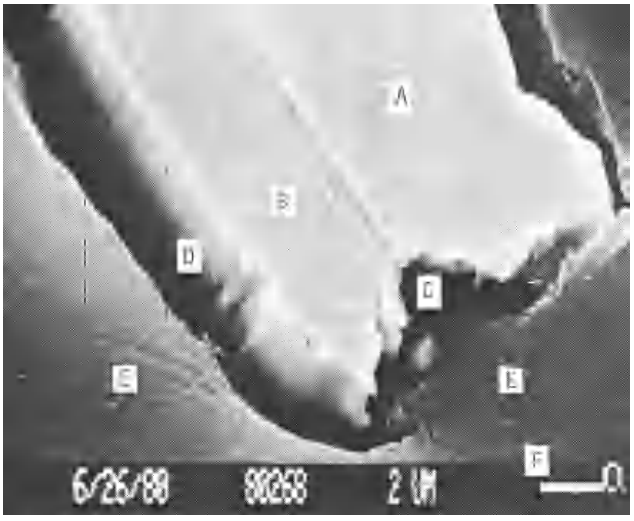

Fig. 5. Cross sectional view of a Gd target that failed during U-ion bombardment. (A) Gd metal; (B) Mo substrate; (C) stress fracture of target; (D) bottom of Mo foil; (E) epoxy mounting; (F) 2-μm fiducial line.

In July of this year, new tests of targets of Gd metal vapor deposited upon substrates of Ta and annealed Mo foils were made by bombardment with U ions. Failure in five of these test targets occurred after being irradiated with only ~5 x 10^{14} particles, which is an integrated flux 50 times less than desired for satisfactory experiments to produce SHE's. Clearly, metallurgical improvements in the targets are pivotal before highly sensitive experiments to produce and detect superheavy elements can be pursued again.

Although the limits we had hoped to set for SHE synthesis fell short of our expectations, we have obtained important information on the production cross sections of nearby actinide isotopes. Our interpretation of these yields bears directly on the problem of estimating the probability of forming superheavy elements because the reaction mechanism for producing the actinides is the same as for synthesizing SHE. Earlier in this report we provided our interpretation of several features in the actinide yields. However, a major conclusion was that, contrary to expectations, a many-nucleon transfer reaction was no more likely to form products in low excitation states than a complete fusion reaction. Specifically, the probability of transferring a large number of nucleons and <30 MeV of excitation energy may be too small to offer appreciable yields of product nuclei in ^{238}U + ^{248}Cm collisions. However, in the context of producing SHE's, this finding may not be fatal because transfer reactions

are capable of reaching areas in the "Island of Stability" which are inaccessible to fusion reactions. The areas attainable in complete-fusion reactions lie on the neutron-deficient side of the "Island" where the fission barriers are expected to be the lowest and, therefore, the chances of surviving their formation in a highly excited state the least. We have reached a crossroad in the quest for superheavy elements and have concluded that if the experimental limitations can be overcome, the potential of the deep-inelastic transfer reaction to produce SHE's should be explored to the edge of our current limits of detection of about 10^{-35} cm^2.

In conclusion, it should be recognized that none of the attempts, including this one, to synthesize and detect superheavy elements have seriously challenged the theory upon which their proposed existence is based. The most detailed estimate of the fission and alpha barriers in the "Island of Stability" (Ref. 13) indicate the half lives for the ground state nuclei producible in all nuclear reactions so far tried are less than the experimental limits of detection. For example, in the $^{238}U + ^{248}Cm$ reaction where the mass-equilibrated primary product for Z = 114 is $^{295}114$, formed with an assumed excitation energy of 40 MeV, de-excitation would lead to the emission of four neutrons to yield $^{291}114$. The spontaneous fission half life predicted for this nuclide (Ref. 13) is within the range of 0.3 s to 5 min or considerably shorter than the day or longer limits established through chemical separations of SHE (see companion paper by G. Herrmann in this symposium). Aside from another question concerning their formation probabilities, it is clear from half-life considerations alone that adequate tests of the theoretical expectations for superheavy elements must involve much more rapid methods for detecting their decay. The techniques for the rapid (to a μs) identification of spontaneous fission nuclides produced by complete fusion reactions are currently available, but they have not been developed for recoil products coming from deep-inelastic transfer reactions. The pathway in the search for superheavy elements must ultimately entail the development of fast, on-line methods for the conclusive identification of superheavy elements with half lives in the microsecond region. Although such an effort may be exceedingly difficult and costly, the impact of their discovery on nearly every field of science would surely repay the investment.

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