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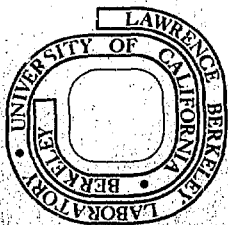
NEW EXPERIMENTAL INSIGHTS INTO THE PRODUCTION OF
SUPERHEAVY ELEMENTS USING HEAVY ION REACTIONS

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NEW SUPERHEAVY ELEMENTS USING HEAVY ION REACTIONS

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ABSTRACT

Attempts at the Lawrence Berkeley Laboratory to produce superheavy elements (SHE) using the reactions of ^{48}Ca with ^{248}Cm and ^{136}Xe with ^{248}Pu have been unsuccessful. These negative results have led us to consider the possibility that these reactions (and their associated mechanisms) do not lead to the formation of superheavy nuclei with sufficiently low excitation energies. Consequently, the number of surviving atoms of superheavy elements is well below our detection thresholds even with using relatively low estimates for prompt fission losses. A summary of some new experimental evidence, supporting the above hypothesis, is presented and possible alternative heavy ion heavy-target combinations are suggested for SHE synthesis.

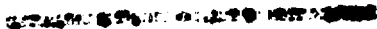
1. INTRODUCTION

Efforts to synthesize and identify superheavy elements (SHE) using heavy ion reactions have been unsuccessful up to the present time. Two different approaches that have been used at the SuperHILAC include the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction [1,2] and the $^{136}\text{Xe} + ^{248}\text{Pu}$ reaction [3]. The first of these reactions

relies on compound nucleus formation, while the second reaction requires the transfer of approximately 50 nucleons from the projectile to the uranium target nucleus in a deep inelastic transfer reaction (DIT). Radioanalytical studies of the above reactions or similar reactions have been pursued in the hope of either finding a suitable reaction pathway to the formation of superheavy elements or eliminating unproductive heavy-ion heavy-target combinations.

We would like to consider several possible reasons why SHE's have not been synthesized and/or identified using heavy ion reactions. One possibility is that none of the reactions used so far provide a reaction pathway leading to the formation of a nucleus having atomic number and mass number within the predicted region of stability known as the island of stability. A second possibility is that "SHE" nuclei are formed in the nuclear reactions but the excitation energies and angular momenta of the SHE nuclei are large and therefore there is no stability provided by the relatively small ground state fission barriers of these nuclei. A third and related possibility is that the true fission barriers for nuclei in the predicted island of stability are in fact closer to the smallest predicted values [4,5] and consequently the spontaneous fission half-lives are very short with respect to the experimental detection time scales used to date.

A fourth possibility that would obscure the identification of the SHE's is the probability of a non-distinctive character for the spontaneous fission process of relatively long lived SHE's. Unless superheavy elements have fission properties that are distinguishable from the fission properties of the known actinide elements, they will remain difficult, perhaps nearly impossible, to identify on the basis of a few events. Such a lack of distinctive fission characteristics in such features as total kinetic energy of fission, neutron multiplicity, fission mass asymmetry, or fission product multiplicity (ternary, quaternary, etc.) could have contributed to the negative results obtained so



far. With respect to this fourth possibility, Kalpakchieva et al. [6] have interpreted results of correlated fragment mass distribution measurements for the $^{40}\text{Ar} + ^{243}\text{Am}$ reaction as possible evidence for the existence of highly mass asymmetric fission of the compound nucleus ($^{283}113$) and they have attributed the observed asymmetry ($A_H/A_L \approx 2.5$) to the preferential formation of a heavy fragment near the doubly magic ^{208}Pb region. Because of the important implications of such distinctive fission properties (i.e., a low total kinetic energy release in fission combined with a unique and easily distinguishable fission energy and mass asymmetry) further experimental evidence was sought and, in contradiction to this interpretation, suggests an alternative explanation for their observations [7].

A final possible reason for the negative results in the search for SHE's is the uncertainty in the assumed chemical properties. This reason is a less likely possibility than the others, because the predicted chemical properties are less uncertain than the predicted nuclear properties and also because a wide range of chemical properties have been utilized in the experimental SHE searches.

In this paper we summarize some experimental results related to the first four possibilities mentioned above.

II. COMPOUND NUCLEAR REACTIONS

A. Competing Deep Inelastic Transfer Process

Although there has been a considerable amount of effort made in the study of heavy ion complete fusion reactions there has been surprisingly less attention given to the important $^{40}\text{Ar} + ^{238}\text{U}$ system. This is in part due to the fact that the evaporation residue ($^{278-xn}110$) has been predicted to have essentially no stability [4,8]. In one of the first studies of this reaction Sikkeland [9] reported a fission excitation function with a threshold of 200 MeV (171 MeV c.m.),

based on the observation of two coincident fragments from the $^{40}\text{Ar} + ^{238}\text{U}$ reaction. It was assumed that all of the binary events associated with full momentum transfer were the result of complete fusion-fission and this was taken as evidence for compound nucleus formation [10]. The similar interpretations of further counter studies by Hanappe et al. [11] and [12,13] have also been based on the full momentum transfer assumption. Oganessian et al. [14] studied the excitation function for the production of gold isotopes in the $^{40}\text{Ar} + ^{238}\text{U}$ reaction, and also obtained a threshold of ~ 200 MeV (171 MeV c.m.) for the production of these products which were again ascribed to the fusion-fission reaction mechanism by assuming that the gold yields were part of a broad symmetric Gaussian-shaped fission product mass distribution. A similar study by Bruchertseifer et al. [15] was also done for the $^{48}\text{Ca} + ^{238}\text{U}$ reaction.

Kratz et al. [16] used radioanalytical techniques to study the mass distribution from the reaction of 288 MeV ^{40}Ar with a thick ^{238}U target. The results of this study [16] in fact showed that approximately 50% of the total reaction cross section resulted in a broad symmetric distribution of products centered at approximately one-half the mass of the compound nucleus. The mass distribution obtained by Kratz et al. is shown in Fig. 1. A contribution from deep inelastic transfer (DIT) was known to exist for the $^{40}\text{Ar} + ^{232}\text{Th}$ reaction [17] but appeared to constitute only a small part of the total reaction cross section in the $^{40}\text{Ar} + ^{238}\text{U}$ reaction [16]. However, studies of ^{40}Ar ions with Au [18-22] and Ag [23] targets showed that it was very difficult to distinguish between products arising from deep inelastic transfer reactions and those products arising from the complete fusion process on the basis of fragment mass and energy distributions alone. In fact the trends in the angular distributions of the light projectile-like products measured as a function of the atomic number of the product have led to a view of the reaction as a dynamical diffusion process in which the formation of a compound nucleus does not need

to be invoked and where "complete fusion" occurs only in the limit at the completion of the diffusion process [24]. Thus such a deep inelastic diffusion process producing a wide range of products may lead to broad symmetric mass distributions centered at half the mass of the composite system [23]. This new and developing view of the deep inelastic reaction process provides ample reason for doubting previously drawn conclusions concerning complete fusion and compound nucleus formation in the $^{40}\text{Ar} + ^{238}\text{U}$ reaction and hence also for similar heavy-ion heavy-target reactions.

A recently developed differential recoil range method [25] can be employed to further test this broad role of the deep inelastic transfer process in the production of a wide range of products from bombardments with ^{40}Ar and similar ions. This method has been used to deduce the general shapes of angular distributions of products ranging from approximately one-half the mass of the compound composite system to products near the target [7]. These recoil range distributions from the reaction of 250 MeV ^{40}Ar with ^{238}U were correlated with a trend in the angular distribution as a function of ΔZ similar to the trend observed in the $^{40}\text{Ar} + ^{197}\text{Au}$ reaction [18-22]. (As indicated above, in the case of the $^{40}\text{Ar} + ^{197}\text{Au}$ reaction this is a trend that has been interpreted as evidence for viewing the deep inelastic reaction mechanism as a dynamical diffusion process.) Complete fusion is ruled out for products with backward or forward peaked angular distribution since the $1/\sin\theta$ type angular distribution is expected for such a process.

Figure 2A shows the experimental recoil range distributions for the Hg(Tl) products from the two reactions $^{48}\text{Ca} + ^{238}\text{U}$ and $^{40}\text{Ar} + ^{238}\text{U}$, corresponding to three different excitation energies [7]. Calculated recoil range distributions for the Hg(Tl) products from these three experiments are shown in Fig. 2B, and are based on three different assumptions about the heavy product angular distribution.

These three functional forms are shown in Fig. 2C. In this study [7] the Hg(Tl) recoil data corresponded most closely to the predictions for the simple backward peaked angular distributions (corresponding to a forward peaked projectile-like fragment angular distribution); however, there was a small discrepancy at the longer ranges indicating the possibility of a small $(1/\sin\theta)$ contribution. To test such an effect a backward peaked angular distribution was used and mixed with a 10% $(1/\sin\theta)$ contribution (actually $1/(\sin\theta + 0.01)$). The experimental results, shown in Fig. 2A, fell between the calculated distributions (shown in Fig. 2B) for a backward peaked angular distribution (solid line, Fig. 2C) and the backward peaked plus $1/\sin\theta$ angular distribution (dotted line, Fig. 2C) and showed that 90% or more of these products are formed in a deep inelastic reaction process.

These data indicated that non-complete fusion (and non-compound nuclear) processes accounted for an unexpectedly large portion of the mass distribution of the $^{48}\text{Ca} + ^{238}\text{U}$ reaction and, of the broad symmetric, previously labeled "fusion-fission" mass distribution of the $^{40}\text{Ar} + ^{238}\text{U}$ reaction [16]. Again, we can see that earlier work on the $^{40}\text{Ar} + ^{238}\text{U}$ system may have overestimated the cross section due to complete fusion processes. Thus we conclude that the use of ^{48}Ca as a projectile with heavy targets, considered a hopeful approach for the production of SHE's, must result in a much smaller production of compound nuclei than had been anticipated.

B. Do Superheavy Elements Fission Asymmetrically?

A very mass asymmetric fission mode has been predicted for SHE's on theoretical grounds by Sandulescu and Greiner [26]. The results of an $^{40}\text{Ar} + ^{243}\text{Am}$ study have been taken as possible evidence for such an effect by Kalpakchieva et al.[6]. At the bombarding energy of 222 MeV corresponding to a compound

nuclear excitation energy of 48 MeV the authors of this study found a double peaked mass distribution associated with full momentum transfer to have a very large mass asymmetry ($A_H/A_L \approx 2.5$) with one of the peaks being near the doubly magic lead region. At higher energies the mass distributions were symmetric and peaked at approximately one-half the mass of the compound nucleus. It is interesting to note that in a preliminary study of the mass distribution of products produced from the reaction of ≤ 300 MeV ^{48}Ca with a thick ^{238}U target we have found a high yield of products in the lead region that should not be attributed to the complete fusion process. A possible interpretation of the relatively few measured yields in the $^{48}\text{Ca} + ^{238}\text{U}$ reaction is shown in Fig. 3. The appearance of the comparatively large "gold finger" (component G) is explained in a similar way as in the ^{84}Kr and $^{136}\text{Xe} + ^{238}\text{U}$ reactions [27,28]. These products represent the remnant of the target-like deep inelastic component not lost to fission. The deduced angular distribution from the recoil range studies of these products in the $^{48}\text{Ca} + ^{238}\text{U}$ and $^{40}\text{Ar} + ^{238}\text{U}$ reaction [7] discussed above, also suggest this interpretation (see Fig. 2).

Since we see little difference between the mechanism of production of nuclei expected in our $^{40}\text{Ar} + ^{238}\text{U}$ and $^{48}\text{Ca} + ^{238}\text{U}$ study and that in the work of Kalpakchieva et al. [6], we might predict that a careful study of the angular distribution trends as a function of Z for fragments in the Pb region, would reveal backward peaked angular distributions for the $^{40}\text{Ar} + ^{243}\text{Am}$ reaction. Such an observation would rule out a complete fusion-fission process leading to the observed mass distribution asymmetry. Perhaps the results of Kalpakchieva et al. [6] could be taken as strong evidence for shell stabilization effects in the deep inelastic process rather than evidence for asymmetric fission of a SHE compound nucleus system. Thus there is no evidence for marked asymmetry in the fission of SHE's, a distinctive property that could be extremely helpful in the search for these elements.

C. Relative Threshold for Complete Fusion and Quasielastic Transfer in the
 $^{40}\text{Ar} + ^{238}\text{U}$ Reaction

The evidence, indicating that the complete fusion process in the ^{40}Ar and ^{48}Ca reactions make up a smaller fraction of the total reaction cross section, σ_R , than previously thought does not eliminate the possibility that compound nucleus formation may still be a large enough fraction of σ_R (on the order of 5 to 10%) to allow production and detection of SHE's. However, the threshold for complete fusion in a reaction such as $^{48}\text{Ca} + ^{248}\text{Cm}$ is of great importance because this threshold will determine the minimum excitation energy achievable in the compound nucleus. The information in Fig. 4 suggests that a difference in the complete fusion threshold of ~ 12 MeV (corresponding roughly to the evaporation of one additional neutron) could easily be the difference between success or failure in the production and eventual detection of a superheavy element, if, for example, the ratio Γ_n/Γ_f has the average value of 10^{-4} . The Bass Model [29,30] and the Proximity Force Model [31] predict complete fusion thresholds for the $^{40}\text{Ar} + ^{238}\text{U}$ reaction and the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction that are 10 to 15 MeV higher than the interaction barrier, where Bass [29,30] defines this interaction barrier energy as corresponding to the potential energy at the "interaction" separation distance where the target and projectile are within the range of their mutual nuclear forces. Quasielastic transfer processes can be expected to occur at the interaction barrier where the resultant of the coulomb plus nuclear forces is still repulsive and complete fusion is not possible.

Oganessian et al. [14,15] have reported complete fusion thresholds for $^{40}\text{Ar} + ^{238}\text{U}$ and $^{48}\text{Ca} + ^{238}\text{U}$ that have been based on the excitation function for the production of Au isotopes in these reactions. However, the results discussed in the previous two sections indicate that these products are part of a deep inelastic process, at least at the higher bombarding energies.

Oganessian et al [14,15] have argued that the broad Gaussian nature of the isotopic distribution of the gold products supports the assumption that these products are formed in a complete fusion-fission process. Figure 5 shows the measured isotopic distribution of gold products for the reaction of ≤ 400 MeV ^{48}Ca with ^{238}U , again taken from our preliminary study of this reaction. The distribution of these products do in fact agree very well with the results of a statistical fission and neutron evaporation calculation shown as a solid line in Fig. 5. In addition a relative shift of the most probable gold isotope to a more neutron-deficient product can be seen for this data when compared to the results of Bruchertseifer et al (dashed line and solid triangles) [15]. The observed difference of approximately 4 neutrons between the peaks is just what is expected for the difference in the effective bombarding energies. However, this type of agreement with a statistical model, interpreted in light of the recoil range data [7] previously discussed, can result from deep inelastic processes as well as from complete fusion-fission. (Note, also, that this supports the idea that charge to mass and energy equilibration occurs more rapidly than mass equilibration in the deep inelastic reaction.)

In order to investigate the behavior of complete fusion as a function of bombarding energy in the $^{40}\text{Ar} + ^{238}\text{U}$ reaction Saint Simon et al [32] have used a method sensitive to all competing reaction channels: the radiochemical measurement of the production cross section of iodine isotopes. By using an iterative procedure to correct for growth and decay along the isobaric mass chains it was possible to obtain the independent yield cross sections shown in Fig. 6. These cross sections correspond to the iodine isotopic distributions for the reaction of ^{40}Ar ions with energies varying from 212 to 340 MeV impinging on thick ^{238}U targets. The isotopic distributions show a two or three humped structure. This structure was explained as primarily due to the superposition of two iodine isotopic distributions from two independent processes, complete

fusion (CF) and quasielastic transfer (QET), both followed by fission. The neutron rich distributions shown as a dashed and dot-dashed line were shown to be formed in QET-induced fission of target-like products having excitation energies in the region of 10-20 MeV. The isotopic distributions shown as solid lines were shown to be from fission of the compound nucleus system $^{278}_{(110)}$ and a fit to these product distributions was calculated using a statistical fission model and the "Overlaid Alice" evaporation code to determine the evaporation of neutrons from the fragments. However, there appeared to be an enhancement of iodine yields around $A \approx 130$ that could not be entirely accounted for by QET or CF induced fission.

These enhanced yields come from the fission of uranium-like nuclei produced in deep inelastic transfer (DIT) reactions. Furthermore, since there is a continuous trend in excitation energy and mass transfer between the QET and DIT processes it is not possible to make a meaningful separation into distinct distributions for the two mechanisms.

Figure 6 shows that the independent yield production cross sections for iodine isotopes from QET-induced fission and CF-induced fission have different behavior at the energies close to the interaction barrier of 200 MeV. From further analysis of these data Fig. 7 was obtained.

In Fig. 7 it can be seen that the complete fusion threshold is at least 12 to 15 (c.m.) higher than the barrier for QET-induced fission which was assumed to be the interaction barrier at 171 MeV (c.m.). This result is to be compared with predicted differences of 13 MeV (c.m.) and 9 MeV (c.m.) for the Bass Model [29,30] and for the Proximity Force Model [31] respectively. The corresponding differences predicted for the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction are 14 MeV (c.m.) (Bass Model) and 10 MeV (c.m.) (Proximity Force Model). The results of Saint Simon et al. [32] suggest that these are conservative lower limits. Thus, they conclude that there is evidence supporting these theoretical models

and that an additional energy of at least 10 to 14 MeV (c.m.) above the interaction barrier is required in the reaction of $^{48}\text{Ca} + ^{248}\text{Cm}$ to produce fusion. Moreover it has been found [33] that by using the "Overlaid Alice" code [34] with "realistic fission barriers" [8] for the superheavy elements, that fission losses in the SHE region increase even more rapidly than might be expected with additional excitation energy because of the rapid drop of the fission barriers of the SHE's as neutrons are evaporated, and the neutron-deficient edge of the island of stability is approached.

Thus the increase in excitation energy of the compound nucleus resulting from the higher threshold for complete fusion of the projectile and target nuclei, augments the difficulty of producing superheavy elements in bombardments utilizing projectiles such as ^{48}Ca .

D. Alternative Complete Fusion Routes?

Radioanalytical studies of heavy-ion projectiles with uranium targets have shown that the fraction of the total reaction cross section going into complete fusion-fission rapidly decreases as ions heavier than ^{40}Ar are used. This effect is dramatically illustrated in Fig. 8 [35] where the percent of the total reaction cross section versus the projectile atomic number is plotted for the three reaction channels, complete fusion-fission (CF), deep inelastic transfer (DIT), and quasielastic transfer (QET). The measured cross sections as well as the parameters B/E_{eff} for each system are contained in Table I. Justification for this comparison is based on the similar B/E_{eff} values for these reactions although the relative cross section ratios for the QET and DIT reactions (particularly for U+U) may not have too much significance. The fraction of the total reaction cross section going into complete fusion

TABLE 1. Thick target cross sections for Z_{HI} plus ^{238}U .

System	E_{lab}	B/E_{eff}	σ_{OET} (mb)	σ_{DI} (mb)	σ_{CF} (mb)	Ref.
$^{40}\text{Ar} + ^{238}\text{U}$	288	0.828	400 ± 120	100 ± 50	620 ± 160	[16]
$^{56}\text{Fe} + ^{238}\text{U}$	538	0.755	810 ± 160	350 ± 55	190 ± 30	[36]
$^{84}\text{Kr} + ^{238}\text{U}$	605	0.859	700 ± 120	470 ± 70	55 ± 15	[27]
$^{136}\text{Xe} + ^{238}\text{U}$	1150	0.800	600	600 ± 125	$\ll 1$	[28]
$^{238}\text{U} + ^{238}\text{U}$	1785	0.874	610	365	$\ll 1$	[37]

is probably even less than indicated, since first, a reevaluation by Kratz et al. [27] of the Kr+U results indicates that complete fusion accounts for less than 4% of the total reaction cross section and secondly, the recoil range distribution data for the $^{40}\text{Ar} + ^{238}\text{U}$ reaction [7] shows a large deep inelastic contribution to the production of products previously attributed to complete fusion-fission.

Thus it is clear from these types of observations that SHE's cannot be produced in a very heavy-ion heavy-target complete fusion-fission reaction where the SHE nuclei are part of the fission mass distribution. An alternative complete fusion reaction with a tightly bound, and somewhat heavier ion (than ^{48}Ca) and lighter target (than ^{248}Cm) such as $^{56}_{26}\text{Fe} + ^{226}_{88}\text{Ra} \rightarrow (^{282}_{114})$, may in fact give a very low minimum excitation energy ($E^* = 18$ MeV) but the fact that the neutron number (168) is far removed from the island of stability makes such reactions less favorable than the $^{48}\text{Ca} + ^{248}\text{Cm}$ combination.

III. DEEP INELASTIC TRANSFER REACTION

A. The $^{136}\text{Xe} + ^{238}\text{U}$ Reaction and Upper Limit Cross Sections for the Production of ^{212}Pb and ^{211}At in the $^{136}\text{Xe} + ^{160}\text{Gd}$ Reaction

The reaction of $^{160}\text{Gd}(^{136}\text{Xe}; ^{84}\text{Kr}, n's)^{212}\text{Pb}$ requires the transfer of 18 protons and 34 neutrons to ^{160}Gd from the ^{136}Xe projectile. This is the number of protons and neutrons required in a transfer from ^{136}Xe to ^{238}U to make ($^{290}\text{110}$) which is predicted to be in the "island of stability." Using a radiochemical separation procedure, an upper limit of $2 \times 10^{-34} \text{ cm}^2$ was observed for production of ^{212}Pb in the reaction of 1150 MeV ^{136}Xe with a thick natural Gd target (21.9% ^{160}Gd) [3]. The upper limit cross section for the reaction $^{160}\text{Gd}(^{136}\text{Xe}, ^{84}\text{Kr}, n's)^{212}\text{Pb}$ is therefore $1 \times 10^{-33} \text{ cm}^2$ or one nanobarn (1 nb). The one nanobarn limit was applied to a similar or greater number of nucleons transferred from ^{136}Xe to ^{238}U by assuming that less than 10% of the Pb fragments fissioned and that the nucleon diffusion rates and the interaction times are nearly the same for the $^{136}\text{Xe} + ^{160}\text{Gd}$ and $^{136}\text{Xe} + ^{238}\text{U}$ reactions. It was pointed out that this limit is consistent with the theoretical prediction that the cross section for transfer of ~ 60 nucleons in the Xe+U reaction is about 1 nanobarn [38]. It was noted, however, that the transfer of 34 neutrons and 18 protons represents a higher n/p ratio than found in ^{136}Xe (n/p = 1.52). It is possible that SHE's would be formed as a result of the transfer of neutrons and protons closer to the ratio n/p = 1.52, and perhaps they would be produced with higher cross sections than the limit set for ($^{290}\text{110}$). Although such nuclides would be more neutron-deficient and further removed from the n=184 closed shell, those with higher atomic number, such as ($^{290}\text{113}$) or ($^{293}\text{114}$) might still be within the island of stability and have half-lives suitable for detection. A test for the transfer of ~ 50 nucleons with such a ratio of n/p (≈ 1.52) was established by observing a limit of $6 \times 10^{-32} \text{ cm}^2$ for

the production of ^{211}At in the $^{136}\text{Xe} + ^{160}\text{Gd}$ reaction [3].

The probability of survival of an excited SHE nucleus can be estimated from the $[\Gamma_n/\Gamma_f]^x$ value which, due to an increasing value of x , is a decreasing function of the excitation energy. However, it is known from studies of Kr and Xe with Ho and Bi targets [38] that the probability for a given number of nucleon transfer increases with the projectile energy damped (converted) into internal excitation of the system. It is the balance of these two factors that will ultimately determine the probability for the production of SHE's. The average value of $[\Gamma_n/\Gamma_f]$ may actually be quite small (we use 10^{-4} as an example above in Section II.C), but in order to set the highest reasonable upper limit for the cross section for the production of SHE's from the $^{136}\text{Xe} + ^{238}\text{U}$ reaction [3] the value $[\Gamma_n/\Gamma_f]^4 = 4 \times 10^{-3}$ was used corresponding to the evaporation of four neutrons [39]. Based on this estimate of $[\Gamma_n/\Gamma_f]^4$ and an upper limit cross section of 1 nb for the production of ^{212}Pb in the $^{136}\text{Xe} + ^{160}\text{Gd}$ reaction, an argument [3] was given that an upper limit for the production of $(^{290}_{110})$ in the $^{136}\text{Xe} + ^{238}\text{U}$ reaction would be 10^{-36} to 10^{-35} cm^2 . It was therefore concluded that the reaction of $^{136}\text{Xe} + ^{238}\text{U}$ does not provide a good test for existence for the island of stability, even with beam intensities 10 to 100 times the levels obtainable at the SuperHILAC. The somewhat higher limits for ^{211}At production in the $^{136}\text{Xe} + \text{natGd}$ reaction would not change significantly the conclusion that the SHE's would be difficult to observe through production by the $^{136}\text{Xe} + ^{238}\text{U}$ reaction. The above arguments, summarized in Fig. 9, were based on the assumption that angular momentum did not significantly increase the fission probability of lead-like products in the $^{136}\text{Xe} + \text{natGd}$ reaction. However, appreciable fission of gold-like products following deep inelastic transfer in the reaction of ^{86}Kr and ^{136}Xe with ^{197}Au [40,41] has been observed. The results of these studies were interpreted as evidence for an increase in the fission probability due to

rotational angular momentum transferred to the gold-like products in the deep inelastic reaction process. Such a process might suggest that the low limit set for the production of ^{212}Pb in the $^{136}\text{Xe} + ^{160}\text{Gd}$ reaction was due to a relatively higher transfer probability combined with large losses of Pb-like products due to fission deexcitation enhanced by an angular momentum effect.

In order to estimate an upper limit cross section for the production of SHE's, using the upper limit for ^{212}Pb produced in the Xe + Gd reaction, it is only necessary to know the relative fission probability for SHE fragments and Pb fragments with similar excitation energy and angular momentum, consistent with the DIT process. Although these properties are not well known, we can expect the fission probability of the SHE's to increase more rapidly with angular momentum than would the Pb fragments. The upper limit cross section of 10^{-35} to 10^{-36} for SHE production in the $^{136}\text{Xe} + ^{238}\text{U}$ reaction is therefore valid for the non-zero angular momentum conditions also.

B. Alternative Multinucleon Transfer Reaction Combinations

Recent studies of the $^{238}\text{U} + ^{238}\text{U}$ reaction at GSI [42,37] show that, for a given average width in the charge dispersion (mass dispersion), the energy damped into internal excitation energy is less than in Xe transfer reactions [36,38]. Such an observation can be interpreted as supporting the idea that there should be significantly larger cross sections for the production of heavy transuranium elements in the reaction of U+U [42] than in the Xe+U reactions [43]. Evidence for such an effect can be seen by making a comparison of the yields of Cf and Es isotopes ($\Delta Z = 6$ and 7 respectively) from these two reactions, where the cross sections for the production of the more neutron-excessive isotopes are 10 to 10^2 times larger from the U+U reaction [42].

Such an effect suggests the use of a very heavy target such as ^{248}Cm , ^{249}Cf , ^{252}Cf , ^{254}Es , or ^{257}Fm with a heavy ion beam of ^{238}U (or possibly ^{197}Ar , ^{208}Pb , or ^{244}Pu) as a way to produce SHE's. Figure 10 shows a number of interesting transfer reactions. The ^{165}Ho and ^{248}Cm reaction shown first could be driven by the closed shell at $Z=50$. However, the diffusion process would probably favor symmetric division into two fragments near ^{208}Pb .

Based on the results of the U+U reaction [37] studies at GSI to produce ^{255}Fm , an analogous reaction of ^{238}U with ^{254}Es is written to suggest the possibility of transfer reactions to produce elements near the SHE region with reasonable cross sections. However, the formation of SHE's requires a transfer with a larger neutron to proton ratio as indicated in the reaction of ^{238}U with ^{257}Fm . It is important to note that the predicted stability of the products in the SHE region for these last two reactions varies from being unstable to having detectable half-lives [4,5]. Although such reactions would have many technical difficulties associated with them, these target-projectile combinations may provide a suitable reaction pathway to the formation of SHE's, not available in the reactions that have been used up to now.

IV. CONCLUSIONS

We have considered several possible reasons for negative results in the synthesis and identification of superheavy elements obtained at heavy-ion accelerator laboratories around the world. Due to the limited choice of targets above uranium, projectiles heavier than ^{40}Ar have been used. However, for projectile ions near and above the mass and charge of ^{40}Ar quasielastic transfer and deep inelastic transfer comprise a significant fraction of the total reaction cross section. The deep inelastic transfer reaction has many characteristics

that tend to obscure the observation of the complete fusion and compound nucleus-fission reactions. As a result, previous measurements of the fraction of the total reaction cross section corresponding to complete fusion can only be taken as upper limits. Furthermore, mass and energy distributions associated with binary events from heavy ion reactions (for example, $^{40}\text{Ar} + ^{238}\text{U}(16)$ or $^{40}\text{Ar} + ^{243}\text{Am}(6)$) in which complete fusion was assumed to occur, cannot be safely interpreted as corresponding to the compound nucleus.

For ions heavier than argon, complete fusion-fission rapidly decreases, eliminating the possibility for production of SHE fission fragments in such reactions as krypton, xenon or uranium with uranium. In spite of the larger contribution from deep inelastic transfer reactions, complete fusion and compound nucleus formation is expected to occur in the $^{48}\text{Ca} + ^{248}\text{Cm}$ reaction. However, the proximity potential model [31] and the Bass model [29,30] predict complete fusion thresholds 10 to 15 MeV higher than the interaction barrier and the work of Saint-Simon et al [32] using the similar reaction $^{40}\text{Ar} + ^{238}\text{U}$ provides experimental evidence for such an effect. As a result the minimum attainable excitation energy for the compound nucleus [$^{296}116$] probably results in large prompt fission losses putting the SHE production level below the experimental level of sensitivity.

The possibility remains that transfer reactions using very heavy ion projectiles such as ^{197}Au , ^{208}Pb , ^{238}U , or ^{244}Pu with ^{254}Es or ^{257}Fm targets could lead to the production of SHE nuclei at relatively low excitation energies. For the $^{136}\text{Xe} + ^{238}\text{U}$ reaction the probability of transferring the required number of protons and neutrons to reach the SHE region appears to be unacceptably low. The success of these experiments rely on the transfer reaction mechanisms and on an extrapolation of broad Gaussian distributions of primary products (before fission) around the target nucleus extending from the millibarn region

into the nanobarn region. Very little is known about the reliability of these extrapolations and, consequently, more radiochemical data are needed.

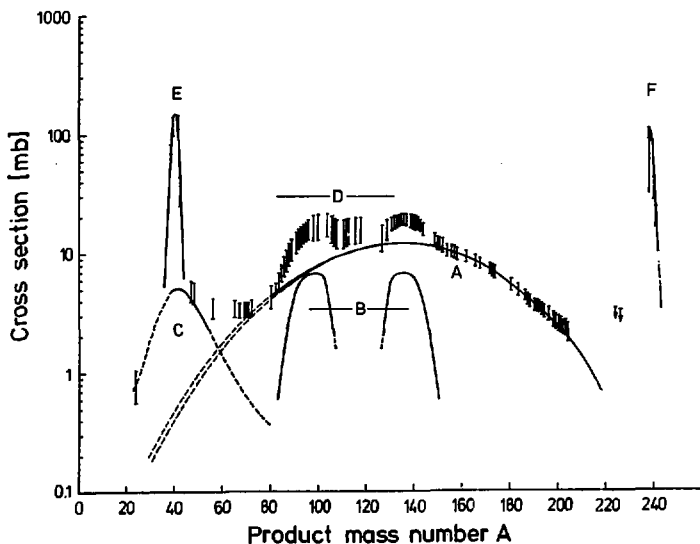
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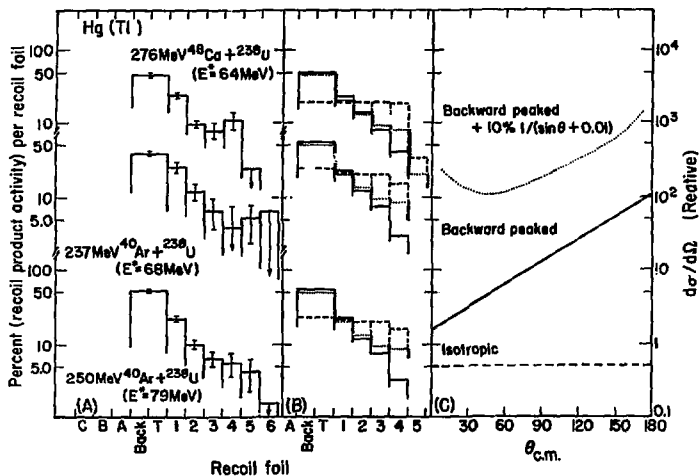
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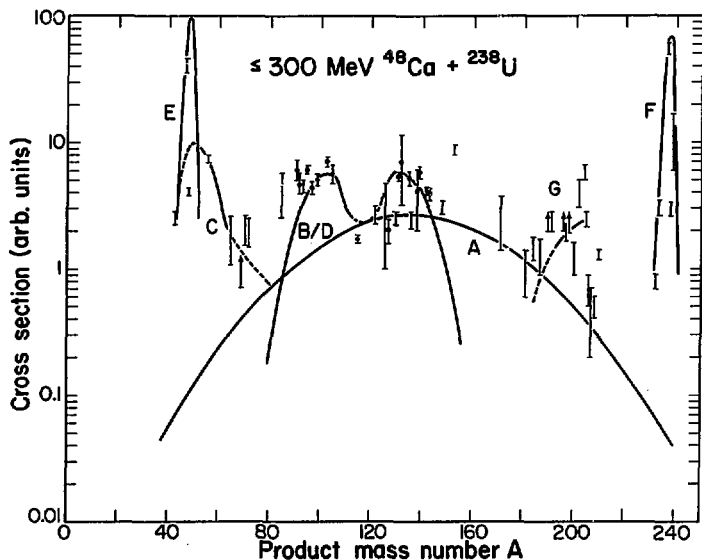
XBL 7511-8975

Fig. 1. Total integrated mass yields (upper and lower limits are indicated at those mass numbers for which experimental data were obtained) and their decomposition into individual components for the reaction of 288 MeV $^{40}\text{Ar} + ^{238}\text{U}$ (Kratz et al. [16]): (A) complete fusion-fission; (B) quasi-elastic transfer-induced fission; (C) deep inelastic transfer; (E) and (F) quasi-elastic transfer ("rabbit ears"). The existence of products from the sequential fission of heavy fragments formed from deep inelastic transfer (D) is also indicated; however, it was not possible to deduce a mass distribution for this component.



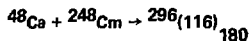
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Fig. 2. (A) The experimental differential recoil range distributions for Hg(Tl) products from the three reactions, 276 MeV $^{48}\text{Ca} + ^{238}\text{U}$ and 237 and 250 MeV $^{40}\text{Ar} + ^{238}\text{U}$. The excitation energies, E^* , for the compound nucleus system are also given. (B) The calculated recoil range distributions for the same reactions as shown in (A). The three calculated distributions shown in (B) for each of the Hg(Tl) distributions were calculated using the angular distributions in (C) denoted by the same type of line.



XBL 778-1702

Fig. 3. Mass yield distribution for the thick target reaction of 300 MeV $^{48}\text{Ca} + ^{238}\text{U}$. The experimental points shown with error bars represent isobaric yield cross sections. A possible interpretation of the data is shown (components A-E). Components E and F result from quasielastic transfer reactions. Component B/D is fit to the neutron excessive isobaric yields shown with solid circles through the error bars, and represents the quasi-elastic transfer and deep inelastic transfer-induced fission component. Component A represents an upper limit for the complete fusion-fission distribution. Component C represents the argon-like deep inelastic transfer distribution, and component G is the surviving remnant of the uranium-like deep inelastic transfer distribution.

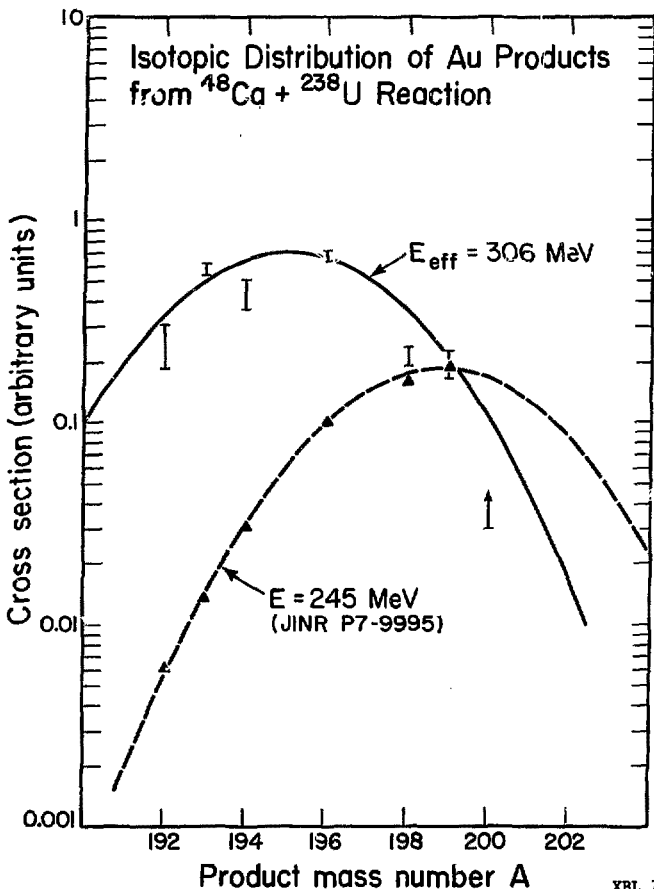


$$\sigma_{\text{ER}} = \sigma_{\text{CN}} \left[\frac{\Gamma_n}{\Gamma_n + \Gamma_f} \right]^x = 10^{-25} \left[10^{-4} \right]^x$$

${}^{48}\text{Ca}$ Energy (Lab)	Excitation Energy (E^*)	x neutrons	σ_{ER} (cm^2)
233 MeV (Coulomb barrier)	25 MeV	~2	$10^{-25} \cdot 10^{-8} = 10^{-33}$
255 MeV (Ave. Exp. Energy)	43 MeV	~3-4	$10^{-25} \cdot 10^{-12} = 10^{-37}$ $10^{-25} \cdot 10^{-16} = 10^{-41}$
$x = E^*/\epsilon_n$	$\epsilon_n \approx 12 \text{ MeV}$		

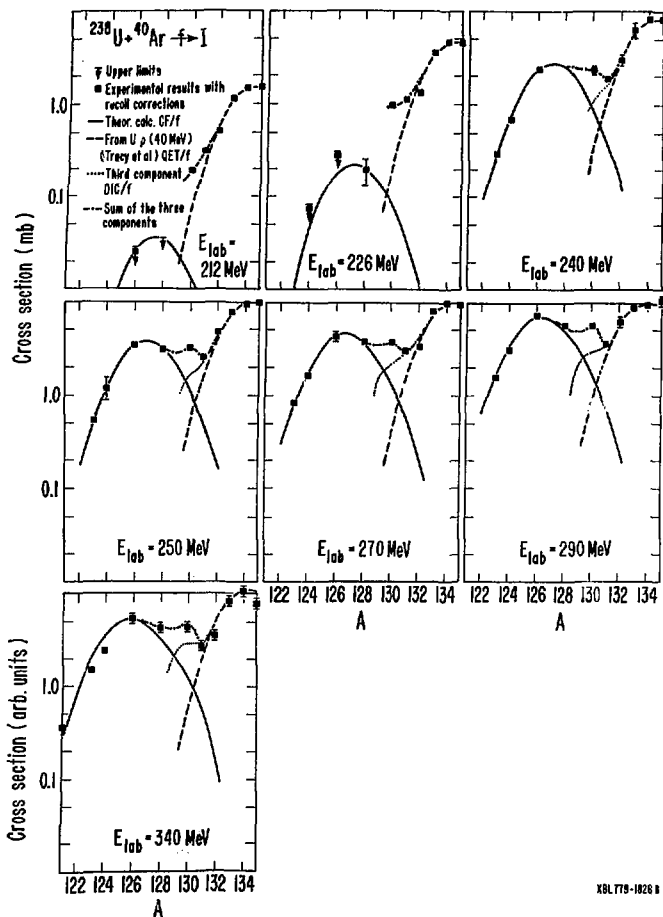
XBL 783-7522

Fig. 4. The relative importance of the threshold energy for fusion in the production of SHE's in the ${}^{48}\text{Ca} + {}^{248}\text{Cm}$ reaction. The Coulomb barrier was calculated using $R = 1.44(A_1^{1/3} + A_2^{1/3})$. The value of 255 MeV represents the average experimental ${}^{48}\text{Ca}$ energy used in searches using the SuperHILAC [1,2]. The number 10^{-4} is used as a reasonable example of an average value for $[\Gamma_n / (\Gamma_n + \Gamma_f)] \approx [\Gamma_n / \Gamma_f]$.



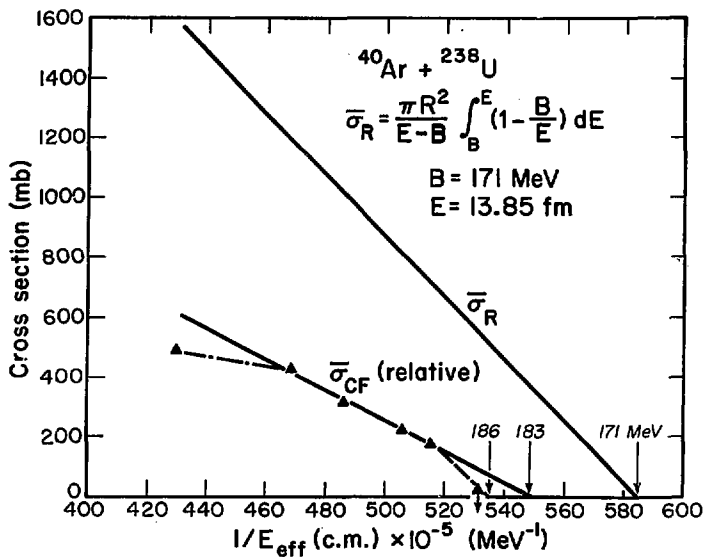
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Fig. 5. Independent yield distributions for gold isotopes from the thick target reaction of <400 MeV ^{48}Ca with ^{238}U ($E_{\text{eff}} = 306$ MeV) and 245 MeV ^{48}Ca with ^{238}U from Bruchertseifer et al.[15]. The solid line represents the results of a statistical fission and evaporation calculation for an effective bombarding energy of 306 MeV. The relative magnitude of the calculated distribution has been fit to the data. The dashed line is the result of a similar statistical calculation by Bruchertseifer et al.[15].



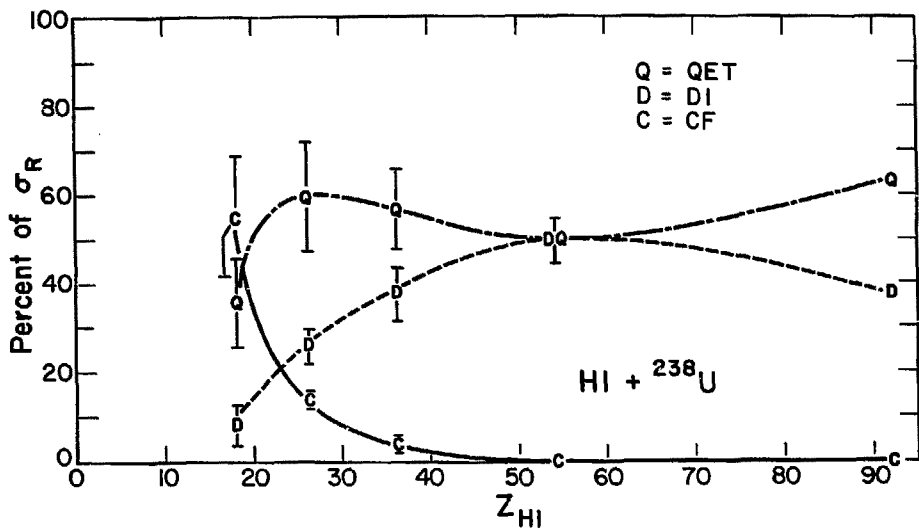
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Fig. 6. Independent yield cross sections for the production of iodine isotopes in the thick target reaction of ^{40}Ar with ^{238}U . The solid lines, dashed lines, dot-dashed lines and dotted lines are explained in the text.



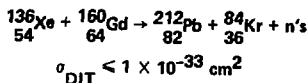
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Fig. 7. A plot of $\bar{\sigma}_R$ and $\bar{\sigma}_{CF}$ vs. E^{-1} where $\bar{\sigma}_R$ is the average total reaction cross section calculated by using the parameters indicated and $\bar{\sigma}_{CF}$ is the average complete fusion cross section. See Ref. 32 for further explanation of the effective bombarding energy, E_{eff} , and the solid, dashed, and dot-dashed lines.



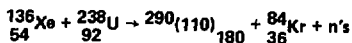
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Fig. 8. Plotted are the percent of the total reaction cross section for the three reaction channels, quasielastic transfer (QET), deep inelastic transfer (DIT), and complete fusion-fission (CF), as a function of projectile Z for the interaction of ^{40}Ar , ^{56}Fe , ^{84}Kr , ^{136}Xe , and ^{238}U with ^{238}U targets.



Transfer to target ($\Delta p = 18, \Delta n = 34$)

Loss by Fission?



$$\sigma_{SHE} < 1 \times 10^{-33} \left[\frac{\Gamma_n}{\Gamma_n + \Gamma_f} \right]^x < 1 \times 10^{-33} \times 4 \times 10^{-3}$$

$$\sigma_{SHE} \leq 4 \times 10^{-36} \text{ cm}^2$$

Fig. 9. An upper limit cross section for the production of SHE's in the ${}_{54}^{136}\text{Xe} + {}_{92}^{238}\text{U}$ reaction based on the experiments using the ${}_{54}^{136}\text{Xe} + {}_{64}^{160}\text{Gd}$ reaction. Values of $x=4$ and $[\Gamma_n/(\Gamma_n + \Gamma_f)]^4 = 4 \times 10^{-3}$ were used [3].

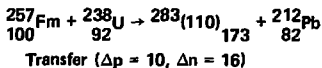
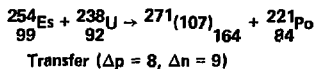
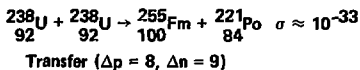
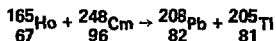
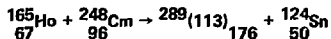


Fig. 10. Hypothesized heavy-ion transfer reactions. Only the U+U reaction has been shown to occur experimentally [37]. No designation of emission of neutrons has been indicated.