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How good are superheavy element *Z* and *A* assignments?

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Abstract. Proton number, Z, and mass number,A, assignments for newly discovered heavy element nuclides have historically been made by observing α decay to a daughter with well-established Z and A, and then observing the well-know decay of that daughter. For all of the new superheavy element isotopes observed in ^{48}Ca irradiations of actinide targets, this correlation technique has not been possible, because the α-decay chains end in spontaneous fission of previously unknown isotopes. Consequently, Z and A assignments have been made by less-direct means. The superheavy element Z and A assignment methods are summarized, and possibilities for how they may be incorrect are explored. While it is highly likely that most of the superheavy element Z and A assignments are correct, there is a real need for a direct proof.

1. History about heavy element *Z* and *A* assignments

For the discovery of elements 104–113 [1–12] proton number (Z) and mass number (A) assignments have been made by observing the α decay of the parent (new element) nucleus followed by the α decay of daughter nuclei with well-established Z and A. These daughter α decays have characteristic decay energies and half-lives. With the assumption of no intervening, unseen decays (such as electron-capture or β decay), the parent Z and A assignment was made by adding two to each of the $Z$and A of the first daughter. The assumption of no intervening decays could be supported by observation of a smooth trend in the α-decay energies, matching that expected by well-established α-decay systematics [13].

For all of the superheavy element (SHE) isotopes produced by ^{48}Ca irradiations of actinide targets [14, 15] the series of α decays (α-decay chains) terminate with spontaneous fission (SF) of the last chain member. The SF decay mode provides little information, other than half-life, to identify the Z and A of the decaying nucleus. SF lacks the characteristic decay energy seen in α decay. Thus, the Z and A of the α-decay chain members cannot be "anchored" by identification of the later chain member.

2. How SHE *Z* and *A* assignments have been made

For the SHE isotopes produced in ^{48}Ca irradiation of actinide targets, and all of the daughter isotopes resulting from successive α-decays, experimenters have resorted to less-direct means of Z and A assignment. These less-direct means are excitation functions, cross bombardments, and decay systematics.

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2.1 Excitation functions

An excitation function for formation of superheavy element isotopes by compound nucleus formation followed by de-excitation by evaporation of neutrons is a record of the production cross section for evaporation residue (EVR) formation as a function of projectile energy. Projectile energies are chosen from near the Coulomb barrier between projectile and target nuclei to a few tens of MeV above the barrier. The production cross sections are typically modelled as a three-step process,

$$\sigma_{EVR} = \sigma_{cap} \cdot P_{CN} \cdot P_x \cdot \prod_{i=1}^{x} \left(\Gamma_n / \Gamma_{tot}\right)_i, \tag{1}$$

where σ_{EVR} is the EVR production cross section. The first step of the process, represented by σ_{cap}, is the cross section for capture of the projectile and target nuclei into a minimum of their mutual Coulomb + nuclear potential to form a di-nuclear complex. The second step is the evolution this di-nuclear complex to an equilibrated compound nucleus, represented in Eq. (1) as P_{CN} (the probability for compound nucleus formation). The third and final step is the de-excitation of this excited compound nucleus by neutron evaporation in competition with fission. This third step is shown in Eq. (1) as the probability of evaporation of exactly x neutrons, P_x, times the product of the probabilities for emitting a neutron, rather than fissioning, in each of x neutron evaporation steps.

At the lowest projectile energies, σ_{EVR} is limited by the Coulomb barrier. Compound nucleus formation ($\sigma_{cap} \cdot P_{CN}$) increases monotonically from zero at far sub-barrier energies (although it may decrease at the highest projectile energies due to critical angular momentum effects). The CN excitation energy results from the center-of-mass kinetic energy and the mass difference of the target + projectile and the CN. The CN is "cooled" during each neutron evaporation step by an amount defined by the neutron separation energy and the kinetic energy of the neutron.

The end result for the formation of SHE in ^{48}Ca irradiations of actinide targets is near Gaussian excitation functions for $2 \leq \times \leq 5$ with FWHM $\approx$ 10 MeV. The experimental excitation functions for SHE formation have been summarized in a recent review [15], and are reproduced here in Fig. 1. In Fig. 1, horizontal error bars indicate the range of projectile energies subtended in the target layer, and vertical error bars indicate statistical errors. Systematic errors in energies and cross sections are not represented.

2.2 Cross bombardments

Cross bombardments, where the same nuclides are produced using different target/projectile combinations, also provide information used in Z and A assignments. Evidence that the same nuclide was produced in each reaction includes observation of decay mode, decay energy, and half-lives for the nuclide in question, and for all α-decay daughters. Cross bombardment information for the SHE has been concisely summarized in the recent review of SHE formation and decay by Oganessian and Utyonkov [15]. Their cross bombardment summary figures are reproduced here in Fig. 2 (even-Z) and Fig. 3 (odd-Z).

Cross bombardments can produce the same nuclide directly. For example, ^{287}Fl (element 114) was produced directly in both the ^{244}Pu(^{48}Ca,5n)^{287}Fl and ^{242}Pu(^{48}Ca,3n)^{287}Fl reactions. When considering these two excitation functions (Fig. 1), it can be seen that ^{287}Fl was produced at the highest CN excitation energies in the ^{244}Pu(^{48}Ca,5n)^{287}Fl reaction and at the lowest CN excitation energies in the ^{242}Pu(^{48}Ca,3n)^{287}Fl reaction. This provides strong evidence that the exit channels (determination of x) have been assigned consistently.

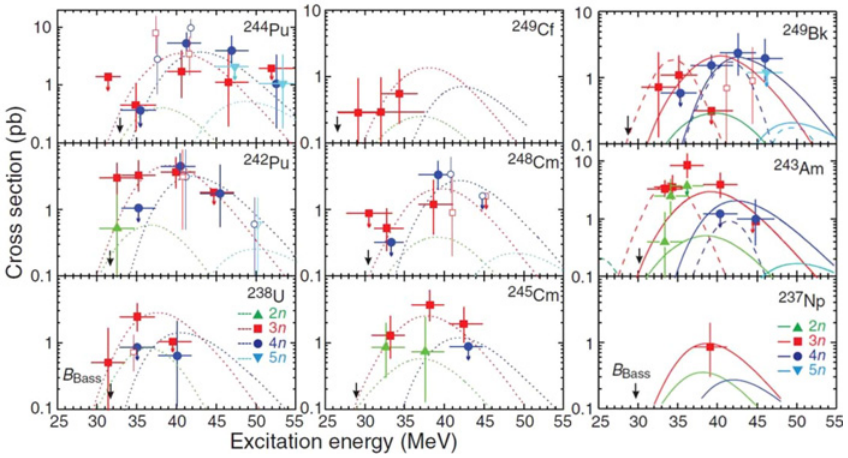

Figure 1. Excitation functions for even-Z SHE (left, center) and odd-Z SHE (right) (reproduced from Ref. [15]).

Another class of cross bombardments has been used, where the same nuclide is produced directly, and as an α-decay daughter. Continuing the example with ^{287}Fl, this element 114 nuclide has also been produced as the α-decay daughter of ^{291}Lv (element 116). This element 116 parent nuclide was produced in both the ^{248}Cm(^{48}Ca,5n)^{291}Lv and ^{245}Cm(^{48}Ca,2n)^{291}Lv reactions. Once again, observing that ^{291}Lv $\rightarrow$^{287}Fl $\rightarrow$^{283}Cn $\rightarrow$^{279}Ds was produced at the highest CN excitation energies in the ^{248}Cm(^{48}Ca,5n)^{291}Lv reaction, and at the lowest excitation energies in the ^{245}Cm(^{48}Ca,2n)^{291}Lv reaction provides strong evidence that these exit channels (determination of x) have been assigned consistently.

Cross-bombardment data indicates that the Z and A assignments have been made consistently: either consistently correct, or consistently incorrect (always off by the same ΔZ and/or ΔA). As may be expected when making Z and A assignments based on a limited number of decay chains, there may be some assignments for individual decay chains, or for certain nuclides, which are in error. As an example, recent data and more comprehensive statistical assessments question the originally suggested α-decay link between the isotopes ^{293}117 and ^{289}115 (cf. Fig. 3) [16–18]. It is expected that these discrepancies will be cleared up as more SHE data become available.

2.3 Decay systematics

Alpha-decay energies tend to increase with increasing Z of the emitting nucleus because the increased Z causes a Coulomb separation energy increase. Additionally, α-decay energies tend to decrease with increasing neutron number, N, because the increased nuclear radius of the α-decay daughter causes a Coulomb separation energy decrease. These $E_\alpha(Z, N)$ systematics have been quantified empirically [13]. The same $E_\alpha(Z, N)$ trends result from calculation of α-decay Q-values, Q_α, using mass models [19]. Q_α trends for even-even nuclides (with even Z and N) are clear, because the decay is dominated by decay from the 0^+ ground-state in the parent to the 0^+ ground state in the daughter. The situation is somewhat less clear in the α-decay of odd and odd-odd isotopes, because the dominant α-decay usually proceeds to an excited analogue-state in the daughter nucleus (with the odd particle(s) in the same Nilsson state as in the parent).

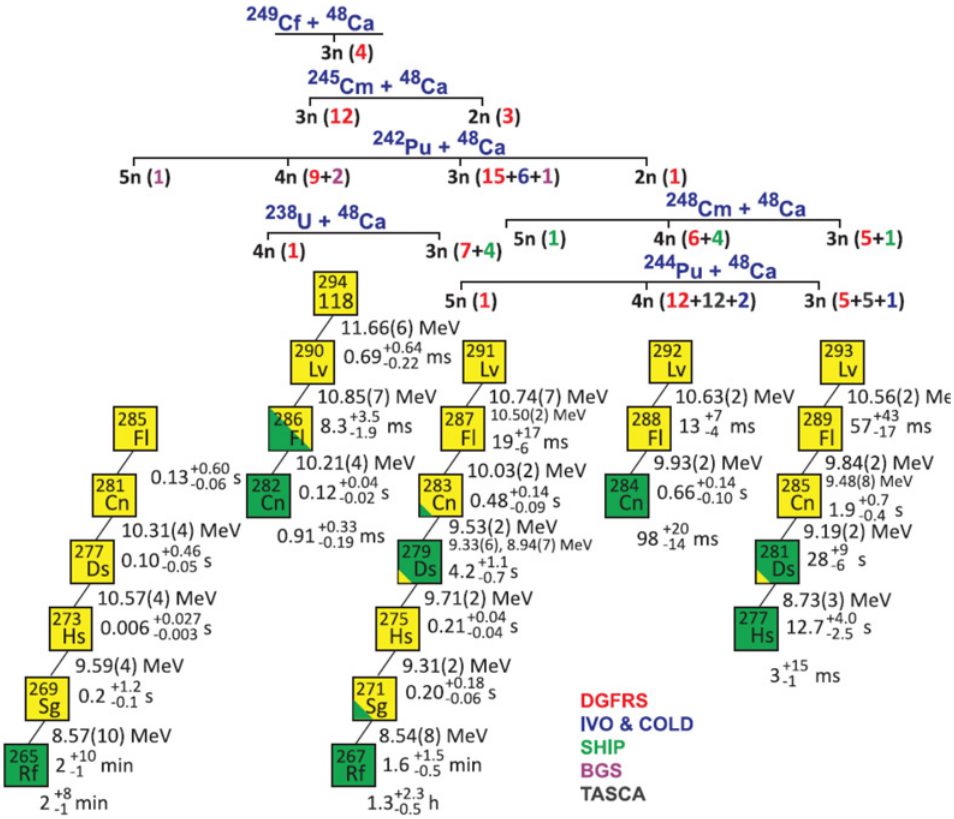

Figure 2. Cross bombardment summary for even-Z SHE (reproduced from Ref. [15]).

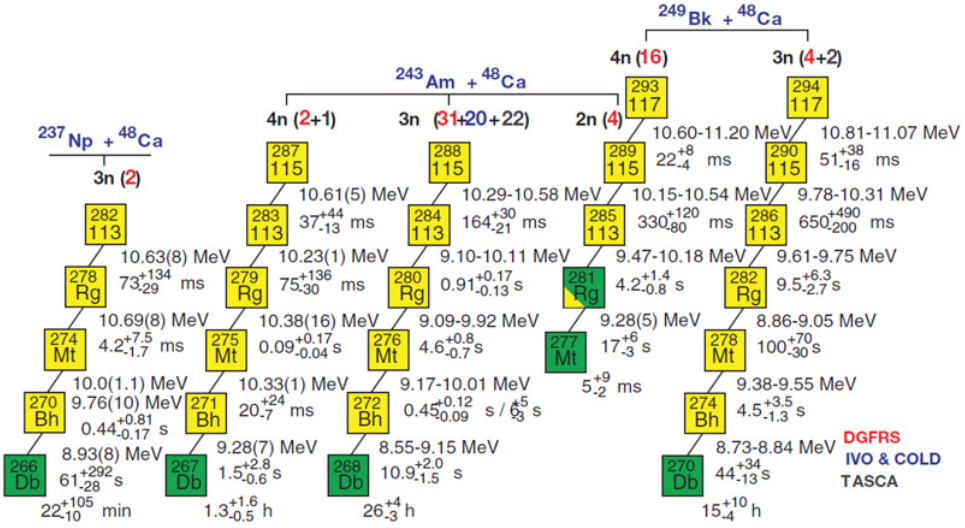

Figure 3. Cross bombardment summary for odd-Z SHE (reproduced from Ref. [15]).

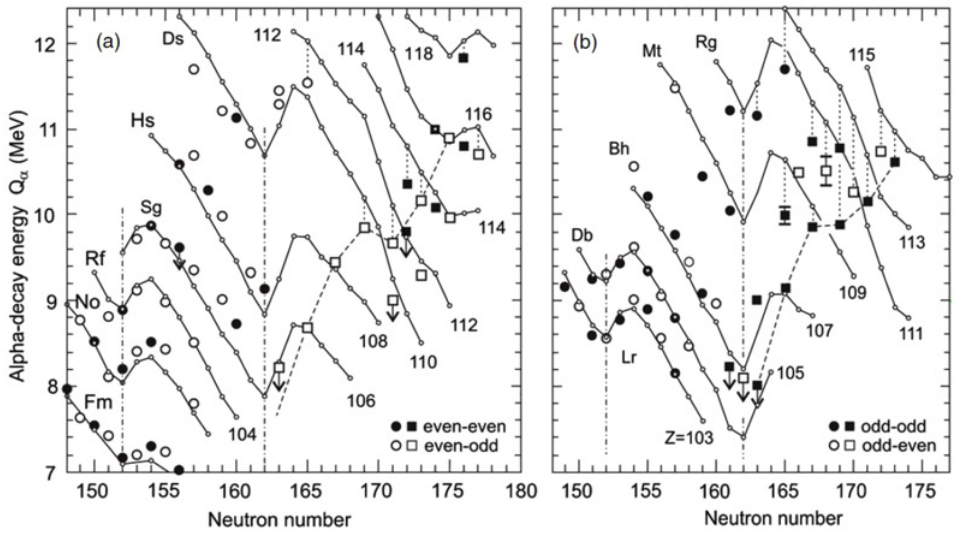

Figure 4. Comparison of α-decay energies with predictions [19] (taken from Ref. [14]).

The maximum α-decay energies for even-Z SHE isotopes are plotted as a function of N in the left panel of Fig. 4. These decay energies have been corrected for the recoil energy imparted to the α-decay daughter nucleus, resulting in α-decay Q-values (assuming a ground-state to ground-state transition). They show a smoothly decreasing α-decay energy with increasing N, giving confidence that the *relative* neutron numbers have been assigned correctly. In addition, the systematic trend of increasing Q_α with increasing Z is readily apparent, giving confidence that the *relative* proton numbers have been correctly assigned.

The maximum α-decay energies (corrected for daughter recoil) for odd-Z SHE isotopes are plotted as a function of N in the right panel of Fig. 4. Once again, the expected trends with increasing Z and N are apparent. The *relative* Z and N for these odd-Z SHE isotopes appear to have been assigned correctly. Also, it should be noted that the odd-Z Q_α values at a given N fall between those for the adjacent even Z. This shows that the Zs of these isotopes do, in fact,fall between those assigned for the even-Z SHE isotopes.

Spontaneous fission (SF) half-life systematics can also be used to test the accuracy of SHE Z assignments. SF is strongly hindered by the presence of odd protons and/or odd neutrons. Even-even isotopes decay by SF with half-lives according to regularly trending spontaneous fission systematics. The SF decays of isotopes with odd numbers of protons or neutrons are hindered by several orders of magnitude compared to even-even neighbouring isotopes. A review of the spontaneous fission half-lives reported for SHE isotopes shows that the even-Z vs. odd-Z and even-N vs. odd-N assignments have been made correctly.

2.4 Summary of accuracy of existing assignments

Evidence provided by measurement of excitation functions, implementation of cross bombardments, and analysis of decay systematics, provides a consistent picture of formation and decay of SHE. The Z and A assignments result in a contiguous set of isotopes and isotones. It has been demonstrated conclusively that, with very few exceptions, the *relative* positions in Z and A are assigned correctly.

Given the strong evidence supporting the contiguous nature of the present Z and A assignments, we must now consider if the whole set of assignments could be wrong. Odd-even effects in the SF half-lives give strong evidence that the set of assignments is not off by one unit in Z or and/or one unit in A. If the set of assignments is incorrect, the Z and/or A assignments must be off by an even number of protons and/or neutrons.

3. Uncertainties in *A* assignments

Mass number, A, assignments have been made based on the energetics of the compound nucleus evaporation residue reaction. The number of neutrons evaporated from the compound nucleus is calculated by summing the center-of-mass frame reaction energy and the Q-value for target plus projectile going to an EVR and x free neutrons. This is further corrected by an estimate for the kinetic energy of the evaporated neutrons. Since the mass defect of the EVR is not experimentally known, predictions from mass models are used.

If these reactions actually proceed via compound nucleus formation followed *exclusively* by evaporation of neutrons, and the mass number is incorrect by at least two neutrons (as shown in the previous section), the mass models must be incorrect by nearly 20 MeV to arrive at incorrect A assignments. Could the mass models, which have been shown to be accurate in the No ($Z = 102$) region, be inaccurate by ~20 MeV in the $Z = 114$ SHE region? If SHE nuclei behave similarly to the lighter heavy elements, cumulative errors in α-decay Q-values, stepping up from the No region (where masses have been measured) to the SHE region could result in 5–10 MeV errors in SHE masses. To reach a 20-MeV mass error, some important new physics, not included in the mass models would be required. The study of SHE is a study at the extreme limits of Z and A. One may expect that the nucleus "finds" innovative ways to stabilize itself. Examples of such new physics (not included in many mass models) could be unusual nucleon density distributions within the nucleus, such as a "bubble nucleus" with a reduced proton or nucleon density at its centre. The result of a calculation including such nucleon density effects [20] is shown in Fig. 5. Another possibility is that the SHE nuclei take on different shapes to increase stability. Many of the mass models consider only axially-symmetric shapes. Non-axially symmetric shapes such as octahedral (stabilized by six Z~20, A~48 clusters) or tetrahedral (stabilized by four $Z = 28$ clusters) could result in enhanced SHE stability, and correspondingly reduced mass. Some plots of these octahedral and tetrahedral deformations [21] are presented in Fig. 6.

As an example, the ^{243}Am(^{48}Ca,xn)$^{291-x}$115 reaction is considered. If element 115 masses are 20 MeV smaller than predicted by mass models, the ^{291}115 compound nucleus will be formed with an excitation energy 20 MeV larger than expected. With this increased excitation energy, approximately two extra neutrons will be evaporated from the compound nucleus to form the fully de-excited evaporation residue. What has been assigned as the ^{243}Am(^{48}Ca,2n)^{289}115 reaction would actually be ^{243}Am(^{48}Ca,4n)^{287}115. Similarly, what has been assigned as ^{243}Am(^{48}Ca,3n)^{288}115 would actually be ^{243}Am(^{48}Ca,5n)^{286}115, and ^{243}Am(^{48}Ca,4n)^{287}115 would be ^{243}Am(^{48}Ca,2n)^{285}115. Conversely, if the element 115 masses are 20 MeV larger than predicted by the mass models, the ^{291}115 compound nucleus will be formed at an excitation energy 20 MeV lower than expected. With this decreased excitation energy, approximately two fewer neutrons will be evaporated from the compound nucleus to form the fully de-excited evaporation residue. What has been assigned as the ^{243}Am(^{48}Ca,2n)^{289}115 reaction would actually be ^{243}Am(^{48}Ca,0n)^{291}115. Similarly, what has been assigned as ^{243}Am(^{48}Ca,3n)^{288}115 would actually be ^{243}Am(^{48}Ca,1n)^{290}115, and ^{243}Am(^{48}Ca,4n)^{287}115 would be ^{243}Am(^{48}Ca,2n)^{289}115. There are two arguments supporting the present assignments, rather than those shifted by plus-minus two neutrons. First, the 0n

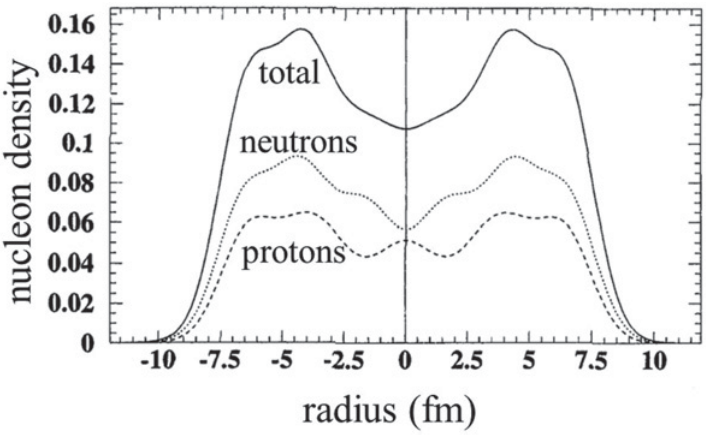

Figure 5. Calculation of radial nucleon density for $^{292}120$ (from Ref. [20]).

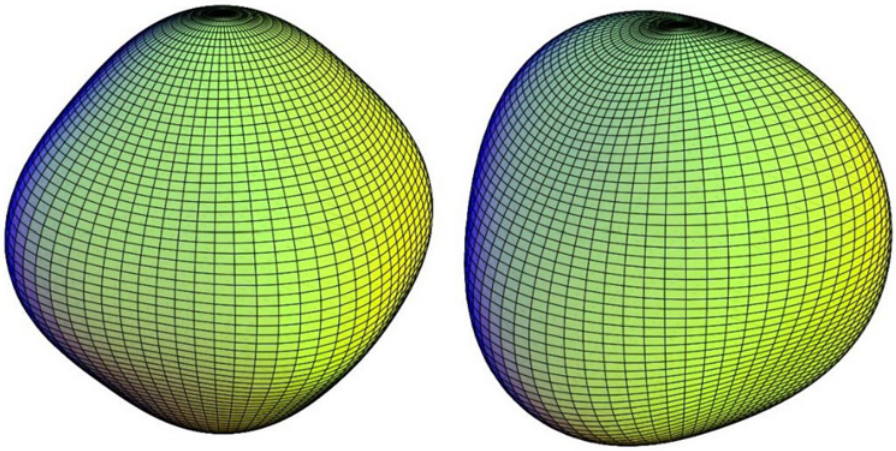

Figure 6. Non-axial nuclear deformations (left: octahedral, right: tetrahedral) (taken from Ref. [21]).

exit channel in the $^{243}\mathrm{Am}(^{48}\mathrm{Ca},0n)^{291}115$ reaction is expected to have a vanishingly-small cross section. However, the sub-picobarn cross sections reported for what has been assigned as SHE produced by two-neutron exit channels may qualify as "vanishingly-small" with this heretofore unobserved exit channel becoming observable. Second, the α-decay systematics presented in Fig. 4 show the effect of the $N = 162$ deformed shell, supporting the present set of A assignments. However, the position of this deformed shell could easily be shifted by plus-minus two neutrons with one neutron level in the Nilsson diagram having a shape dependence different than calculated.

4. Uncertainties in *Z* assignments

Z-assignments for the contiguous set of SHE isotopes have been made largely on the assumption that SHE formation proceeds by compound nucleus formation, followed by de-excitation *exclusively* by neutron emission. There are other possibilities. In the formation

of neutron-deficient isotopes of lighter elements ($Z = 50$–80) by compound nucleus – evaporation reactions, de-excitation by emission of α-particles (and neutrons) is quite common, accounting for a large fraction of the EVR cross section. Such αxn exit channels have not been found to be significant in the production of elements with $102 \leq Z \leq 113$ in "cold fusion" reactions using projectiles from ^{48}Ca to ^{70}Zn with Pb and Bi targets. However, αxn exit channels could become more important at even higher Z for formation of SHE. Also, processes, such as αxn exit channels could have been proceeding at picobarn levels in the $102 \leq Z \leq 110$ region, which are insignificant compared to the microbarn – to – nanobarn cross sections found for the formation of elements 102–110. These charged-particle evaporation processes could dominate the picobarn cross sections found for formation of SHE at even higher Z. For example, the energetics of (^{48}Ca, $3n$) reactions are similar to those for (^{48}Ca, αn) reactions, and what has been interpreted as ^{248}Cm(^{48}Ca, $3n$)^{293}116 could, in fact, be ^{248}Cm(^{48}Ca, αn)^{291}114.

There is an argument against dominance of αxn exit channels in the production of SHE. SHE produced in αxn exit channels will have broader recoil velocity and angular distributions than SHE produced in xn exit channels. Similar SHE production cross sections have been measured with three gas-filled recoil separators and a velocity filter. These separators, are, respectively, The Dubna Gas Filled Recoil Separator at Dubna, the Berkeley Gas-Filled Separator in Berkeley, and the TransActinide Separator and Chemistry Apparatus at GSI, and the Separator for Heavy Ion Products (SHIP) at GSI. These four separators have different angular acceptance characteristics, and therefore have different relative efficiencies for SHE produced in xn vs. αxn exit channels. A consistent set of SHE production cross sections have seemingly been measured at all four separators, suggesting that αxn exit channels do not dominate SHE production.

One must also consider evaporation of protons during de-excitation of SHE compound nuclei. The same arguments apply here. Processes such as pxn exit channels may become more important at the high-Z of SHE. While pxn exit channels have been insignificant compared to the microbarn – to – nanobarn cross sections found for the formation of elements 102–110, they could have been proceeding at picobarn levels. These picobarn levels could dominate the picobarn level cross sections for formation of SHE. For example, the energetics of (^{48}Ca,$3n$) reactions are similar to those for (^{48}Ca,$2p$) reactions, and what has been interpreted as ^{248}Cm(^{48}Ca,$3n$)^{293}116 could, in fact, be ^{248}Cm(^{48}Ca,$2p$)^{294}114. Once again, the similarity in SHE cross sections measured with four different recoil separators provides some evidence against the predominance of $2pxn$ reactions in SHE production, although this evidence is weaker than in the case of αxn reactions, because the kinematics of $2pxn$ recoils are more similar to those of xn recoils.

Finally, compound nucleus formation followed by neutron evaporation has been the assumed reaction mechanism for SHE formation using ^{48}Ca projectiles with actinide targets. The possibility of pre-compound nucleus particle emission has been ignored. Pre-compound emission of high-kinetic-energy neutrons could be occurring at picobarn cross section levels. Because of the high neutron kinetic energy, the compound nucleus will be cooled much more than expected. Similar arguments can be made for emission of high-kinetic energy pre-compound proton or pre-compound α emission.

5. Conclusion

Excitation functions, cross bombardments, and decay systematics show that the Z- and A-assignments for SHE produced in ^{48}Ca irradiations of actinide targets, and all of their decay daughters form a contiguous block of isotopes. If the formation mechanism, nuclear masses,

and radioactive decay of SHE isotopes behave as in the lighter heavy elements, the position of this contiguous block of isotopes in Z and A is – by and large – correctly assigned.

If mass models diverge from true masses when extrapolating from regions of Z and A with well-known masses to the SHE isotopes, excitation functions could be improperly interpreted, leading to incorrect A-assignments. This divergence between model masses and true masses would have to be unexpectedly large: 20 MeV. Such a large divergence would indicate interesting new physics in nuclear shapes and/or nucleon density distributions.

The production mechanism for SHE isotopes is formation of an excited compound nucleus, followed by de-excitation by neutron evaporation in competition with fission. The Z and A assignments have assumed there is no charged particle emission during the de-excitation process. CN de-excitation involving charged particle emission has been observed in other parts of the chart of nuclides, but has had an insignificant impact on formation of heavy element isotopes close to $Z = 102$. If insignificant charged particle evaporation continues to the SHE, the SHE Z-assignments are correct. However, the SHE lie at the high-Z limit of nuclear stability, and a resurgence of charged particle emission during CN de-excitation is a possibility.

A strong case has been made for the present set of SHE Z and A assignments. Excitation functions, cross bombardments, and decay systematics show that, with very few exceptions, the present Z and A assignments are correct relative to each other. This results in a contiguous set of isotopes. There is a small probability that the Z and/or A assignments for the whole group of SHE isotopes is incorrect by an even number (–2) of protons and/or neutrons.

Before using the measured Q_α values from SHE decay to adjust mass models, the Z and A assignments must be proven to be correct. Otherwise, the mass models could be adjusted to produce mass defects that support the incorrect assignments. If the group of Z and A assignments for SHE is incorrect, there is important and exciting new physics to be learned. If the A assignments are incorrect, this new physics could be new variations in nuclear shapes or nucleon density distributions. If the Z assignments are incorrect, new heavy element formation reaction mechanisms which become important in the heaviest elements on the picobarn cross section scale could be explored.

6. Future work

Experiments are being carried out to measure the Z of SHE by looking for X-ray emission with characteristic energies coincident with SHE α decay [16, 22]. Alpha decay from an odd or an odd-odd isotope typically proceeds to an excited analogue state with odd particle(s) in the same state(s) as in the parent. If the excitation energy of this analogue state is above the K-shell edge, de-excitation by internal conversion of a K electron usually results in a K Xray. These K X rays have well-known energies and relative intensities which are unique for each Z. The first direct proof of SHE Z will probably come from a future α-K-X-ray coincidence experiment in a SHE isotope where the α-decay analogue state in the daughter lies above the K-shell edge and decays by internal conversion with a $M1$ multipolarity.

There are also some possibilities for a direct proof of SHE Z and A by observing α-decay chains that end in isotopes with well-characterized Z and A. Target-projectile combinations that are more neutron-deficient than those that have been successful so far are required. Unfortunately, these reaction combinations seem to have sub-picobarn cross sections, and the α-decay chains tend to terminate by SF before reaching daughters with well-characterized Z and A. Another difficulty results because electron capture decay may become dominant in these neutron-deficient decay chains, shifting the decay chain away from the known daughters.

Chemical separations will play only a minor confirmatory role in the determination of SHE proton numbers. Chemical separations can be performed with SHE to show similar chemical behaviour to periodic table homologs, but the chemical information gained will not be sufficient for assignment of Z. An example is the gas-phase chemistry with what is presumed to be element 112, where atoms of 112 was shown to be volatile and to adsorb strongly onto a gold surface [23, 24]. This behaviour was claimed to be similar to that for the periodic table homolog element, Hg, and different from noble gas behaviour of Rn. However, this chemistry experiment cannot be used to assign Z, as it used atoms with uncertain Z in a chemical separation system that has not been proven to be selective for element 112 over other nearby SHE elements.

An apparatus for direct measurement of SHE mass numbers is being constructed at the Lawrence Berkeley National Laboratory's 88-Inch Cyclotron. A RF Gas Catcher is being installed at the Berkeley Gas-filled Separator focal plane to stop the SHE recoils in ultra-pure He, where they will retain a positive charge state. RF fields and a DC gradient will move the SHE ions toward an exit orifice. SHE ions will be swept through the orifice and into a radiofrequency quadrupole trap where they will be trapped and cooled. The SHE ions will then be ejected from the trap, accelerated to $\sim$10 kV, and passed through a mass separator. The device will have a mass resolution of $A/\Delta A = 1000$, sufficient to determine SHE A from single events.

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