Consistent Interpretation of the Secondary-Reaction Experiments in W Targets and Prospects for Production of Superheavy Elements in Ordinary Heavy-Ion Reactions

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A consistent interpretation is presented of spontaneous fission events previously observed in Hg sources separated from CERN W targets. The energy spectra are interpreted in terms of fission to four fragments with $E(\text{total}) \simeq 320$ MeV. The value of $\bar{\nu}$ is estimated to be between 3 and 5. The measured masses of the fissioning nuclei are consistent with various molecules of element 112 with about 160 neutrons. The prospects of producing superheavy elements with use of targets around W and projectiles like Sr and Zr are discussed.

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In recent years many efforts have been made to produce superheavy nuclei with use of heavy-ion reactions. So far these experiments have been unsuccessful.[1] It is the purpose of this Letter to show, on the basis of the experimental results obtained in the studies of various fractions separated from CERN W targets, that with heavy-ion experiments the prospects of producing the superheavy elements are quite reasonable.

In several papers[2–6] some evidence was presented for the possible production of superheavy elements in W targets that were bombarded with 24-GeV protons. The experiments were based on the idea that in such irradiations the superheavy elements may be produced via secondary reactions.[2–6] It was, however, difficult to interpret the positive evidence obtained in a consistent manner. Recently[7–9] evidence has been presented for the production of long-lived isomeric states of the neutron-deficient nuclei ^{236}Am and ^{236}Bk produced in a W target which had been irradiated with 24-GeV protons. In addition, Hartree-Fock calculations[8, 10] made for the ^{236}Cm nucleus predicted the existence of low-lying oblate isomeric states. In the following we will show that the results obtained in the superheavy-element region can be consistently interpreted if one assumes that, as in the actinide region, the formation of neutron-deficient isotopes is not possible. According to this interpretation it also will become clear that the results obtained with the W targets are not in contradiction with the negative results obtained with U and Th targets,[11] nor with the experiment of Katcoff and Perlman[12] concerning the cross sections for energetic fragments. Preliminary results of this work have been published elsewhere.[13]

Let us now describe the experimental results and their possible interpretation:

(a) Spontaneous fission activity has been found in mercury sources which were separated from two tungsten targets labeled as W2 and W3.[2–4] The kinetic energy spectra of singles events and the sum energy spectra of two fragments were measured[3] for the Hg(W2) source, and after correction for the known contamination[3] were found to be different from the known spectra in the actinide region. The singles spectrum showed indications of three groups and the sum spectrum of two groups. A two-dimensional spectrum of the correlated events was also measured[3] and because of its importance it is presented in Fig. 1. In this spectrum evidence for three groups of coincidences, which are outside the region where most of the ^{252}Cf fragments would have been found, is observed. These coincidence groups are as follows: 69-MeV fragments in the front detector in coincidence with 62-MeV fragments in the back detector, 100 MeV in coincidence with 67 MeV, and 83 MeV in coincidence with 116

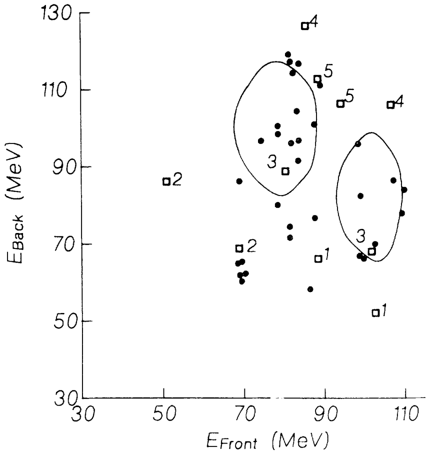

FIG. 1. Correlated energies of fission fragments from the Hg source. The pairs of numbered points show the alternative interpretations when the analysis of the events is ambiguous (Ref. 3). The contours enclose the region in which 90% of ^{252}Cf fragments would be found and are asymmetric because of energy loss in the source backing.

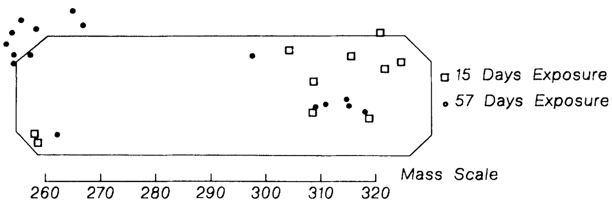

FIG. 2. Positions of fission fragments observed in the Ni foil by use of polycarbonate foils. The heavy line represents the Ni foil. The distance between two mass units was 2 mm. (The points on the left-hand side are due to edge effects of the polycarbonate foils.)

MeV. A statistical analysis, done with the formula given in Ref. 6, shows that the probabilities of these groups being due to accidental concentration of points are very small: 1.7×10^{-5}, $\leq 1.2\times10^{-2}$, and 3.0×10^{-4}, respectively.

The low-energy group of coincidences between two fragments of about 65 MeV is of particular interest. The energy in both detectors is well defined and therefore it is impossible to explain this group as due to energy loss in the source. It is, however, difficult to understand this group in terms of binary fission since then a total kinetic energy of about 130 MeV is expected from isotopes of Hg or nearby elements. Spontaneous fission in the Hg region is very unlikely.[12,14] It seems more likely, therefore, that the two 65-MeV fragments are connected to the others at around 180 MeV (average of 100 and 116 MeV) and 83 MeV. As mentioned above a coincidence between 67- and 100-MeV fragments is actually indicated in Fig. 1. Under the assumption of a four-particle breakup the total measured kinetic energy of the fragments would be about 320 MeV. The Z and A values of the fragments may be estimated with use of the following assumptions: (a) The fissioning nucleus is $^{272}_{112}X_{160}$ (the chemical homolog of Hg; the reason for considering such a neutron-deficient isotope will be clear below). (b) The masses of the fragments are inversely proportional to the measured kinetic energies as is the case in binary fission. (c) The N/Z values of the fragments are equal to the N/Z value of the fissioning nucleus. With these assumptions the four fragments turn out to be ^{80}As, ^{81}As, ^{63}Fe, and ^{48}Ca. The appearance of the doubly closed-shell nucleus ^{48}Ca should be noticed. A fragment scheme like ^{78}Ge (two fragments), ^{68}Ni, and ^{48}Ca (two closed-shell nuclei) is also consistent with the data. The value of $\bar{\nu}$ was estimated to be between 3 and 5 using the arguments of Nix[15] and the predicted masses.[16] This value is in accord with the crude experimental result[3] of between 2 and 5. It also should be mentioned that from the point of view of total energy release, fission to four fragments is prefered[17] in the superheavy-element region.

(b) Part of the Hg(W2) source was run through a mass separator.[4] The technique used in the ion source has been described by Freeman *et al.*[18] The separated atoms or molecules were collected on a thin Ni foil and fission activity was detected on this foil, with use of polycarbonate foils and also photographic emulsions. Figure 2 represents as an example the results of 15-d and 57-d polycarbonate exposures. (The mass determination was good to within one mass unit.) A significant concentration of events is seen in the mass region 308 to 318 .

These fission fragments could not be due to molecules of contaminants like ^{252}Cf or ^{238}U because of the short half-life of the activities, which was estimated by us to be about several weeks. A statistical analysis similar to the one done in Ref. 6 shows that the probability that these events are accidental or spurious is very small. (The background on these measurements was zero. The chances of obtaining two spurious events on the same line, for instance the 308 events, are very small. They are even much smaller if one considers events which appeared in two or more different exposures, such as the events at mass 308 or the four events at mass 317–318 which appeared in four different exposures.) It is, however, also difficult to understand these results in terms of atomic ions because of the large measured masses. An interpretation of the measured masses (from all the exposures) in terms of molecules is given in Table I. The molecules chosen are to be common molecules found with Hg and may be formed by combination of element 112 with various impurity molecular ions of O, N, C, or Cl present in the ion source. (Only events which appeared at the center of the focal plane of the mass separator were considered. However, taking into account the few off-center events does not basically change the discusssion or the conclusion.) It is seen that five different molecules can be related to the isotopes of element 112 with 160–161 neutrons and three molecules to the isotope with 164 neutrons.

(c) Six groups of coincidences between α particles and characteristic x rays of superheavy elements have been found[5] with Au, Tl, and Pb sources which were separated from CERN W targets. It was shown[6] that the probabilities of their being due to an accidental concentration of events are very small. These coincidence groups may be interpreted as due to α decay, which follows by the internal conversion process, of isotopes of elements 111, 113, and 114 to excited states of the corresponding daughter nuclei.

Since the CERN W target may be considered as a "multibeam laboratory" the exact reactions are not known. However, two things may be said about our results: (a) The target was W. (b) The reactions seem to lead to the production of superheavy isotopes with neutron number around $N=160$. It is interesting to note that quite a unique situation, namely, the radiative capture process, is possible under these conditions. The Q values for reactions like

$$^{88}\mathrm{Sr}+{}^{184}\mathrm{W}\rightarrow {}^{272}_{112}X_{160} \text{ or } {}^{86}\mathrm{Sr}+{}^{186}\mathrm{W}\rightarrow {}^{272}_{112}X_{160}$$

are -282.4 and -275.8 MeV, respectively,[16] while the Coulomb barrier between the targets and the projectiles (if we assume that $r_0=1.4$ fm) is around 285 MeV. The situation is the same for similar combinations of targets and projectiles which may lead to cold fusion of isotopes of elements 111, 113, and 114 in the same mass region. The chance of survival of superheavy nuclei produced by the radiative capture process is much larger compared with the various neutron-evaporation processes (because of the small expected values of Γ_n/Γ_f). It is therefore possible that this process manifested itself in our experiments. It should be mentioned that under these conditions our results are not in contradiction with those of Katcoff and Perlman[12] since large cross sections, on the order of a few tens of millibarns, are possible for the radiative capture process. In this case only a few nanobarns are needed for the production cross section of, for instance, Sr ions with energies of $E \geqslant 4.8$ MeV/nucleon, in order to produce about 500 atoms of the isotope $^{272}_{112}X$. In addition, because of the restrictions made by Katcoff and Perlman[12] to fragment energies of $E \geqslant 5.5$ MeV/nucleon and to the angular region between 0° and 45°, their value of $\sigma_{\mathrm{fragment}} \leqslant 6$ nb cannot be considered as a relevant limit for the W targets.[19,20] (The total production cross section of, for instance, ^{86}Sr ions is estimated to be about 1 mb.[21]) It should also be mentioned that the above conditions cannot be fulfilled in U or Th targets.[11] The production cross sections of the neutron-deficient fragments needed are very small[22] and the excitation energies of the formed compound nuclei are very high.

From the point of view of the cluster model[10] isotope $^{272}_{112}X$ may be considered (in addition to the fragments mentioned above) as consisting of four ^{68}Ni nuclei. It seems that the prospects of producing the superheavy element nuclei with use of targets around $Z=74$ and presently available Sr, Zr, or similar beams are quite reasonable. It, however, should be mentioned that the conditions which ex-

TABLE I. Results of mass separator measurements on the Hg(W2) source. Number of fission tracks are given in parentheses for each mass. The masses are arranged according to various possible molecules of element 112 (see text).

A^+	$A^{16}\mathrm{O}^+$	$A^{35}\mathrm{Cl}^+$	$A^{12}\mathrm{C}^{14}\mathrm{N}^{16}\mathrm{O}^+$, $A^{14}\mathrm{N}_3^+$	$A^{14}\mathrm{N}^{16}\mathrm{O}_2^+$	N
269(1)					157
272(1)	288(1)	308(3)[a]	315(2)	317–318(4)	160–161
276(1)	292(1)	311(1)[b]			164

[a] Mass 308 may also be interpreted as $^{276}A\mathrm{O}_2^+$.

[b] Mass 311 may also be interpreted as $^{269}A\mathrm{N}_3^+$ or $^{269}A\,^{12}\mathrm{C}^{14}\mathrm{N}^{16}\mathrm{O}^+$.

ist in the CERN W target cannot be absolutely reproduced in heavy-ion reactions. In the W target the second step of the reaction takes place within about 5×10^{-14} sec after the fragment has been formed. During this short time the fragment is still at high excitation energy and probably quite deformed. Both the Q values of the reactions and the Coulomb barriers are different as compared to normal ions.

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