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The evidence for production of the superheavy element with $Z = 112$ via secondary and direct heavy-ion reactions

A Marinov[a], **S Gelberg**[a] **and D Kolb**[b]

[a]Racah Institute of Physics, The Hebrew University, Jerusalem, Israel
[b]Department of Physics, Kassel University, Germany

ABSTRACT. The evidence for production, via secondary reactions, of a superheavy element with Z=112, in CERN W targets which were irradiated with 24-GeV protons is described. Spontaneous fission events with a half-life of several weeks were observed in Hg sources separated from two W targets. The measured masses of the fissioning nuclei are consistent with various molecules of element 112 with about 160 neutrons. The energy spectra of the fission fragments are interpreted in terms of non-binary fission. A radiative capture process between highly excited and deformed Sr nuclei and corresponding W nuclei is suggested, taking into account subbarrier fusion and extra-push energy phenomena. Indications for production of element 112 via direct $^{88}Sr+^{184}W$ reaction are given.

1. Introduction

The search [1-5] for production, via secondary reactions, of superheavy elements in CERN W targets which were irradiated with 24-GeV protons has several basic advantages: a) Large variety of isotopes, both stable and radioactive are available as "projectiles". b) The second stage of the reaction takes place between highly excited and deformed fragment and another W nucleus in the target. This may affect strongly the fusion cross section due to the subbarrier fusion effect. The detection and identification of the superheavy elements rely on their predicted [6] chemical properties that, for instance, element 112 is the chemical homologue of Hg. In two Hg sources separated from two W targets fission fragments were found and the masses of the fissioning nuclei and the energy-spectra of the fission fragments were measured [1-4]. These experiments, as well as the indications obtained [5] with the direct $^{88}Sr+^{184}W$ reaction are described, and interpreted in terms of the interplay between the extra-push energy and the subbarrier fusion phenomena.

2. Observation of spontaneous fission in Hg sources

Spontaneous fission events were detected [1-3] from two well separated [1] Hg sources, Hg(W2) and Hg(W3) obtained from two W targets (W2 and W3) using polycarbonate films. These fission fragments could not be due to any actinide nucleus with $Z \geq 95$ in general or ^{252}Cf in particular for the following reasons: a) By a study of α-particle spectra [1] the decontamination factor of the Hg(W2) source from any actinide nucleus with $Z \geq 95$ was determined [1,5,7] to be about 3.7×10^9, while the intensity of fission activity in Hg was larger [2] than in the whole actinide fraction. b) The characteristic α-particle group of ^{252}Cf of 6.12 MeV was not seen in the

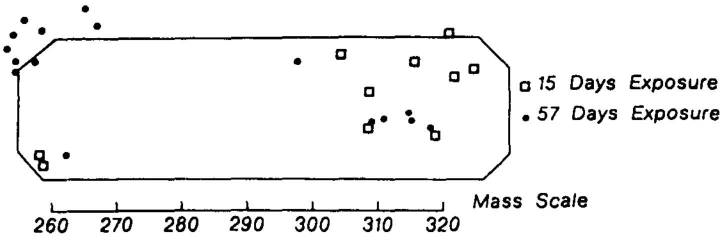


FIG. 1. Positions of fission fragments observed in the Ni foil by use of polycarbonate foils. The heavy line represents the Ni foil. The distance between two mass units was 2 mm. (The points on the left-hand side are due to edge effects of the polycarbonate foils).

TABLE 1. 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.

A^+	$A^{16}O^+$	$A^{35}Cl^+$	$A^{12}C^{14}N^{16}O^+$, $A^{14}N_3^+$	$A^{14}N^{16}O_2^+$	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 $^{276}AO_2^+$.
[b] Mass 311 may also be interpreted as $^{269}AN_3^+$ or $^{269}A\,^{12}C^{14}N^{16}O^+$.

Hg fractions [1,8]. c) The deduced half-life (with large uncertainty) of the fission fragments seen with the Hg(W3) source [2] was found to be about 47 days [5], and by a study of X-ray spectra it was proved that they were not due to any actinide nucleus.

The observed fission fragments could also not be due to Hg itself. a) The expected lifetime for fission of Hg itself is extremely long. b) In two very sensitive experiments where W and Hf were bombarded [9] with ^{12}C and $^{16,18}O$ respectively, and U target was irradiated with 28 GeV protons [10], fission fragments were searched for with negative results.

3. Mass measurements of the fissioning nuclei

Part of the Hg(W2) source was pass through a mass separator [3,4]. 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. Four such measurements were made during 5, 15, 57 and 130 days. The results of the 15 and the 57 days exposures are shown in Fig. 1. In between the second and third measurements, α-particles were searched for [3,5] during 27 days using photographic emulsion. A significant concentration of spontaneous fission events is seen in Fig. 1 in the mass region of 308 to 318. These events could not be due to contamination of ^{252}Cf since the deduced half-life (with large uncertainty) of the activity is about 33 d. (If the fission events were due to ^{252}Cf about 22 events should have been seen in the 130 d exposure at masses 308 and 318 while zero events were observed.) Furthermore no α-particles were observed [3,5] in the photographic emulsion, in the whole mass region of 290 to 320, while more than 80 were expected. The probability that these events are accidental or spurious is very low [4]. The procedure [11] in the ion source of the mass separator, where the Hg fraction was electroplated (without applying any voltage) on a small Cu wire and then evaporated at about 300°C, eliminated any element of $90 \le Z \le 111$.

Table 1 gives an interpretation [4] of the measured masses (from all the exposures) in terms of different molecules which may be formed by combination of element 112 with various impurity molecular ions present in the ion source. It is seen that 11 events can be related to 5 different molecules of the isotope(s) of element 112 with N=160-161.

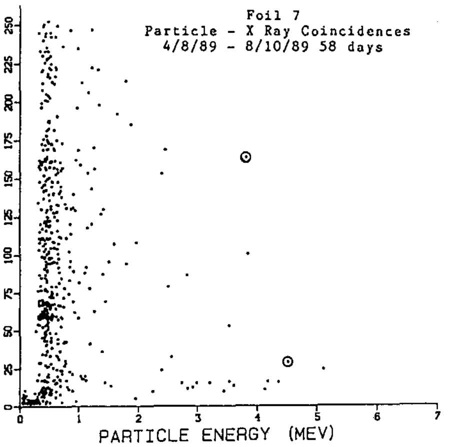
Recently quite large fission barriers have been predicted [12] for superheavy elements with $N \approx 160$. However the predicted alpha particle lifetimes [12] are quite short. It seems that probably long-lived isomeric states in the neutron-deficient superheavy isotopes with Z=112 and $N \approx 160$ were formed. Note that in the study [13] of the actinide nuclei, produced in the same W target, evidence for the production of long-lived isomeric states in the neutron deficient ^{236}Am and ^{236}Bk nuclei has been obtained, and also unidentified particle groups were found [14]. Furthermore various new shape isomers have been predicted by Hartree-Fock [15] and also by Hartree-Fock-Bogoliubov [16] calculations.

4. Energy spectra of the fission fragments

The energy spectra of the fission fragments, of both singles and coincidences between two fragments, were measured [2,4] with part of the Hg(W2) source using Si detectors. Fig. 1 of Ref. [4] shows a two-dimensional spectrum of the correlated events. Evidence for three groups of coincidences which are outside the region where most of the ^{252}Cf fragments would have been found, was observed. Of particular interest is the low-energy group of coincidences between two fragments of about 65 MeV each. It is difficult to understand this group in terms of binary fission. According to $Z^2/A^{1/3}$ low, a total kinetic energy of about 130 MeV is expected [17] from isotopes of Hg or nearby elements. However as was mentioned above a spontaneous fission process in the Hg region is very unlikely and was not found experimentally using two very sensitive methods [9,10]. The data were interpreted [4] in terms of fission to four fragments with $E_{kin}(\text{total}) \approx 320$ MeV. Ternary or quarternary fission processes are possible [18] for the superheavy elements. The value of $\bar{\nu}$ for the four particle breakup was estimated [4] to be between 3 and 5 in accord with the crude experimental result [2] of between 2 and 5.

5. Possible reaction mechanism

The exact reactions in the CERN W target are not known. However, two things may be said. a) The target was W. b) The reactions seem to lead to the production of superheavy isotopes with Z=112 and $N \approx 160$. Under these conditions the radiative capture process is in principle possible [4]. The Q values [19] for reactions like $^{88}\text{Sr}+^{184}\text{W} \rightarrow {}^{272}112$ or $^{86}\text{Sr}+^{186}\text{W} \rightarrow {}^{272}112$ are -282.4 MeV and -275.8 MeV, respectively, while the Coulomb barrier is around 285 MeV. The chance of survival of superheavy nuclei produced very cold by the radiative capture process is much larger compared with the various neutron-evaporation processes. Large fusion cross sections around 1 mb, which are needed in order to interpret the experimental results, are in principle possible (see below). The above conditions cannot be fulfilled [4] in U or Th targets.



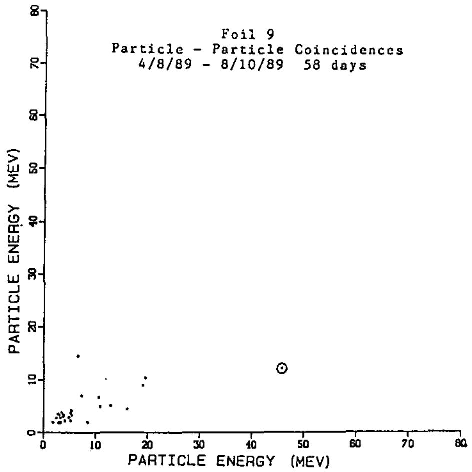


FIG. 2. (Left) A coincidence spectrum of X- and γ-rays versus particles measured with catcher foil No. 7. The resolving time was 1 μs.

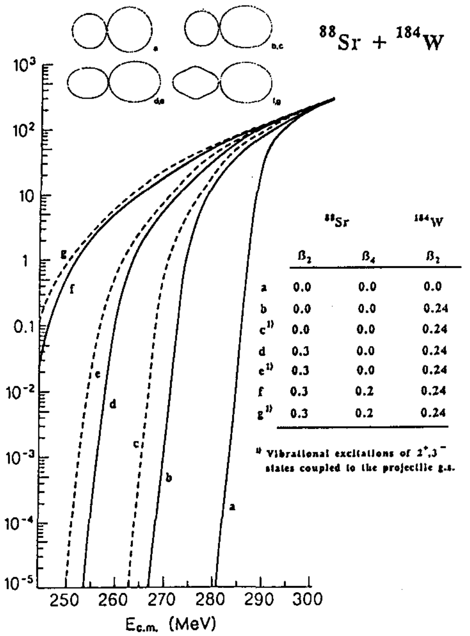
FIG. 3. (Right) A coincidence spectrum of particle versus particle measured with catcher foil No. 9. The resolving time was 5 μs.

6. Study of the ^{88}Sr+^{184}W reaction at 5.1 MeV/u

The ^{88}Sr+^{184}W reaction has been studied [5] with 5.1 MeV/u ^{88}Sr beam provided by the UNILAC accelerator in GSI, using the degrader-catcher foil technique. The expected excitation energies [19] of the compound nucleus were between 0 and 21 MeV, such that the radiative capture and the 1n evaporation reactions were in principle possible.

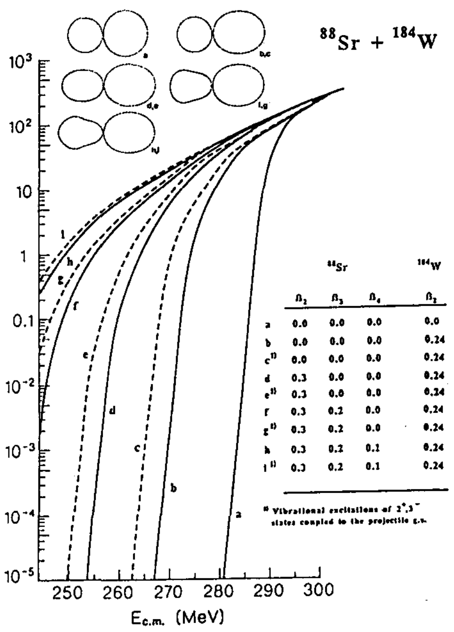
A search for fission fragments using polycarbonate track detectors gave negative results with an upper limit of about 0.8 nb for half-lives of between 10 h to 30 d. Some positive indications were obtained in particle versus X- and γ-ray and particle versus particle coincidence measurements. In Fig. 2 two coincidence events between 3.81 and 4.51 MeV particles and 163.7±0.5 keV and 29.1±0.5 keV X- or γ-rays respectively, are seen. These energies fit with the predictions made for $K_{\alpha 1}$ (163.50 keV [20] or 163.37 keV [21]) and $L_{\beta 4}$ (28.98 keV [20] or 28.94 keV [21]) X-rays of element 112. The probabilities that these two events are due to accidental background were estimated [5] to be 5% and 10% respectively. A possible interpretation may be that an isomeric state or states were formed in ^{272}112 or in ^{271}112 and decayed by internal conversion (which was followed by characteristic X-ray of element 112) to another excited state of the same isotope, which then decay by emitting protons of 3.8 or 4.5 MeV. It should be noted that $L_{\beta 4}$ is the strongest expected L X-ray line related to M1 or M2 transitions. The estimated production cross section is about 20 nb for half-lives of between 10 to 50 days.

In Fig. 3 a coincidence event between a $12.16^{+0.3}_{-0.1}$ MeV alpha particle and a $45.8^{+20}_{-0.4}$ MeV fission fragment was observed. (About 10% pulse height defect of the detector should be considered for the second energy. The large uncertainties toward the higher energies reflect the thickness of the catcher foil and the large solid angle). The probability that this event is due to accidental background is about 1%. An energy of $50^{+20}_{-0.4}$ MeV for the fission fragment is in accord with the secondary reaction experiments [4] where a group of coincident events of about 65 MeV energy for both fragments was observed. An alpha particle energy of



	^{88}Sr β_2	^{88}Sr β_4	^{184}W β_2
a	0.0	0.0	0.0
b	0.0	0.0	0.24
c[1)]	0.0	0.0	0.24
d	0.3	0.0	0.24
e[1)]	0.3	0.0	0.24
f	0.3	0.2	0.24
g[1)]	0.3	0.2	0.24

[1)] Vibrational excitations of $2^+, 3^-$ states coupled to the projectile g.s.



	^{88}Sr β_2	^{88}Sr β_3	^{88}Sr β_4	^{184}W β_2
a	0.0	0.0	0.0	0.0
b	0.0	0.0	0.0	0.24
c[1)]	0.0	0.0	0.0	0.24
d	0.3	0.0	0.0	0.24
e[1)]	0.3	0.0	0.0	0.24
f	0.3	0.2	0.0	0.24
g[1)]	0.3	0.2	0.0	0.24
h	0.3	0.2	0.1	0.24
i[1)]	0.3	0.2	0.1	0.24

[1)] Vibrational excitations of $2^+, 3^-$ states coupled to the projectile g.s.

FIG. 4. (Left) Calculated fusion cross sections using the code CCDEF [24] for the ^{88}Sr+^{184}W system assuming quadrupole and hexadecapole deformations for the ^{88}Sr nucleus.

FIG. 5. (Right) The same as Fig. 4 but including also octupole deformations for the ^{88}Sr nucleus.

12.16 MeV fits the predictions made [19,22,23] for element 112 with 160 or 159 neutrons (see Table 2). This result may indicate the production of a long-lived isomeric state in one of the above mentioned isotopes of element 112. This isomeric state decayed by γ-rays or internal conversion process to the ground state which then decayed by emitting characteristic alpha particle in coincidence with a fission fragment of the daughter nucleus. A cross section of about 0.7 nb was estimated from this coincidence event for half-lives of 10-50 days.

TABLE 2. The differences (in MeV) between the measured alpha particle energy of 12.16 MeV and various predictions made [19,22,23], for the isotopes ^{271}112 and ^{272}112.

	Liran Zeldes	Möller Nix	Möller et al.	Tachibana et al.	Spanier Johannson	Patyk Sobiczewski
^{271}112	0.046	0.40	0.89	-0.26	0.62	0.05[a]
^{272}112	0.22	0.51	0.99	-0.014	0.77	0.27

[a]The average of the values for ^{270}112 and ^{272}112 was taken.

7. Calculated fusion cross sections for the ^{88}Sr+^{184}W system

Fusion cross sections have been calculated for the ^{88}Sr+^{184}W system using the code CCDEF [24]. Fig. 4 shows the results assuming various quadrupole and hexadecapole deformations for the ^{88}Sr nucleus. Since it is possible that the shape of the "projectile" (fragment) in the secondary reactions is not axially symmetric, octupole deformations have been included in the code by us, and the results are given in Fig. 5. Curve "c" in both figures is probably the relevant one for the direct $^{88}Sr+^{184}W$ reaction. A cross section of about 20 nb at 300 MeV is obtained by assuming extra-push energy of about 37 MeV (shifting this curve to the right by this amount). Cross sections of about 1 mb at energies around 290-295 MeV is obtained by shifting curves "f" or "g" in Fig. 4, or curves "f", "g", "h" and "i" in Fig. 5 to the right by an extra-push energy of about 40 MeV. An energy of 40 MeV is consistent with extrapolation of experimental [25] and theoretical [26] extra-push energies to the $^{88}Sr+^{184}W$ system. Smaller extra-push energy is possible for highly deformed ^{88}Sr fragment. It is seen that at least qualitatively it is possible to understand the relatively large deduced fusion cross sections in the secondary reaction experiments in terms of the the large deformations expected for the fragments.

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