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The chemical separation of Eka-Hg from CERN W targets in view of recent relativistic calculations

In 1971 evidence for the production of element 112 via secondary reactions in CERN W targets was obtained. The evidence was mainly based on the observation of fission fragments in Hg sources separated from the W targets, on the measured masses of the fissioning nuclei and on the assumption that element 112 (Eka-Hg) actually behaves like Hg in the chemical separation process. This assumption is analyzed in view of recent relativistic calculations of the electronic structure of element 112. It is shown that in the superheavy element region only the chemistry of element 112 is similar to that of Hg.

Die chemische Trennung von Eka-Hg aus CERN W Targets unter Berücksichtigung neuer relativistischer Berechnungen. *1971 erhielt man experimentelle Evidenz für die Erzeugung des Elements 112 aus Sekundärreaktionen in W Targets am CERN. Die Evidenz basierte hauptsächlich auf der Beobachtung von Spaltfragmenten in Hg Fraktionen, die aus den W Targets chemisch abgetrennt worden waren, den gemessenen Massen der spaltenden Kerne und auf der Annahme, dass Element 112 (Eka-Hg) sich in dem chemischen Trennprozess ähnlich wie Hg verhält. Diese Annahme wurde unter Berücksichtigung neuer relativistischer Berechnungen zur elektronischen Struktur des Elements 112 analysiert. Es wird gezeigt, dass in dem Bereich superschwerer Elemente nur die Chemie des Elementes 112 ähnlich der von Hg ist.*

1 Introduction

Back in 1971 long-lived fission fragments have been found in Hg sources separated from two CERN W targets which were irradiated with 24 GeV protons [1, 2]. Based on the extrapolation of the periodic table the fission activity was interpreted as due to production, via secondary reactions, of the superheavy element with Z = 112. The masses of the fissioning species were measured and heavy masses like 272, 308 and 317–318 were found and consistently interpreted as due to element 112 with 160–161 neutrons and various molecules of it [3]. From the measured mass of the produced superheavy nucleus cold fusion reactions like $^{88}Sr + {}^{184}W \rightarrow {}^{272}112$ and $^{86}Sr + {}^{186}W \rightarrow {}^{272}112$ were deduced [3]. The ordinary heavy ion reaction $^{88}Sr + {}^{184}W$ has been studied and characteristic X-rays of element 112 in coincidence with low energy particles (interpreted as protons) and a very high-energy α particle (E = 12.16 MeV) in coincidence with a fission fragment were found [4]. The results have been summarized in Refs. [5–7].

For many years it was difficult to understand these data and in particular the long lifetime of several weeks of the fission activity, and the large deduced fusion cross sections of several mb in the secondary reaction experiment, and in the nb range in the ordinary heavy ion reaction. However, in recent years new long-lived isomeric states with new radioactive decay properties have been discovered [8–11]. These states are long-lived high spin super- and hyperdeformed isomeric states. The evidence for the existence of the long-lived high spin superdeformed isomeric states is based in the first place on the observation of relatively low energy and about five orders of magnitude enhanced (as compared to lifetime-energy relationship [12]) α particles, where the enhancement is consistent with the calculated penetration through a barrier of a superdeformed nucleus, and the α particles themselves were found to be in coincidence with superdeformed band γ-ray transitions [8]. Secondly, very retarded proton activities were observed which were interpreted as due to superdeformed to normal states transitions [9]. The evidence for the existence of similar hyperdeformed isomeric states is based on the observation of high energy and 13 orders of magnitude retarded α particles in coincidence with superdeformed band γ-rays, where the energy of the α particles fit with theoretical predictions for hyperdeformed to superdeformed transition [10], and, in addition, on the observation of low energies and about seven orders of magnitude enhanced α particles, where the low energies fit with theoretical predictions for hyperdeformed to hyperdeformed transitions, and the large enhancements fit with penetration calculations for such transitions. The lifetimes of the observed high spin super- and hyperdeformed isomeric states have been found to be much longer than that of their corresponding normal ground states [11].

It was shown that the existence of the super- and hyperdeformed isomeric states enables one to consistently interpret the production of the long-lived superheavy element with Z = 112 [11]. The long observed lifetime shows that isomeric state(s) rather than the normal ground state has been produced in the reaction, and the large fusion cross section in the ordinary heavy ion reaction is due to the production of the compound nucleus in a super- or hyperdeformed isomeric state. The shape of such a state is similar to that of the projectile-target combination in their touching point [11]. Therefore much less overlapping and interpenetration are required in this case as compared to the production of the compound nucleus in its normal deformed, much more compact, shape. Secondly, in the secondary reaction an additional effect is taking place [5–7]. The projectile in this case is not a nucleus in its ground state, but rather a fragment that has been produced just within 2×10^{-14} s before interacting with another W nu-

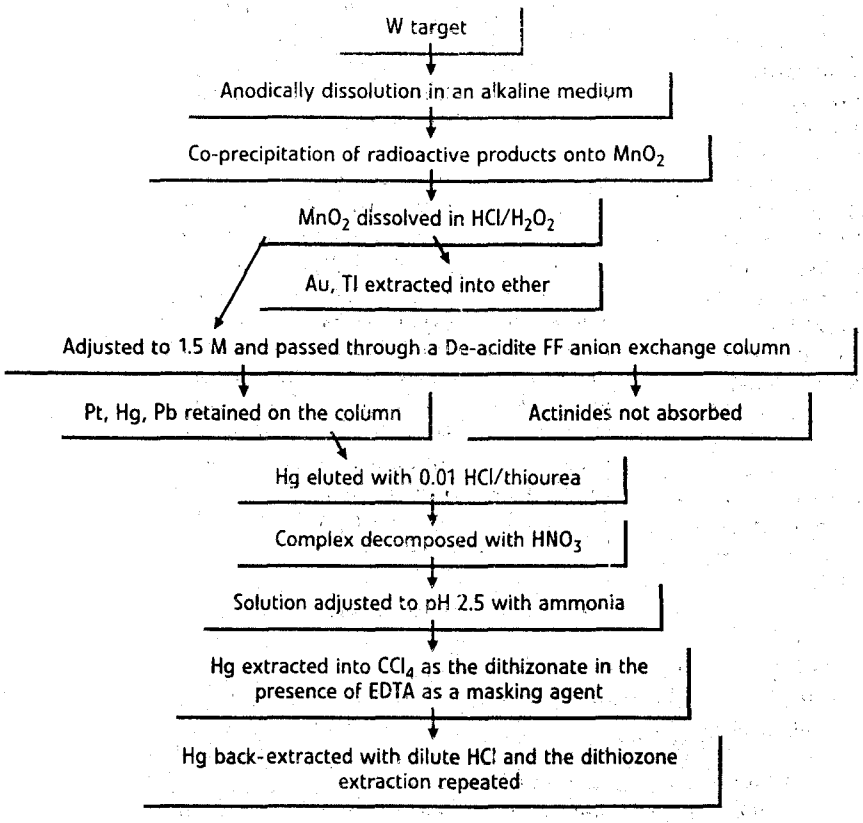

Fig. 1. Block diagram of the chemical separation of Hg from the CERN W targets

cleus in the target. During this short time it is still at high excitation energy and quite deformed. Since the Coulomb repulsion energy between the projectile and the target nuclei, for tip to tip configuration, decreases as a function of deformation, deformation has a large effect on the fusion cross section as is well known from the sub-barrier fusion effect [13, 14].

It is seen that a consistent interpretation to both the secondary reaction experiment in CERN W targets and the ordinary $^{88}Sr + {}^{184}W$ reaction is possible, in terms of production of the superheavy element with $Z = 112$ and $N \simeq 160$ in an isomeric state(s), provided that the chemical properties of element 112, which were used in the chemical separation procedure, are similar to those of Hg. Recently the question was raised if it is justified to expect that element 112 will act like Hg, since some relativistic calculations indicate that it might show properties more like a noble gas [15] (see also Refs. [16, 17] and references therein). The purpose of this paper is to show that, including relativistic effects, only element 112 is similar enough to Hg to follow it in the chemical separation.

2 The chemical separation procedure that isolated the fission activity from the CERN W targets

In the original experiments [1–3] the above fission fragments were found in Hg sources that were separated from two CERN W targets irradiated with 24 GeV protons, but not in Au, TI and Pb sources [18]. The chemical procedure was described in [1] and for the sake of completeness it is summarized in Fig. 1. Fission fragments were seen several times from the produced Hg sources [1, 2]. The Hg source from the W2 target was then electroplated, without applying any voltage, on a small piece of Cu wire that was put in the ion source of a mass separator and heated to about 300 °C [19]. Based on experimental data and on extrapolation of the periodic table this low temperature eliminates any element with $90 \leq Z \leq 111$. The masses of the fissioning species were measured and interpreted as due to the atom and various molecules of the $^{\sim 272}112$ nucleus, where the molecules are known to be common molecules formed with Hg itself [3].

3 The chemical separation procedure in view of recent relativistic calculations

It is clear that the measured fission activity basically followed the chemistry of Hg, otherwise one would have readily lost about 500 atoms, which where responsible for the observed fission fragments, out of about 30 g of W material, in the complex chemical procedure. It must be due to a particular element. As mentioned above, since in the periodic table element 112 is in the same column as Hg having a closed s-d shell, it was concluded that the fission fragments are due to this element. It should be mentioned that this conclusion is also in accord with recent relativistic calculations [15]. It is shown [15] that the $p_{1/2}$ and $s_{1/2}$ level energy distance even increases from Hg to element 112 and thus makes partial $p_{1/2}$ occupancy at least as improbable as in Hg. Occupation of the $p_{1/2}$ shell by one electron or more, ($Z = 113$ and higher) or removal of at least one electron from the s-d shell (Z below 112) would drastically change the chemical and physical properties. E.g. from the experience with s-d atomic level structures in the periodic table we see that the evaporation temperatures stay high as long as the s-d shell is not closed, with a strong drop for the closed $d^{10}s^2$ configuration as in Hg and $Z = 112$. On the other hand the $ns_{1/2}$ and $(n\text{-}1)d_{5/2}$ which were quite apart in Hg are now almost energetically degenerate, because the $7s_{1/2}$ orbital becomes more bound due to a direct relativistic effect and the $6d_{5/2}$ less bound due to indirect relativistic effects (stronger shielding of the nuclear charge by the relativistically enhanced deeper binding of the $s_{1/2}$ and $p_{1/2}$ orbitals). The less bound $6d_{5/2}$ makes it more reactive and the more bound $7s_{1/2}$ makes it more noble than in Hg. These two effects may compensate and make chemical behaviors of Hg and element 112 similar. (For instance, according to the relativistic calculations [15] the binding energies (D_e) of Hg and element 112 on Cu are almost the same). On the other hand the chemistry of Hg cannot be similar to that of any other superheavy element in this Z region. Therefore it may be concluded that the chemical separation procedure described in Fig. 1, which was followed by electroplating, on Cu (without applying any voltage) and evaporation at low temperature of about 300 °C, did isolate element 112 and no other. It is essential to note that the chemical procedure of Fig. 1 was done an Hg and element 112 at various oxidation states, and not at on elemental state like in [20, 21] (see also [22, 23]) where one is basically sensitive to the volatility and the adsorption on Au properties of the element. These properties might be more similar to those of Rn than to Hg.

4 Summary

The chemical separation procedure that isolated Hg sources from the CERN W targets is analyzed in view of recent relativistic calculations for the atomic energy levels of element 112. It is shown that the observed fission activity in the Hg sources with its high measured masses of the fissioning species can be associated only with this element.

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