

## Radiochemical Investigations at the FLNR

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The paper considers a programme of development of radiochemical investigations at the FLNR in the light of recent results on the synthesis of isotopes of elements 108–116 in reactions between  $^{48}\text{Ca}$  and  $^{238}\text{U}$ ,  $^{242}\text{Pu}$ ,  $^{244}\text{Pu}$ ,  $^{248}\text{Cm}$ . Three main lines of the investigation are discussed: chemical identification and study of chemical properties of superheavy elements (SHE), on-line mass separation for the detection of SHE, and the setting of new experiments on the search of SHE in Nature.

### 1. Introduction

The results of experiments aimed at the synthesis of superheavy elements in the reactions between  $^{48}\text{Ca}$  and  $^{248}\text{Cm}$  (Ref. 1),  $^{244}\text{Pu}$  (Ref. 2),  $^{242}\text{Pu}$  (Ref. 3), and  $^{238}\text{U}$  (Ref. 4) carried out at the FLNR in 1999–2001 point to a substantial increase in the stability of nuclides with  $Z = 108$ – $116$  and  $N = 170$ – $175$ . The observed decay chains of nuclei of superheavy elements consist of successive  $\alpha$  decays and are terminated by spontaneous fission.

Relatively long half-lives of isotopes of elements 108–116 obtained in reactions with  $^{48}\text{Ca}$  and chemical properties of SHE predicted theoretically provide possibilities for new experiments devoted to the chemical identification of SHE, study of their chemical properties, on-line mass separation for the detection of SHE, and search of SHE in Nature.

### 2. Chemical Identification of Elements 112 and 114

The experiments on chemical identification of elements 112 and 114 and study of their chemical properties are planned as the first priority. The  $^{238}\text{U}(^{48}\text{Ca}, 3n)^{283}112$  reaction is chosen for the first experiment. The  $^{283}112$  nuclide decays by spontaneous fission with the half-life  $T_{\text{SF}} = 3^{+2.8}_{-1.0}$  min.<sup>5</sup> The cross section of the above mentioned reaction is equal to about 5 pb.<sup>4</sup>

According to classical concepts, element 112 belongs to group IIB of the Periodic Table and is a homologue of Zn, Cd, and Hg. Theoretical works predict that element 112 has an enhanced volatility (Hg-like),<sup>6</sup> and like Hg it must adsorb on metals due to the formation of quite strong intermetallic bonds.<sup>7</sup> However, a number of works predict that element 112 is chemically inert like Rn.<sup>8</sup>

These theoretical concepts became the basis for the experiment, developed at the FLNR, on the chemical identification of element 112. A general scheme of the experiment is presented in Figure 1.

The  $^{238}\text{U}$  ( $\sim 2$  mg/cm<sup>2</sup>) target is irradiated with a  $^{48}\text{Ca}$  beam 264 MeV in energy and  $\sim 6 \times 10^{12}$  s<sup>-1</sup> (1 pμA) in intensity. The recoils are thermalized in helium at atmospheric pressure. Then recoils are transported by helium through a Teflon capillary to the detection system consisting of two subsequently linked blocks: Au-coated PIPS-detectors and the ionization chamber. Both blocks are surrounded with  $^3\text{He}$ -counters registering neutrons from spontaneous fission. Thus the transport line (ca. 25 m) will provide separation of the volatile 112 element from the non-volatile reaction products, detection of SF events by PIPS detectors will indicate that the 112 element is adsorbed on the Au surface (Hg-like), and detection of SF event by the ionization chamber in the gas phase will indicate that the 112 element is inert gas (Rn-like). Optimistic estimates predict the

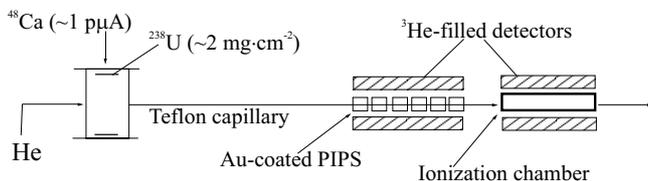
registration of one nucleus per 2–3 days, thus the sensitivity of the chemical experiment may be by  $>5$  times higher than that of physical experiments with using kinematic separators. Technical details of the chemical experiment and the results of model experiments with short-lived Hg isotopes are described in Reference 9.

Experiments on chemical identification of element 114 will be the next stage. We will use the  $^{244}\text{Pu}(^{48}\text{Ca}, 3,4n)$  reaction. The choice of effective methods of separation and detection will depend on the experimental results of the experiment on the chemistry of element 112 and results of model experiments with short-living Pb isotopes. One can expect that element 114 will also have an enhanced volatility. Then the scheme of separation and identification will be similar to that used in the case of element 112 and described above. The identification of element 114 is possible by its daughter isotopes of element 112:  $^{244}\text{Pu}(^{48}\text{Ca}, 4n)^{288}114 (\alpha; 2.6 \text{ s}) \Rightarrow ^{284}112 (\alpha; 19 \text{ s}) \Rightarrow ^{280}110 (\text{SF}; 7.5 \text{ s})$  and  $^{244}\text{Pu}(^{48}\text{Ca}, 3n)^{289}114 (\alpha; 30 \text{ s}) \Rightarrow ^{285}112 (\alpha; 15.4 \text{ min}) \Rightarrow ^{281}110 (\alpha; 1.6 \text{ min}) \Rightarrow ^{277}108 (\text{SF}; 16.5 \text{ min})$ . In the identification of decay chains, the background created by short-living isotopes undergoing  $\alpha$  decay, first of all the isotopes of Rn and Po, is of primary importance. One of possible schemes of the low-background experiment is shown in Figure 2. The scheme allows for carrying out the on-line experiment. The recoil nuclei are thermalized, then are carried out by a gas flow and adsorbed on the first column at a set time (the second, third, and fourth ones are closed). Then — on the second column (the third and fourth ones are closed and the first column is blown by He cleaning it out from Rn); then — on the third column (the fourth one is closed, He is blown through the second column, the first column is heated up to a certain set temperature and desorbed nuclei of elements 112 and 114 are transported by He into the first detection block). And finally — on the fourth column (the first column is cooled down to a temperature required for the adsorption of elements 112 and 114, the second one is heated and the nuclei are transported into the second detection block, the third one is blown by He). Then the cycle is repeated. It is possible to unite the detection blocks into a single one. It is just a general scheme, its realization requires detailed elaboration.

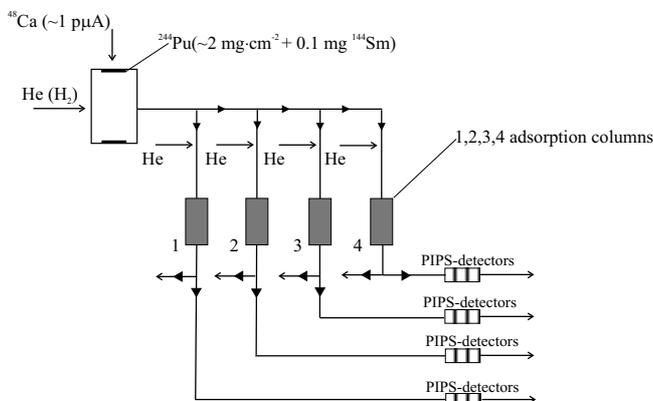
### 3. On-line Mass Separation for the Synthesis of SHE

Because of the short lifetimes of superheavy nuclides (100 μs–10 ms), kinematic separators have been traditionally used for their synthesis. Such separators work as velocity (or energy) selectors providing in-flight separation of nuclides during the time  $\sim 1$ – $10$  μs. At present, six similar setups work all over the world, namely, at the GSI (Darmstadt, Germany), RIKEN (Wako-Shi, Japan), GANIL (Caen, France), LBL (Berkeley, USA), and JINR (Dubna, Russia, two separators). All of them separate reaction products, but do not measure masses of separated atoms. The masses are determined indirectly by means of

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**Figure 1.** Scheme of the 112 element identification experiment.



**Figure 2.** Scheme of "low background" experiment for chemical identification of 114 element.

investigating the properties of atoms implanted in a focal plane detector.

Since the lifetimes of nuclides located within (or in the vicinity of) the "island of stability" are considerably longer, one should revise the concept of separation itself. Instead of kinematic separators it is more reasonable to use classical separators, which are less fast-acting and measure masses of atoms with a high accuracy. A design of such a separator is presented below.

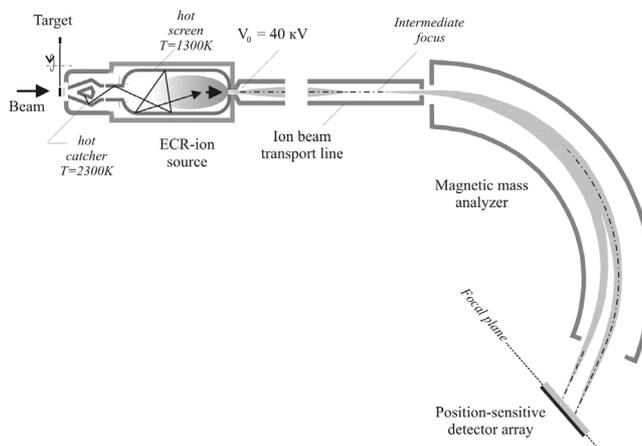
Schematic view of the analyser is shown in Figure 3. We gave it the name of MASHA (Mass Analyzer of Super Heavy Atoms).

Recoil nuclei emitted from the target are stopped in a hot Ta catcher and heated up to the temperature  $T \sim 2300$  K. Implanted in the catcher atoms drift, due to thermodiffusion, towards its surface and enter the chamber of the ECR ion source ( $f = 2.45$  GHz). In the ion source, the atoms are ionized with  $Q = +1$  and then are extracted from plasma, forming a beam with the energy  $E = 40$  keV. Having passed through magnetic and electrostatic elements of the separator and mass analyzer, the atoms reach the separator focal plane which is at a distance of approximately 14 m from the ion source. In the separator, the ECR exit slit (the object) is transformed into a dispersive image, the mass resolution is as high as  $M/\Delta M \sim 1000$ –1200. A focal plane detector (FPD) installed in the separator focal plane has the position resolution  $\Delta X \sim 1$  mm to maintain the above mentioned mass resolution. The detector has a semiconductor strip structure.

Atoms of superheavy elements are to be separated from  $^4\text{He}$  atoms (the ion source gas),  $^{48}\text{Ca}$  ions (primary beam ions), target fission fragments with  $A = 70$ –150, and target-like atoms, the products of transfer nuclear reactions, with  $A = 200$ –250. No background can be allowed in the registration zone so that one could observe decays of the superheavy nuclides in positions along the focal plane, which are determined by the nuclide masses.

The setup described above has the following advantages for the separation of superheavy nuclei:

- (i) Unlike kinematic separators, in which the maximum target thickness is limited by the necessity of preserving kinematic characteristics of a reaction and thus cannot be more than 0.2–0.3 mg/cm<sup>2</sup>, in a classical separator it can be increased up to 1.5–2.0 mg/cm<sup>2</sup>. This circumstance allows synthesizing two (or probably even three) isotopes simultaneously.
- (ii) All the elements with  $Z = 112$ –118, being chemical ana-



**Figure 3.** Mass Analyzer of Super Heavy Atoms.

logues of Hg, Tl, Pb, Bi, Po, At, and Rn, have low boiling points and high volatility. Their fast transport from the hot catcher to the ion source chamber and high ionization efficiency in the ion source plasma can be organized.

Taking into account the above mentioned, one can expect that the total efficiency of the new setup will be few times higher than those of existing kinematic separators.

#### 4. Search of SHE in Nature

Theoretical predictions concerning the existence of the "island of stability" made in the 1960s predetermined the theoretical research on predicting the chemical properties of SHE as compared with their more light homologues as well experimental investigations connected with the search of SHE in Nature. The most active stage of this work was in the 1970s and 1980s. Special attention was paid to a group of nuclides with  $Z$  and  $N$  in the vicinity of magic numbers  $Z = 114$  and  $N = 184$ , for which the lifetimes were assumed to be the longest ones and could be compared with the age of Earth. That is why the properties of element 114 (Eka Pb) were predicted most fully. Keller et al.<sup>10</sup> wrote that the chemistry of element 114 should be similar to that of  $\text{Pb}^{+2}$ , with a more distinguished tendency to formation of complexes in solutions. Pitzer<sup>8</sup> noted that element 114 would be a volatile liquid or gas due to the effect of the closed shell  $7p_{1/2}^2$ . Similarly to theoreticians, experimentalists initially concentrated their attention on the search of element 114 in Nature, and later included elements 110–117 into the orbit of search.

In the search for SHE, the highest sensitivity of  $10^{-13}$ – $10^{-14}$  g/g was reached in the registration of events of neutron emission multiplicity using detectors with  $^3\text{He}$  counters<sup>11</sup> and neutron detectors with liquid scintillators.<sup>12</sup> A detailed description of the obtained results is presented in review works by Seaborg,<sup>13</sup> Flerov,<sup>14</sup> Hermann,<sup>15</sup> and Kratz.<sup>16</sup> Note that no unambiguous data pointing to the existence of SHE in Nature was obtained. In the 90s, the above mentioned work was practically stopped due to lack of positive experimental results as well as due to some new (for that time) theoretical predictions made by Möller et al.<sup>17</sup> The prediction was that the nuclides  $^{288}110$  and  $^{290}110$  would be the most stable of all SHE, and their half-lives would be just 200 days.

The results on the synthesis of isotopes of elements 112, 114, and 116 obtained at the FLNR in combination with theoretical calculations based on the macroscopic-microscopic model of the stability of even-even isotopes of elements 108–116 (Smolańczuk<sup>18</sup>) allow one to revitalize the experiments aimed at the search of SHE in Nature.

The setting of new experiments presupposes estimation of previous ones. Speaking about the search for SHE in Nature, analysis of the known experiments (so far leaving apart the problem of formation mechanism in the process of nuclear synthesis)

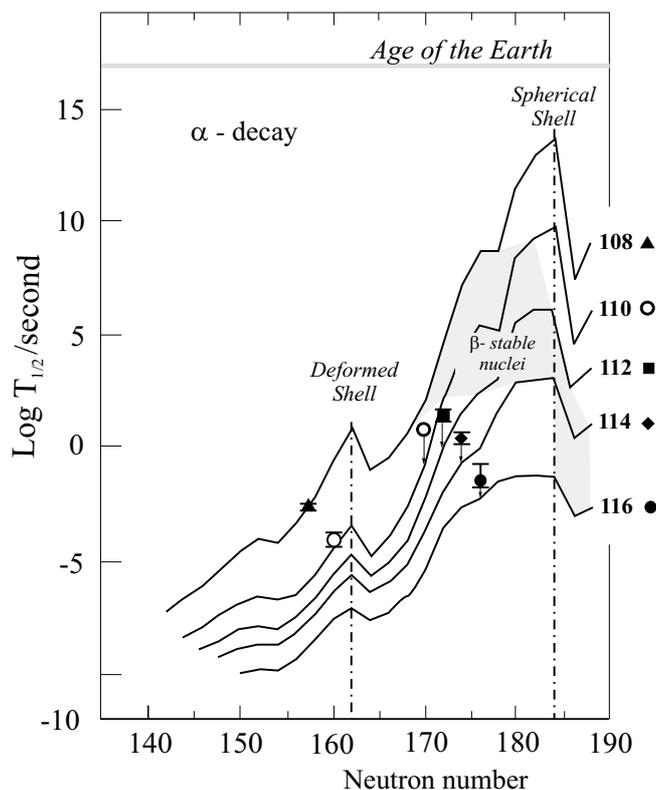


Figure 4. Half-life of the 108–116 elements (theory and experiment).

comes to two principal questions: the choice of the registration method and the choice of the object of search.

The data obtained in Reference 19 on the neutron multiplicities in the fission of superheavy nuclei (SHN) from a weakly excited state make it possible to state that the neutron multiplicity ( $\nu$ ) in the fission of SHN from the ground state will be equal to no less than 8. Thus a method of registration of SHE using scintillation and  $^3\text{He}$  counters employed in previous works seems to be justified, and the obtained results — quite correct.

It has been noted above that in the search for SHE the experimentalists were mainly focused on the investigation of samples promising from the point of view of finding elements 110–117, i.e. from Eka-Pt to Eka-At. According to modern concepts, the most stable nuclides will have atomic numbers  $Z \leq 110$ . Figure 4 shows the calculation data<sup>18</sup> on the lifetimes of even-even isotopes of elements 108–116. It is predicted that the most long living one will be the nuclide  $^{292}108$  ( $\log T_{1/2\text{SF}}(\text{s}) = 12.01$ ;  $\log T_{1/2\alpha}(\text{s}) = 13.73$ ). Figure 4 also shows experimental values for the half-lives of isotopes of elements 108–116 synthesized to date. One can see that the lifetimes of nuclides  $^{280}110$ ,  $^{284}112$ ,  $^{288}114$ , and  $^{292}116$  exceed the calculation values by 1–2 orders of magnitude. One cannot exclude that this difference will be growing with increasing  $N$  and that the lifetime of the nuclide  $^{292}108$  will be comparable with the age of Earth.

However, note that the problem of the  $\beta$  stability of these nuclei has not been solved yet. According to Smolańczuk's calculations the nuclei  $^{276-286}108$  should be stable ones. But he does not exclude a possibility of existence of more  $\beta$ -stable nuclei in this region.

The search of element 108 (Eka-Os) in Nature is regarded by us as an extremely interesting though problematic task. In the first series of experiments we will investigate Os samples. A sample of metallic Os (about 1 kg) will be produced from the

products of copper-nickel concentrate sublimation. Then, using a cascade of gas centrifuges, a heavy fraction in the form of  $^{192}\text{OsO}_4$  will be extracted. Although the content of the most heavy isotope  $^{192}\text{Os}$  in the natural mixture is 41%, one can expect that tetra-oxide of element 108 will be added to the first portions and thus the concentration co-efficient will be no less than 1000. Analysis of the obtained concentrate will be performed in a low background laboratory with the use of ionization chamber. The expected sensitivity of the experiment in assuming the lifetime  $T_{1/2} = 10^9$  years of the sought nuclide is about  $10^{-16}$  g/g.

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