

E7-2002-64

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ON-LINE MASS SEPARATOR
OF SUPERHEAVY ATOMS

Submitted to «Nuclear Instruments and Methods A»

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I. EXPERIMENTAL APPROACH TO THE SYNTHESIS OF SUPERHEAVY ELEMENTS

The investigations in the field of heavy nuclei have led during the past few years to the synthesis of a series of isotopes of superheavy elements with atomic numbers 110-116 [1]. It has been shown experimentally that increasing the neutron number in the nuclei with $Z \geq 110$ brings forth a rise in their lifetime with respect to different radioactive decay modes (α -, β -decay and spontaneous fission). The obtained results can be taken as evidence for the existence of "islands of stability" in the region of superheavy nuclides, arising from the structural properties (the nuclear shells) of nuclear matter.

The formation cross sections for heaviest nuclei in fusion reactions amount to a few picobarns, for the heaviest nuclides - fractions of a picobarn. As a result, in long-term experiments (lasting for several months) only one or few atoms of the new nuclides are produced.

During the last 25 years, the so-called "cold fusion reactions" [2] were used for the synthesis of new elements. In these reactions the isotope ^{208}Pb ($Z = 82$) has been used as target material, bombarded by accelerated ions of mass A from 40 to 70. In reactions of this type the 6 heaviest elements up to $Z = 112$ have been produced.

The lifetime of the nuclei with $Z = 110-112$ is 10^{-4} to 10^{-3} s [3].

Both, the small cross section and the short lifetime require adequate experimental approach, which in turn imposes certain restrictions to the possibilities to synthesize new elements.

One of the most important factors in these investigations is the question of identifying the nuclides and the determination of their radioactive decay.

For the heavy nuclei with $Z \leq 102$ the determination of the atomic number followed from radiochemical experiments. The experiments were based on a branch of inorganic chemistry - the "chemistry of actinides", which got developed during the 1950-70s. In other cases, the identification of the new nuclides was based on their radioactive properties, observed in the sequential α -decays leading to known nuclides. It was assumed that the radioactive properties of the final nucleus were very well defined, and by means of the decay chain it was possible to establish the mass and atomic number of the new, formerly unknown, parent nucleus. This method has been used in the synthesis of all elements with $Z \geq 102$.

In the given approach, the problem is divided into two parts: separation of the superheavy nuclei from an enormous number of reaction by-products and a fast delivery to the detectors; measurement of the decay of the new α -radioactive nuclide and all of its daughter products until in the decay sequence a known nucleus is observed.

The separation of the nuclei, produced in the reactions, is based on the kinematic characteristics of the reaction products - the energy of the recoil nuclei and the emission angle with respect to the beam direction. The selection of the reaction products according to these parameters defines the selectivity of the

separator. It has been shown that a kinematic separator allows suppressing the effect of background products of the reaction by a factor of 10^4 - 10^6 . The efficiency for transporting the evaporation residues - atoms of the new element - to the focal plane of the separator, for a period equal to their time of flight from the target to the detector ($t \sim 1$ - $10 \mu\text{s}$), is 30-40%. Use is made of this principle in all 6 separators engaged in experiments on the synthesis of new elements: in LBL (Berkeley, USA) [4], GSI (Darmstadt, Germany) [5], RIKEN (Tokyo, Japan) [6], GANIL (Caen, France), and the two setups in JINR (Dubna) [8,9].

The necessity to preserve the kinematic characteristics of the heavy nuclei in the process of separation imposes definite limitations to the thickness of the target: $\Delta x \leq 200$ - $500 \mu\text{g}/\text{cm}^2$ depending on the ion mass.

Unfortunately, the suppression of the by-products of the reaction by a factor of 10^6 is far from satisfactory in reaching background-free conditions in the separator focal plane.

So as to be able to distinguish the decay sequence of a nucleus produced at a rate of 1 atom/month, from the signals loading the detector at a rate of 10-50 Hz (the ratio is $\leq 2 \cdot 10^7$), it is necessary to use a sophisticated detector array, which is capable of registering the consequent decay with high resolution in energy, time and position. The reliability of distinguishing the decay sequences in such conditions strongly depends on the half-life both of the parent nucleus and of its daughter products. The increase in the lifetime considerably complicates the data analysis.

Whatever one may say, the traditional experimental approach to the synthesis of new elements is far from optimal in the synthesis of neutron-rich nuclei located on the "islands of stability" of superheavy elements.

- The limitation on the target thickness hinders increasing the yield of superheavy nuclei. The increase of the yield owing to the increase of the beam intensity (keeping in mind the high price of the initial material ^{48}Ca and the necessity of a considerable upgrade of the accelerator) does not bring forth an improvement of the effect-to-background ratio.
- The nuclei in the new region of stability may undergo other radioactive decay modes (β -decay or spontaneous fission). The background conditions in this case are also different.
- The increase in lifetime complicates the data analysis and imposes a limit to the time interval for registering the decay sequence of the heavy nucleus. This limit, without additional systems for suppressing the background from other reaction products reaching the focal plane of the kinematic separator, amounts to $t_{\text{lim}} = 1$ - 2 hours.

Finally, last but not least, comes the following circumstance. The whole decay chain of the heavy nucleus starts and ends at an unknown nuclide. The identification of the charge and mass numbers of the new nuclides is done on the basis of their decay properties and the data from additional experiments (cross reactions, probability of different decay channels of the compound nucleus etc.), which, in the case of small cross sections, is a rather difficult problem.

II. IDENTIFICATION OF SUPERHEAVY ATOMS

(Measurement of the atomic mass)

As it is seen in Fig. 1, the half-life for α -decay of the even-even isotope with $Z = 114$ ($N=174$) amounts to about 2 s. Its daughter nucleus - the isotope with $Z = 112$ ($N=172$) - is also an α -emitter with $T_{\alpha} \sim 0.7$ min. Finally, the grand-child-nucleus - $Z = 110$ ($N=170$) - undergoes spontaneous fission with $T_{s.f.} \sim 7.5$ s. The whole decay sequence lasts about 1.5 min. The neighbouring odd isotopes of element 114 with $A = 287$ and 289 are more long-living - their decay sequences last about 4 min and 32 min, respectively.

One may certainly assume that the isotopes of the yet unknown elements with odd atomic numbers 111, 113 and 115 and neutron numbers $N \geq 170$ will be more stable than their even-even neighbours.

Thus the measured and expected half-lives of the neutron-rich nuclei of interest with $Z = 111-115$ will be of the order of seconds, in some cases - even tens of seconds.

It should be noted that elements with $Z = 112-120$ in their chemical properties are homologues to the easily volatile metals Hg-Ra. The boiling temperature of these elements lies in the region of $T_{\text{boil}} \leq 1700^{\circ}\text{C}$. According to calculations of the atomic structure the elements EkaHg - EkaRa should be more volatile than their stable analogues [10].

It is noteworthy that in the synthesis of all artificial elements heavier than uranium the mass of the new atoms was never measured. This is due to the properties of the actinide nuclei with $Z = 89-103$, the separation efficiency of which is small. The elements with atomic numbers $Z = 104-111$, which are the chemical analogues of Hf, Ta, W, ... Au, are also not promising what concerns mass-separation because of their high boiling temperatures reaching values up to 3000°C .

Having in mind the properties of the superheavy atoms - chemical analogues of the heavy metals - the experimental approach to their synthesis in heavy ion induced reactions can be radically changed.

Instead of separating the reaction products by their kinematic characteristics (using in-flight recoil separators), they may be separated by their mass (using on-line mass-separators).

The operating principle of fast mass-separators, directly determining the mass of the separated atoms, is well known. The reaction products are stopped in a heated catcher (it can be the target itself), escape from the catcher into the plasma of the ion source as a result of diffusion, get ionized in the plasma, are extracted by an external electric field and, then are analyzed by their mass in a magnetic spectrometer of high resolution. Such a scheme, with some modifications, could be used for separating superheavy atoms with $Z = 112-120$.

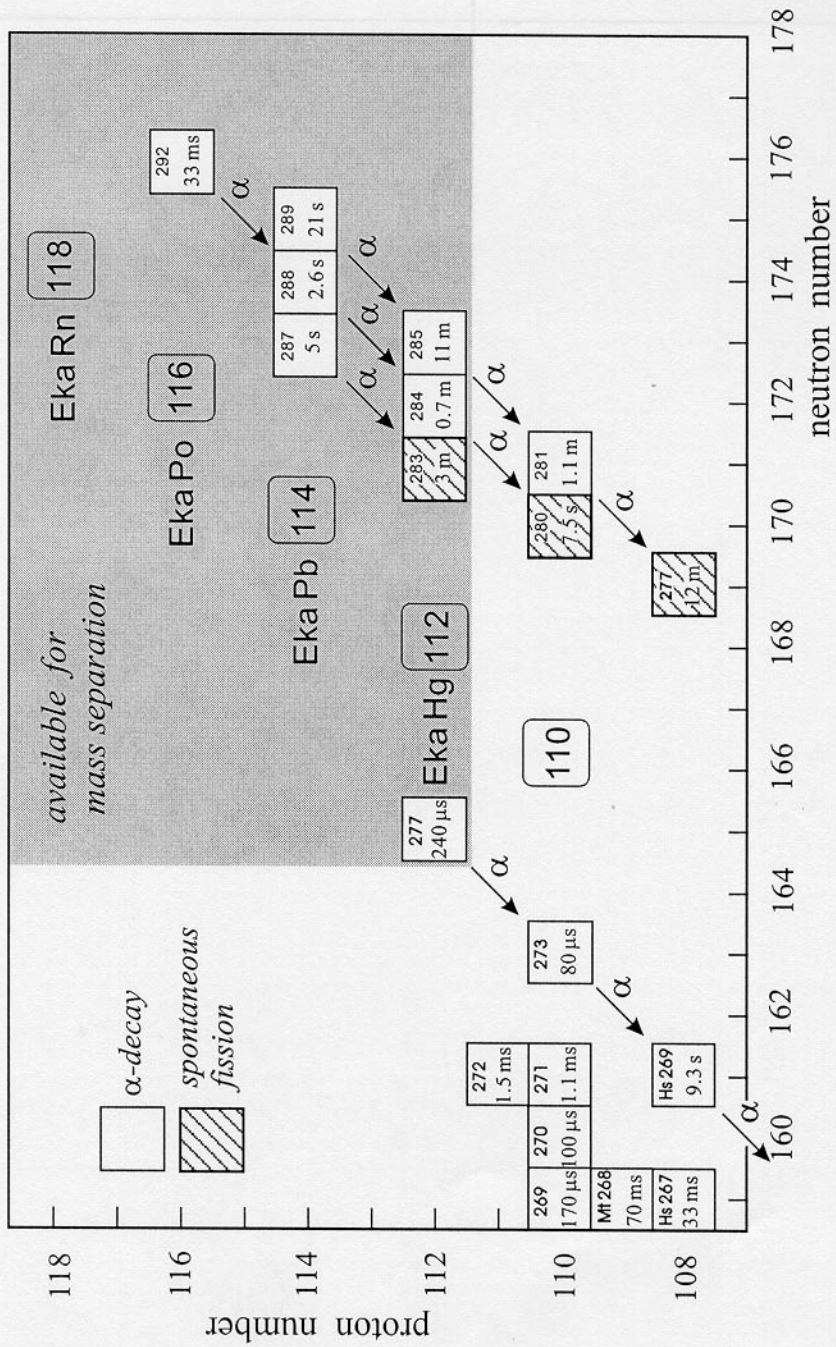


Fig. 1. The end of the chart of nuclides. The decay chains are shown for the heaviest atoms, synthesized in heavy ion induced reactions.

III. MASS-ANALYZER FOR SUPERHEAVY ATOMS (MASHA)

The basic principles of the mass-separator (mass-analyzer) of superheavy atoms can be seen from the schematic presentation, given in Fig. 2. The recoil nuclei, flying out of the target, are implanted into a catcher heated to a temperature $T_{\text{coll}} \sim 2000^\circ\text{C}$.

The range of the recoil nuclei in the working layer defines the target thickness. It depends on the kinetic energy of the heavy atom, formed in a fusion reaction. For the $^{244}\text{Pu} + ^{48}\text{Ca}$ reaction more than 80% of the atoms of element 114 will leave a target layer 1.5 mg/cm^2 thick into an angular cone $\theta_L = \pm 10^\circ$ with respect to the beam direction. The energy loss of the beam in the target layer is about 15 MeV, a value exceeding the width of the excitation function leading to the formation of nuclei with a given mass. Close to the Coulomb barrier in this energy interval, two neighbouring isotopes can be synthesized simultaneously. Increasing the projectile mass allows using target layers even thicker than 1.5 mg/cm^2 , which, in turn, will correspond to a larger excitation energy range in the compound nucleus.

The residual range in the catcher material defines the depth of embedding of the atoms in the catcher. For the reaction considered here, $^{244}\text{Pu} + ^{48}\text{Ca}$, and a Ta catcher the depth of implantation of the atoms will amount to $\Delta x \sim 1\text{-}2 \text{ }\mu\text{m}$. When using a profiled catcher, this value can be reduced a few times. Then the diffusion time for Pb and Bi atoms from a heated Ta catcher ($T \sim 2000^\circ\text{C}$) according to calculations and the available experimental data [11] will be $\tau_{\text{diff}} \leq 1 \text{ s}$.

The atoms, diffused from the heated catcher, are injected into an ion source.

Other possibilities exist when the function of the target and catcher are combined. The diffusion of the reaction products takes place from the powder granules of uranium carbide or plutonium oxide at high temperature with the subsequent motion in the thermo gradient column towards the ECR source. In this case, an "infinitely thick" target may be used, which will cover the whole energy range from the Coulomb barrier to the maximum desirable value.

In the present setup, it is supposed to use a source of the ECR type, operating at a frequency of 2.45 GHz. In order to increase the efficiency of ionization of neutral atoms, in the ion chamber a hot screen is to be placed so as to reflect the atoms falling on its surface. The effusion time of the atoms in the interaction with the surface of the screen depends, as is well known, on the physical and chemical properties of the elements to be separated. At a screen temperature $T_{\text{scr}} \geq T_{\text{boil}}$ the effusion time is $\tau_{\text{effus}} \leq 1 \text{ ms}$. The less the temperature, the higher the effusion time τ_{effus} . For the Pb and Bi atoms at $T_{\text{scr}} \approx 1400^\circ\text{C}$, following the data from ref. [11], $\tau_{\text{effus}} \approx 0.1 \text{ s}$. For all other elements from Hg to Ra at the same temperature the effusion time is significantly less.

According to the measurements, which we have performed for He^+ and Kr^+ ions with the ECR source at $f_0 = 2.45$ GHz and the data for Ar^+ ions with the source at $f_0 = 5.1$ GHz [12], we may assume that at a frequency of 2.45 GHz and power $W \sim 10\text{-}50$ W, the spectrum of charge states of the heavy atoms will be determined mainly by the $q = 1^+$ component. The following measurements with atoms of Xe and Hg will allow a more accurate determination of the yield of single-charged ions.

The ionization and extraction times from the plasma under an external electric field, following the results from ref. [13], will amount to $\tau_{\text{extr}} < 1$ ms.

The presented considerations make it possible to conclude that the ion sources operating on the principle of the electron cyclotron resonance may compete with sources of other types (plasma or with surface ionization), which are traditionally used in the existing mass-separators.

Determination of the mass of superheavy atoms with accuracy $\Delta m = 0.25\text{-}0.30$ mass units ($M/\Delta M \approx 1500$) is carried out according to their magnetic rigidity in a permanent magnetic field. The parameters of the magnetic analyzer, the "image" size on the focal plane are defined by the characteristics of the "object" in the intermediate focus of the setup (Fig. 2). The object size is in turn connected with the emittance of the beam extracted from the ion source, which will be determined experimentally at the prototypes and at the presently operating ion sources.

In the focal plane of the magnetic analyzer detectors will be placed, which will register the position and decay of the separated atom. The focal plane detector covers 18 mass units. This will make it possible to register and determine the masses both of evaporation residues and of their daughter decay products. At a relatively low kinetic energy, $E \sim 40$ keV, the signal, denoting the arrival of a heavy atom to the front position-sensitive detector, will arise only when the atom decays. In the case of spontaneous fission this signal corresponds to the energy deposition of the fission fragment entering the sensitive layer of the detector.

In the case of α -decay, the signal in the front detector will arise only when the α -particle is emitted in the sensitive layer of the detector (the probability is 50%). After the α -decay takes place, the recoil nucleus ($E_{\text{rec}} \sim 120$ keV) will leave the detector and the side detectors without position measurement will register the consequent decay. On the contrary, if the α -particle is emitted in the back hemisphere, the nucleus is embedded in the detector and the front detector will register the consequent α -particle again with 50% probability. As a result, in the absence of position signals from the α -particles (one or more) indication of the position is given by the spontaneous fission.

In these conditions, in order to increase the sensitivity of registration of consequent decays, the front detector should be surrounded by side detectors (without position sensitivity). Such detectors, used at present at the kinematic separators, have efficiency of about 90% [8,9].

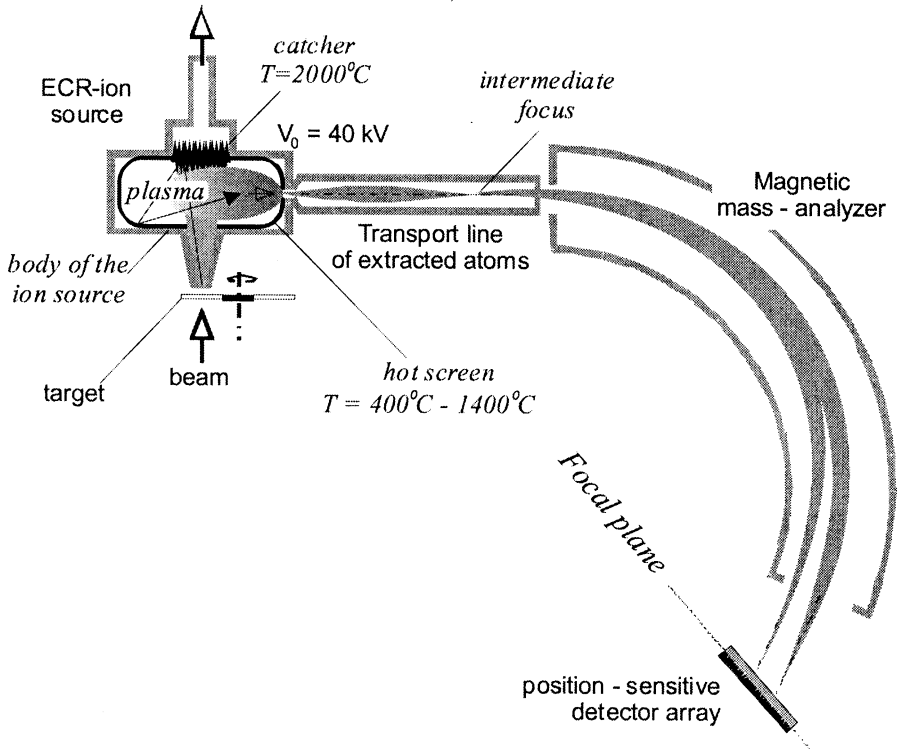


Fig. 2. Schematic presentation of the magnetic analyzer of superheavy atoms (MASHA).

The registration of the atoms in the focal plane of the separator requires exclusion of the α -particle background from the decay of target-like nuclei, especially from the decay products of light isotopes of actinide elements (Th and U), produced in deep-inelastic collisions or quasi-fission. These nuclei are some 40-60 a.m.u. away from the mass of the superheavy atom and can be separated already at the intermediate focal plane.

We are inclined to expect that their appearance in the working area in the form of molecular compounds, imitating the large mass of the studied atom, will be strongly suppressed by the formation probability of similar molecules in the chamber of the ECR-ion source, working at a partial pressure of the parasite atoms of 10^{-8} torr.

The characteristics, which in principle may be achieved in the mass-separator for superheavy atoms, are compared with the characteristics of the existing setups (kinematic separators) in Table 1.

Table 1. Planned characteristics of MASHA compared to those of existing separators.

Parameters	Recoil Separator	MASHA (Projected)
Target thickness (mg/cm ²)	0.3	1.5 or more
Angular acceptance	± 2.5°	± 10° or more
Energy range (MeV)	3	15 or more
Ion charge of the recoiling atoms	20±2	1
Transmission efficiency	30%	≥20%
Suppression of the target-like recoil atoms	10 ³ - 10 ⁵	>10 ⁸
Precision of the mass measurement	± 25	± 0.25
Mass range (mass units)	1	18
Number of the synthesized isotopes	1	2 or more
Relative yield at given luminosity	1	4-5

CONCLUSIONS

The mass analyzer for superheavy atoms (MASHA) in its operating principle and expected characteristics is foreseen for the synthesis of elements with $Z = 112\div 120$. The limitations of the possibilities of the setup are determined by the minimal value of the half-life of the superheavy atom $T_{1/2} \geq 1.0$ s.

The setup MASHA can surpass its analogues both in efficiency of obtaining superheavy atoms and in the information, obtained upon identification of their mass and decay characteristics. It also opens new possibilities to study the chemical properties of superheavy elements.

The shortening of the duration of experiments by using this setup and the improvement of the obtained information will make it possible to widen the field of research in the region of superheavy elements.

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Received on April 3, 2002.

Концептуально рассмотрен «on-line» масс-сепаратор сверхтяжелых атомов (MASHA), предназначенный для сепарации и определения свойств радиоактивного распада атомов новых элементов и изотопов, образующихся в реакциях с тяжелыми ионами. Новые нуклиды с периодом полураспада $T_{1/2} \geq 1$ с транспортируются в ECR-источник, работающий на частоте 2,45 ГГц, и разделяются по массе с массовым разрешением $M/\Delta M \sim 1500$.

В фокальной плоскости спектрометра расположен фронтальный стриповый детектор, окруженный боковыми детекторами, определяющий массу по сигналам от зарегистрированных α -частиц или осколков спонтанного деления с эффективностью около 90 %. По сравнению с существующими «in-flight» кинематическими сепараторами ядер отдачи новая установка может иметь более высокую эффективность и высокую селективность от фоновых продуктов реакции. Установка MASHA может быть использована также в исследованиях ядерных реакций различного типа с пучками стабильных и радиоактивных ионов.

Работа выполнена в Лаборатории ядерных реакций им. Г. Н. Флерова ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 2002

The concept is presented of an on-line Mass Analyzer of SuperHeavy Atoms (MASHA) dedicated to the separation and determination of the mass and decay properties of new elements and isotopes produced in heavy-ion induced reactions. The new nuclides with half-lives $T_{1/2} \geq 1$ s are transported to an ECR-source working at a frequency of 2.45 GHz and are separated by mass with a mass resolution of $M/\Delta M \sim 1500$.

In the focal plane of the magnetic analyzer a front strip detector surrounded by side detectors will be placed to determine the mass according to the signals from the detected α -particles or fission fragments with efficiency of about 90 %. In comparison to other existing in-flight recoil separators, the present setup will be characterized by higher efficiency and high selectivity relative to background reaction products. The setup MASHA may be used also in the investigation of nuclear reactions of different type induced by stable and radioactive beams.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

Макет *Т. Е. Попеко*

ЛР № 020579 от 23.06.97.

Подписано в печать 03.04.2002.

Формат 60 × 90/16. Бумага офсетная. Печать офсетная.

Усл. печ. л. 0,81. Уч.-изд. л. 1,17. Тираж 350 экз. Заказ № 53210.

Издательский отдел Объединенного института ядерных исследований
141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.