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Mendeleev's principle against Einstein's relativity: news from the chemistry of superheavy elements

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Abstract. The review briefly considers the problems of synthesis and chemical identification of superheavy elements. The specific features of their properties are determined by the relativistic effects. The synthesis and chemical investigations into bohrium and element 112 are discussed as examples. The bibliography includes 24 references.

I. Introduction

Mendeleev's first Periodic Table from 1869 was based on atomic masses rather than atomic numbers and had empty positions that stimulated attempts to search for those missing members. As a consequence, several new elements, *e.g.*, gallium, germanium and scandium were then discovered.

Until 1955, chemists have discovered all elements up to atomic number 101 (mendelevium, Md). Expanding to still higher atomic numbers then became a domain in physics — in some cases with the help of chemists: rutherfordium and dubnium. The heaviest element that has been approved by IUPAC has the atomic number 111 (roentgenium) and the heaviest element that has been claimed discovered has the atomic number 118 — though the element with atomic number 117 is still missing.

Chemists have — at least once — investigated chemical properties of all elements up to atomic number 108 (hassium) and the two elements with atomic numbers 112 and 114 (ongoing), respectively.

For elements of most Groups of the Periodic Table, chemical similarity describes their behaviour. Examples are the noble gases, alkaline earth elements or the halogens, respectively. Of course, this rule of thumb is not always equally strict. For, *e.g.*, elements of the Group 14, starting with carbon and ending currently with lead, the term similarity is not really justified. For this group, the valence electron configuration is ns^2np^2. For carbon, both p electrons are positioned with parallel spin in the degenerate sublevels $2p_{1/2}$ and $2p_{3/2}$, respectively (the Hund rule). In addition, the energetically close $2s^2$ level makes for carbon the sp^2 and sp^3 hybridizations possible — the basis of organic chemistry! Increasing spin-orbit coupling for heavier members of this group leads to increasing energy gaps between subshells preventing hybridization.

Chemistry of heaviest elements is usually assigned to atomic numbers of the 'second hundred' (*i.e.*, elements heavier than fermium). Due to the by Z^2 increasing Coulomb attraction between the positively charged nucleus and the atomic electrons, their orbital velocities approach the velocity of light. This becomes true even for valence electrons that define the chemical property. As a consequence, three effects emerge that describe the influence of relativistic effects on the atomic electron orbital energies.[1]

The primary relativistic effect is caused by the increased stability of spherical electron shells. Due to the high orbital velocities, electron masses increase. This leads to a contraction of the orbital radius and an increase in the energy of the electron level. For heavy elements, this holds for s and the $p_{1/2}$ orbitals.

The secondary relativistic effect emerges as a consequence of the primary relativistic effect on the non-spherical orbitals with high angular momentum ($p_{3/2}$, d and f orbitals). Screening effects caused by the spherical orbitals lead to a destabilization of higher angular momentum orbitals.

Spin-orbit coupling for heavy elements increases leading to energetically significantly different energies. A classical example is element 114, below lead, that has been predicted to possibly behave more like a noble gas rather than being lead-like. The reason is the calculated approximately 4 eV energy gap between the filled $7p_{1/2}$ and the empty $7p_{3/2}$ subshells.

Theory and experiments with heaviest elements has therefore developed to a test ground for chemists to evaluate the 'influence of Einstein' on the basic principles of the Periodic Table. Of course, relativistic effects influence chemical properties already for much lighter elements. One

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example is gold: its yellow color is a consequence of the reduced energy gap between the $5d^{10}6s$ and the $5d^{9}6s^{2}$ level.

For elements above atomic number 100 (fermium), much attention received lawrencium and rutherfordium as well as the elements 112 and 114. For lawrencium (Lr), the last member of the actinide series with atomic number 103, and for rutherfordium (Rf), the first transactinide with atomic number 104, a p-element behaviour was predicted, with a $[Rn]5f^{14}7s^{2}7p_{1/2}$ ground state rather than the expected $[Rn]5f^{14}6d7s^{2}$ for Lr (Ref. 2) and a $[Rn]5f^{14}7s^{2}7p^{2}$ ground state rather than the expected$[Rn]5f^{14}6d^{2}7s^{2}$ for Rf (Ref. 3), respectively. As a consequence, elemental lawrencium and rutherfordium should behave like volatile metals, which is typical of p-elements, while d-elements have a very low volatility. Experiments aiming at a separation of volatile lawrencium and rutherfordium, however, failed.[4,5]

For elements 112 and 114 with a filled 6d shell ($Z = 112$) and a filled $7p_{1/2}$ subshell ($Z = 114$), respectively, noble gas-like behaviour was postulated.[6]

Heaviest elements are not only interesting for chemists, but also as many-body nuclear systems (nuclei with more than approximately 250 nucleons) that have essentially no macroscopic nuclear stability anymore. Due to shell effects, nuclei around atomic (proton) number 114 and neutron number 184 should be rather stable against nuclear decay. For nuclei in the vicinity of such magic shells — named 'island of superheavy elements' (SHE) — calculated half-lives reach values of up to 10^{9}s (Ref. 7). Given the large uncertainties of calculated half-lives which may reach many orders of magnitude, the existence of SHE in nature cannot fully be ruled out. Many groups have extensively searched for such elements in nature, however, without success. One drawback is the unknown chemical property that has to be extrapolated in order to separate possible SHE from natural samples.

Therefore, heavy elements have so far been synthesized and chemically investigated only at the single atom level at large accelerators by fusing heavy ions with appropriate target elements. Products formed in complete fusion processes are then ejected from a thin target due to the high recoil momentum and collected in a gas. After being stopped, the products are subjected to chemical reactions that enable synthesis of specific chemical compounds, followed by a chemical analysis, usually by chromatography. Finally, products have to be identified at the single atom or molecule level. All these steps need to proceed on-line, continuously and as fast as possible. The current technological limit reaches an overall separation time of one second and a production rate of about one atom per week of bombardment. The shortest-lived nuclide that has been used for chemical study has a half life of 0.5 s ($^{287}114$).

One obvious question to be answered is the reliability of any chemical information if it is based on single (or very few) atoms only. Chemical laws (*e.g.*, equilibrium constants, reaction rates, kinetic constants, *etc.*) describe the behaviour of a large number of educts. For few atoms, no information may be gained on macroscopic properties such as, *e.g.*, melting or boiling point, solid-state behaviour, colour of compounds or of the element, respectively. To overcome such restrictions, every single atom or molecule has to be subjected to multiple chemical steps. Chromatographic separations fulfil such requirements. In liquid or gaseous phase, atoms, ions or molecules are transported through columns filled with a stationary phase that subject the species to multiple interactions. In heavy element research most successful was gas adsorption chromatography. Species contained in a carrier gas are transported inside a column where they interact with the surface in form of adsorption/desorption processes. The interaction energy (adsorption enthalpy) defines the temperature-dependent adsorption time τ by a Frenkel-like equation according to

$$\tau_{ads} = \tau_0 \exp \frac{\Delta H_{ads}}{RT},$$

where τ_0 is a constant, R is the gas constant, T is the temperature and ΔH_{ads} is a compound-specific adsorption enthalpy on a given surface.

If this interaction is repeated many times along the column, the total transport time (retention time) yields a very unique signature of a given element or molecule. The trajectory of every atom or molecule is usually described by a Monte Carlo model.[8] Typical separation procedures that have been applied in heavy element chemistry subjected every single atom or molecule to roughly 10^{5} single surface interaction steps. The information that may be gained from the study of one single atom reflects then the same statistical accuracy as if 10^{5} atoms were subjected to one single chemical process.

Two gas adsorption chromatographic techniques have been mostly applied: isothermal chromatography and thermochromatography. Isothermal chromatography measures the transport time (retention time) of an atom or molecule through a column that is kept at a fixed temperature. Thermochromatography determines the deposition temperature of a volatile species inside a chromatography column along which a stationary negative temperature gradient is established. The volatile species are injected into the column from the high-temperature side. In both cases, from measured quantities (retention time as a function of the isothermal temperature or deposition temperature) adsorption enthalpies can be deduced. Based on empirical correlations, from such microscopic adsorption enthalpies, macroscopic thermochemical quantities such as sublimation enthalpies may be deduced.[9]

Finally, after chemical isolation unequivocal identification of separated products at the single atom level is mandatory. To achieve this goal, use is made of the well established nuclear physics fact that isotopes in this region of elements have rather unique radioactive decay properties: they emit α-particles with very high energies that form decay products, which also undergo α-emission. This leads to so-called decay chains, quite often ending in a final nuclide that decays by a spontaneous-fission process. Separated samples are therefore usually positioned between silicon semiconductor detector pairs to assay α-particle energies and the total kinetic energy of fission fragments in an event-by-event counting mode. In this way, the decay of every single atom results in an ensemble of information such as energies of the α-particles and of fission fragments as well as time correlations between consecutive decays. This makes identification of single atoms extremely reliable with typical error probabilities far below 1%.

In the following, two examples of recent studies are summarized, the first chemical investigation of bohrium (element 107, Bh) using isothermal gas chromatography and of element 112 (yet unnamed) applying thermochromatography. The first example describes a study where six atoms of bohrium were sufficient to prove its similarity with

other members of Group 7 of the Periodic Table such as technetium (Tc) and rhenium (Re) in form of a volatile molecule, the oxychloride. The second experiment proved the similarity of element 112 with mercury from the same Group of the Periodic Table on the basis of five atoms that also behaved as a very volatile noble metal.

II. Bohrium chemistry

In an experiment at the Philips cyclotron of the Paul Scherrer Institut (PSI), the first successful chemical isolation and identification of bohrium was accomplished.[10] A highly radioactive target of ^{249}Bk (670 μg cm^2) — supplied by the Lawrence Berkeley National Laboratory — covered with a thin layer of terbium was irradiated for about four weeks with typically 1.6×10^{12} particles of ^{22}Ne ions per second at a beam energy in the target of 118 MeV, producing ^{267}Bh, which has a half-life of 17 s in the reaction ^{249}Bk(^{22}Ne,4n). ^{169}Re, which has a half-life of 16.3 s, was simultaneously produced in the reaction with the terbium layer and served as a yield monitor for the chemical separation process. Nuclear reaction products recoiling from the target were attached to carbon aerosol clusters and transported with the carrier gas flow through a capillary to an on-line gas chromatography apparatus (OLGA;[11] Fig. 1). At the entrance to OLGA, the clusters were collected on a quartz filter heated with a pre-column oven to 1000 °C and subjected to a mixture of the reactive gases HCl and O_2. The reactive gases converted the carbon clusters to gaseous products, hereby releasing the attached atoms of bohrium, which then interacted with both reactive gases to form volatile oxychloride molecules. Thermodynamic modelling point to the molecule MO_3Cl, with M = Tc, Re or Bh, as the most stable compound to be formed under the experimental conditions.

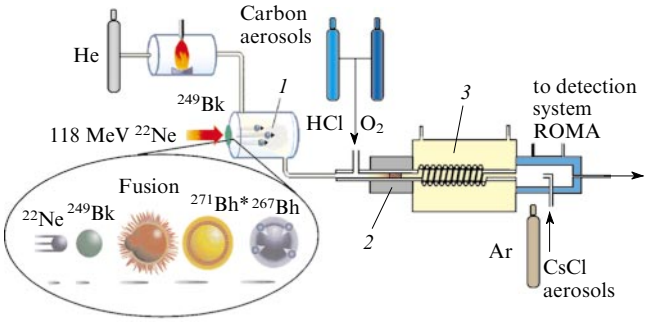


Figure 1. On-line isothermal Gas chromatography Apparatus (OLGA) used for the first chemical investigation of bohrium (element 107) at the Paul Scherrer Institute.
(*1*) Reaction products, (*2*) reaction oven, (*3*) chromatography column. Reaction products formed in fusion processes between a ^{22}Ne beam and the ^{249}Bk target are collected in a gas chamber, then attached to carbon clusters and finally transported to an oven system where volatile oxychloride molecules are formed. Behind the column products are re-attached to particles (CsCl) and transported to a counting system (ROMA) able to detect single atoms.

After chemical separation, *i.e.*, directly behind the isothermal column, products were attached to new aerosol particles and transported to a rotating wheel detection system where α-particle and spontaneous-fission (SF) decays were registered in a 4π counting geometry. Three experiments were performed at isothermal temperatures of 180, 150 and 75 °C, respectively. At each isothermal temperature, the same beam integral of 10^{18} ^{22}Ne particles was accumulated on the target. A total of six genetically linked decay chains attributed to the decay of ^{267}Bh were observed; four at an isothermal temperature of 180 °C, two at 150 °C and none at 75 °C. The averaged decay properties of the observed six decay chains are shown in Fig. 2.

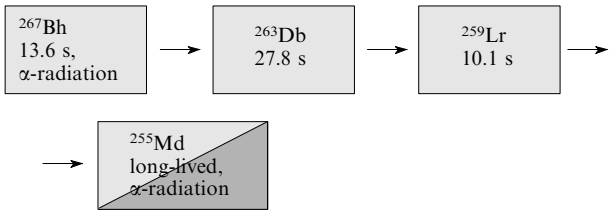


Figure 2. The average decay properties of the six observed atoms of bohrium (^{267}Bh) decaying *via* ^{263}Db and ^{259}Lr to the long-lived nuclide ^{255}Md (not detected). The times represent half-lives deduced from the measured life times.

Relative yields of the compounds $^{108}TcO_3Cl$ (measured in an independent experiment using ^{108}Tc with a half-life of 5.2 s), of $^{169}ReO_3Cl$ and of $^{267}BhO_3Cl$ (measured in the same experiment) as a function of isothermal temperature are shown in Fig. 3. The deduced enthalpies of adsorption ($-\Delta H^0_{ads}$) on the column surface are

Compound	$^{108}TcO_3Cl$	$^{169}ReO_3Cl$	$^{267}BhO_3Cl$
$-\Delta H^0_{ads}$ /kJ mol^{-1}	51 ± 3	61 ± 3	75^{+6}_{-9}

Therefore, the sequence in volatility is:[10]

$TcO_3Cl > ReO_3Cl > BhO_3Cl$.

This sequence in volatility agrees well with predictions from fully relativistic density-functional calculations for Group 7. The results of these calculations have shown that the electronic structure of BhO_3Cl is very similar to that of TcO_3Cl or ReO_3Cl. Increasing dipole moments and electric dipole polarizabilities in the Group suggest a decreasing volatility in the sequence:[12] $TcO_3Cl > ReO_3Cl > BhO_3Cl$.

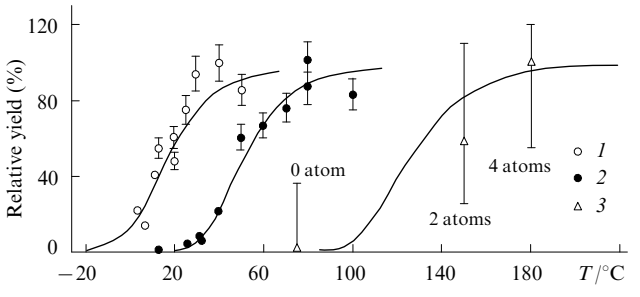


Figure 3. Relative yields of the compounds $^{108}TcO_3Cl$ (*1*), $^{169}ReO_3Cl$ (*2*) and $^{267}BhO_3Cl$ (*3*) as a function of isothermal temperature of the OLGA device.
The lines through the experimental data indicate the result of simulations with a Monte Carlo model[8] with the listed adsorption enthalpies.
Half-life /s: (*1*) 5.2, (*2*) 16, (*3*) 17;
$-H_{ads}$ /kJ mol^{-1}: (*1*) 51, (*2*) 62, (*3*) 75^{+9}_{-6}.

Also classical extrapolations down the Groups of the Periodic Table making use of empirical correlations of thermochemical properties predict BhO_3Cl to be more stable and less volatile than ReO_3Cl or TcO_3Cl.[9] The experimentally determined ΔH^0_{ads}-value can be used to estimate a macroscopic sublimation enthalpy (ΔH^0_{sub}) of BhO_3Cl using an empirical linear correlation function yielding $\Delta H^0_{sub}(BhO_3Cl) = 89^{+21}_{-18}$ kJ mol^{-1} (Ref. 9).

III. Chemistry with element 112

From the systematic order of the Periodic Table, element 112 is a representative of Group 12 together with zinc, cadmium and mercury and is expected to have a closed-shell electronic ground state configuration [Rn]5f^{14}6d^{10}7s^2. Thus, a noble metallic character can be expected. However, relativistic calculations of atomic properties suggest a significant contraction of the spherical 7s-electron orbital and endow element 112 with inertness much similar to that of the noble gas radon[6] although recent relativistic calculations predicted that it should form a semiconductor-like solid.[13] These diverse predictions require experiments able to distinguish between a noble volatile metal and a noble-gas-like behaviour. For this purpose, an experimental study was performed using the thermochromatography technique. Products formed in fusion reactions between ^{48}Ca and an actinide target were collected in a recoil chamber that was continuously flushed with a He + Ar gas to transport volatile species at room temperatures along a Teflon capillary to a chromatographic column. This column consisted of an array of 32 silicon detector pairs, covered by a thin gold layer and each pair kept at a different temperature covering the range between + 35 °C and − 185 °C, respectively (Fig. 4). The carrier gas containing the atoms of element 112 was flowing between the detectors mounted opposite to each other forming a rectangular channel. Hence, the detectors served as chemical sensors since the separation occurred directly on their surface.

In two experiments using the ^{48}Ca + ^{238}U reaction that forms element 112 in a complete fusion process with a total beam dose of 5.9×10^{18} , no decay of an element 112 atom (*i.e.*, of ^{283}112 with a half-life of 4s) was observed at a cross section limit of 1.3 pb.[14] This sensitivity was not sufficient to be in disagreement with physics experiments performed at FLNR in Dubna [15] and at GSI in Darmstadt,[16] both able

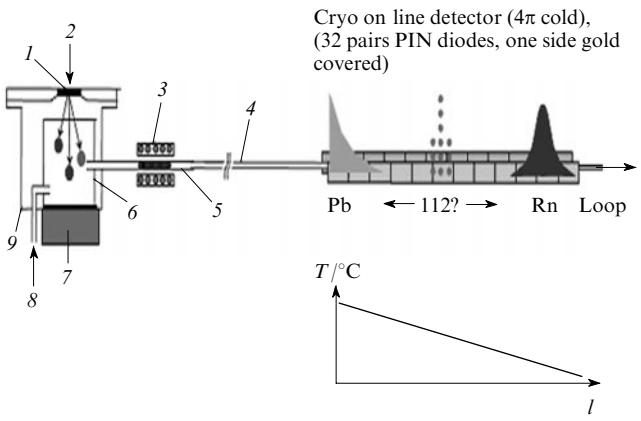


Figure 4. Thermochromatography device (IVO) (*in situ* Volatilisation and on-line detection) used for the first chemical study of element 112 at the Flerov Laboratory of Nuclear Reactions (FLNR) in Dubna.
(*1*) Window/target, (*2*) ^{48}Ca beam, (*3*) SiO_2-filter, (*4*) teflon capillary, (*5*) quartz column, (*6*) quartz inlay, (*7*) beam stop, (*8*) carrier gas, (*9*) recoil chamber.
Recoiling fusion products are transported in a carrier gas to the detector array along which a stationary negative temperature gradient is established.

to successfully synthesize this isotope with an average cross section of about 1.5 pb.[15] It was then decided to produce element 112 in an 'overshoot' reaction where the synthesized nuclide was a very short-lived isotope of element 114 that decayed to element 112 *via* α-emission prior to reaching the detector array after about 2s. The choice was ^{48}Ca + ^{242}Pu, because in this reaction the isotope ^{287}114 with a half-life of 0.5 s is produced with a cross section of about 4 pb.[15] This nuclide decays by α-emission to ^{283}112.

In the course of two bombardments, each lasting three weeks — during which a total beam dose: 6.2×10^{18} ^{48}Ca particles was accumulated on the ^{242}Pu target — five genetically linked α-SF decay chains were observed (Fig. 5). The detected atoms of element 112 on the gold surface were found at very low temperatures between − 5 °C and − 39 °C, respectively. This deposition pattern could be modelled with a species that interacts with an adsorption enthalpy on Au of -52^{+20}_{-4} kJ mol^{-1} (Refs 17 and 18).

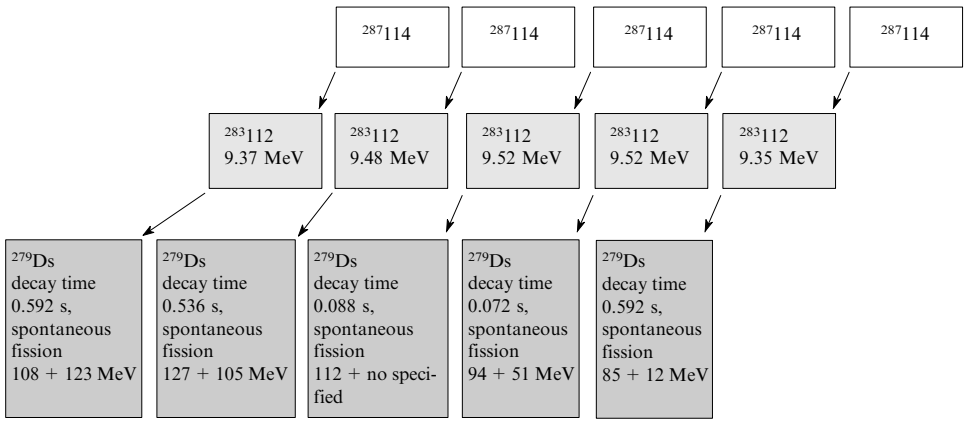


Figure 5. Decay chains from element 112 observed in two chemistry experiments performed in 2006 and 2007 at the Flerov Laboratory of Nuclear Reactions (FLNR) in Dubna using the ^{48}Ca + ^{242}Pu reaction at a total beam dose of 6.2×10^{18}.

Applying an empirical correlation between adsorption enthalpies of s- and p-elements at the single atom level on a gold surface with their respective sublimation enthalpies yielded a sublimation enthalpy for element 112 of 39^{+23}_{-10} kJ mol^{-1}. This value is in line with an extrapolation within Group 12 (Fig. 6). It clearly points to a still metallic and not to a noble gas-like behaviour of element 112. One might speculate that macro amounts of element 112 are gaseous under standard conditions, due to the very low sublimation enthalpy.

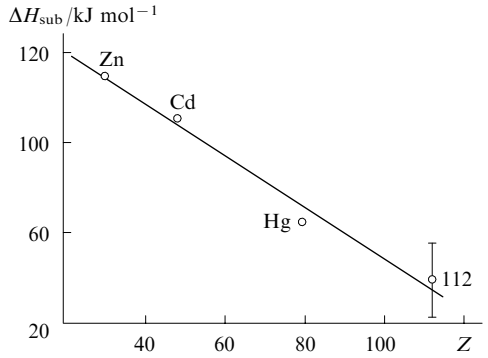


Figure 6. Sublimation enthalpies for Zn, Cd and Hg from the literature and empirically deduced sublimation enthalpy for element 112 from its measured deposition temperatures on Au.

This successful investigation of element 112 was the first time chemists have reached the island of superheavy elements that exists due to nuclear shell effects at the proton number 114 and the neutron number 184 (see Fig. 7).

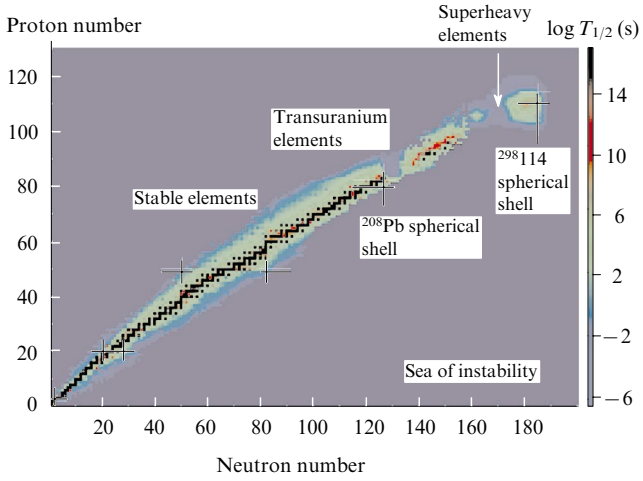


Figure 7. Chart of nuclides with the island of superheavy elements at the upper right corner (Courtesy: Yu Ts Oganessian). The arrow points to the nuclide used for the first chemical study of element 112.

IV. Conclusion

Currently, experiments are underway to proceed to even heavier elements, hence, to study chemical properties of 7p-elements. In a first step, the focus is on element 114. Indeed, in recent experiments performed in 2007–2008 at the Flerov Laboratory of Nuclear Reactions in Dubna, using the fusion reactions ^{48}Ca + ^{242}Pu and ^{244}Pu indication for the formation of one atom of ^{287}114 ($T_{1/2}$ = 0.5 s) and two atoms of ^{288}114 ($T_{1/2}$ = 0.8 s), respectively, were found applying an improved, faster IVO set-up.[19] Surprisingly, these three atoms behaved even more volatile than element 112 in the same set-up used for the chemical study of element 112, in support of the early predictions[6] and modeled in Ref. 20. This observation is, however, quite unexpected according to more recent predictions[21,22] and points to non-lead-like behaviour of element 114, probably caused by very strong relativistic effects on the spherical $7s^2$ and $7p^2_{1/2}$ shells efficiently shielding the filled $6d^{10}$ shell.

Due to the current limit of approximately one second separation time, nuclides with half-lives down to about 0.5 s are accessible to chemical study. Actually known isotopes for 7p elements show that only element 113 remains accessible using the isotope ^{284}113 with a half-life of 0.5 s. However, still unexplored elements above atomic number 108 (hassium) (the last chemically identified transactinide besides element 112) are meitnerium (Mt; Z = 109), darmstadtium (Ds; Z = 110) and roentgenium (Rg; Z = 111). For these elements, the presently known longest-lived isotopes are ^{276}Mt ($T_{1/2}$ = 0.7 s), ^{281}Ds ($T_{1/2}$ = 9.6 s) or ^{280}Rg ($T_{1/2}$ = 3.6 s). All these nuclides have been discovered in reactions with ^{48}Ca beams on actinide targets at the Flerov Laboratory of Nuclear Reactions in Dubna.

Very fascinating would be to enter the element region where the 5g electron shell starts to be filled. This is predicted to begin with element 125.[23] Currently it is speculative, whether isotopes of such elements exist with half-lives sufficiently long for chemical study. Recent theoretical calculations point to a much more extended island of superheavy elements that is not restricted to the magic proton shell at Z = 114 but exhibits also shell closures at Z = 120, 122 or even Z = 126 (see, *e.g.*, Ref. 24).

Finally, given the current limitations caused by the very low production rates — reaching actually the level of about one atom per month of beam time for the heaviest elements — it is mandatory to invest in accelerator and target technology. Needed are next generation heavy ion accelerators that deliver DC beams of utmost intensity, possibly in the mA range. As a consequence, improved target technologies need to be developed that can withstand such high intensity bombardments.

The results described in this manuscript have been obtained in collaborations of nuclear chemistry groups mainly from the Paul Scherrer Institute (Villigen), Bern University, Gesellschaft für Schwerionenforschung (Darmstadt), Lawrence Berkeley National Laboratory, Oak Ridge National Laboratory, Lawrence Livermore National Laboratory and Flerov Laboratory of Nuclear Reactions (Dubna).

Delivery of high intensity ion beams of ^{22}Ne and of ^{48}Ca by the operators of the Philips cyclotron at PSI and the 4m cyclotron at FLNR, respectively, is highly appreciated.

Careful reading of the manuscript by Robert Eichler is highly acknowledged.

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