

# Superheavy element chemistry at GSI – status and perspectives

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**Abstract.** Superheavy elements have been synthesized and chemically characterized one-atom-at-a-time up to element 108. Presently, the quest for identification and investigation of element 112 is one of the hottest topics in this field. The transactinide elements 104 to 108 are members of group 4 to 8 of the periodic table and element 112 belongs into group 12. For some of these elements detailed chemical properties have been revealed which show stunning deviations from simple extrapolations within their respective group while others exhibit great similarities with their lighter homologues. All presently known chemical properties of seaborgium (Sg, element 106) — the heaviest element whose behavior was investigated in aqueous solution — and hassium (Hs, element 108) were obtained in experiments performed at the GSI in large international collaborations. Recently, the highly efficient and very clean separation of Hs was applied for nuclear studies of various Hs nuclides investigating their cross section and their nuclear decay properties in the region of the  $N = 162$  neutron shell. To overcome certain limitations of the presently used on-line chemical separations the new TransActinide Separator and Chemistry Apparatus (TASCA) — with a gas-filled recoil separator as a front-end tool — was designed and built at the GSI in a collaborative effort. Presently in its commissioning phase, TASCA shall be a key instrument for a big leap into quantitatively and qualitatively new experiments in the region of superheavy elements.

**PACS.** 27.90.+b  $220 \leq A$  – 25.70.-z Low and intermediate energy heavy-ion reactions – 25.70.Jj Fusion and fusion-fission reactions – 23.60.+e Alpha decay

## 1 Introduction

The number of chemical elements has increased considerably over the last decades. But how many of these elements are characterized chemically to allow comparing their chemical properties with expectations (i) derived from their lighter homologues in the periodic table or (ii) from theoretical model calculations? To what extent are these chemical properties affected by increasingly strong relativistic effects? These are key questions chemists like to answer in addition to the central question of how many elements can exist. Already the first pioneering chemical studies showed that rutherfordium and dubnium (Rf, element 104, and Db, element 105, respectively) belong into group 4 and 5 of the periodic table. These elements, which can only be produced one-atom-at-a-time in heavy-ion reactions, mark the beginning of a remarkable series of chemical elements: from a chemical point of view they are transactinide elements and from a nuclear point of view they can be called superheavy elements (SHE) [1]. It was only in recent years, that a large number of experiments did not only shed light on many fascinating and unexpected chemical properties of Rf and Db but allowed stepwise climbing up the exceedingly difficult path to the

subsequent elements Sg, Bh and Hs; see references [1–4] for a comprehensive coverage of this development.

Most chemical experiments of lighter transactinides depend on the cluster (aerosol) jet-transport. This technique was applied to investigate Rf (see [5,6] and references therein for a large number of very detailed, recent Rf studies), Db and Sg in aqueous solution. For Sg, the heaviest element investigated in aqueous solution [7,8], two studies were performed at GSI. They will be described in an exemplary way. Contrary to the transport of lighter transactinides with a cluster jet volatile compounds of hassium (Hs, element 108) are directly formed with the reactive gas oxygen in the recoil chamber and are transported to the chemistry/detector device. This unique approach allowed unprecedented chemical studies of Hs [9,10] and yielded exciting nuclear results for the most neutron-rich Hs isotopes and their daughter nuclides. Chemical and nuclear aspects of Hs studies will be discussed in more detail. Most recently the quest for element 112 and beyond became one of the hottest and most challenging topics in heavy element research with chemical methods [11–13]. For chemists, this big leap from Hs to element 112 is attractive not only because nuclides became available (see Sect. 2) with presumably similar half-lives and cross sections as exploited in the Hs studies but also due to

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The figure shows a periodic table of elements. The main body of the table is arranged in 7 rows and 18 columns. The columns are numbered 1 to 18. The elements are arranged in order of increasing atomic number. The first row contains H (1) and He (2). The second row contains Li (3), Be (4), B (5), C (6), N (7), O (8), F (9), and Ne (10). The third row contains Na (11), Mg (12), Al (13), Si (14), P (15), S (16), Cl (17), and Ar (18). The fourth row contains K (19), Ca (20), Sc (21), Ti (22), V (23), Cr (24), Mn (25), Fe (26), Co (27), Ni (28), Cu (29), Zn (30), Ga (31), Ge (32), As (33), Se (34), Br (35), and Kr (36). The fifth row contains Rb (37), Sr (38), Y (39), Zr (40), Nb (41), Mo (42), Tc (43), Ru (44), Rh (45), Pd (46), Ag (47), Cd (48), In (49), Sn (50), Sb (51), Te (52), I (53), and Xe (54). The sixth row contains Cs (55), Ba (56), La\* (57), Hf (72), Ta (73), W (74), Re (75), Os (76), Ir (77), Pt (78), Au (79), Hg (80), Tl (81), Pb (82), Bi (83), Po (84), At (85), and Rn (86). The seventh row contains Fr (87), Ra (88), Ac+ (89), Rf (104), Db (105), Sg (106), Bh (107), Hs (108), Mt (109), Ds (110), Rg (111), 112, 113, 114, 115, 116, and 118. Below the main table, there are two rows of elements: the first row contains Th (90), Pa (91), U (92), Np (93), Pu (94), Am (95), Cm (96), Bk (97), Cf (98), Es (99), Fm (100), Md (101), No (102), and Lr (103); the second row contains Ce (58), Pr (59), Nd (60), Pm (61), Sm (62), Eu (63), Gd (64), Tb (65), Dy (66), Ho (67), Er (68), Tm (69), Yb (70), and Lu (71). The elements from Th to Lr are collectively labeled as 'Actinides' and 'Lanthanides'. The elements from Rf to 118 are collectively labeled as 'Transactinides = Superheavy Elements'.

**Fig. 1.** Periodic table of the elements. The IUPAC accepted transactinide elements 104 through 111 shall take the positions of the seventh period transition metals below Hf in group 4 and Au in group 11. While chemical studies have justified placing the elements Rf through Hs into group 4 to 8 of the periodic table, the “chemically unknown” heavier elements (chemical symbols for “known” elements and open symbols for yet unconfirmed reports) still need to be investigated. The arrangement of the actinides reflects that the first actinide elements still resemble, to a decreasing extent, the chemistry of *d*-elements.

a possibly strong influence of relativistic effects on the element 112 volatility (see Sect. 3), and the possibility to apply existing, successfully tested chemical separation techniques and detectors. Studies with element 112 constitute a transition from a more traditional approach in SHE chemistry — to form a chemical compound and investigate it chemically — towards physicochemical studies of an element which is present in the gas-phase in its atomic state and interacts with the wall material. The success in SHE chemistry was only feasible because of an enormous progress in many fields. Experimental developments, their scientific outcome and perspectives for SHE chemistry research will be discussed from a GSI’s point of view.

However, at the same time these very successful chemistry experiments demonstrated clearly the limits of these techniques; e.g. increasing beam intensities result in strongly decreasing transport yields of cluster jets. To move on to new frontiers, including heavier elements and so far inaccessible chemical systems, “physical” recoil separators are presumably ideal front-end “tools” for SHE chemistry. Ground-breaking experiments began at the Berkeley Gas-filled Separator (BGS) [14, 15]. A large international community is presently building and commissioning TASCA, the TransActinide Separator and Chemistry Apparatus, at the GSI [16]. Characteristic features of this gas-filled recoil separator, which will be very instrumental in chemical and in nuclear oriented experiments in the SHE region, will be outlined together with the discussion of perspectives of the SHE research.

## 2 Nuclear syntheses and properties of superheavy elements

Nuclides produced in heavy-ion reactions of medium-heavy projectiles with Pb and Bi targets lead to short-

lived nuclides which are, with one or two exceptions [14], usually not usable in chemical investigations. In contrast to these so-called “cold-fusion” reactions superheavy element chemistry experiments usually exploit an advantage of “hot-fusion” reactions which lead to the synthesis of the most neutron-rich, longest-lived isotopes of a given element. Usually the most neutron-rich available isotope of an actinide element, e.g.  $^{248}\text{Cm}$ , is irradiated with a neutron-rich light ion beam like  $^{18}\text{O}$ ,  $^{22}\text{Ne}$  or  $^{26}\text{Mg}$ . Such reactions produce SHE one-atom-at-a-time — decreasing from about one atom per minute for element 104 and 105, and a few atoms per hour for element 106, to about one atom per day for element 108 [1, 2]. This leads to the most extreme situation one can envision in chemistry: only single atoms or molecules, which are rapidly vanishing due to their radioactive decay, are at hand for chemical investigations. As all nuclides used and investigated in recent chemical studies of transactinides at GSI decay by emission of characteristic  $\alpha$ -particles a clear nuclear identification of the separated product was feasible after chemical isolation.

Most pleasantly, more elements and longer-lived nuclides are on the horizon and provide a challenge for chemical studies. In reactions between  $^{48}\text{Ca}$  as a projectile and targets made of  $^{233,238}\text{U}$ ,  $^{242}\text{Pu}$ ,  $^{248}\text{Cm}$  and  $^{249}\text{Cf}$  Oganessian et al. [17, 18] reported on the observation of elements up to 116 and of element 118. This opens up a thrilling region for heavy element chemists to probe the influence of increasingly strong relativistic effects and to test the structure of the periodic table at its extreme limit; see Figure 1. As all nuclear decay chains observed from these  $^{48}\text{Ca}$  induced reactions are terminated by spontaneous fission (SF) no “genetic” links into a region of known nuclides can be established. Therefore, an unambiguous identification of these new elements still remains open. Chemistry — in addition to unraveling exciting chemical

properties of these elements — may again become a crucial tool in the identification of a new element. First steps towards a chemical separation and identification of element 112 — directly produced or as a decay product of element 114 — have been made [11–13]. Those chemical experiments to search for  $^{283}112$  which were made by a large collaboration at the GSI made use of the  $^{48}\text{Ca}$  on  $^{238}\text{U}$  reaction [12]. They will be discussed in the next section together with chemical aspects of element 112.

In recent years, chemistry experiments at the GSI made use of  $^{22}\text{Ne}$  and  $^{26}\text{Mg}$  beams from the UNILAC to synthesize and investigate  $^{265}\text{Sg}$  and  $^{269,270,271}\text{Hs}$  in reactions with  $^{248}\text{Cm}$ . While the main emphasis in the Sg experiments was to unravel chemical properties of Sg [7, 8] recent Hs experiments exploited the, by now well established, unique and very clean Hs chemistry to study nuclear reaction and nuclear structure aspects [19]. A rotating  $^{248}\text{Cm}$  target wheel [1] was irradiated with high intense  $^{26}\text{Mg}$  beams at five different energies between  $E_{\text{lab}} = 130$  MeV and 150 MeV covering the range from below the maximum of the  $4n$  channel producing the new isotope  $^{270}\text{Hs}$  [19] to an energy well beyond the maximum of the  $5n$  channel ( $^{269}\text{Hs}$ ). In this energy range cross sections for  $^{269}\text{Hs}$  and  $^{270}\text{Hs}$  are in the range between 1 pb and 6 pb. This is in good agreement with calculations using the code HIVAP [20]. At the lower irradiation energies, surprisingly enough, evidence for the observation of  $^{271}\text{Hs}$  — produced in the  $3n$ -evaporation channel(!) — was obtained [19]. Most interesting, the decay of these Hs isotopes — their  $\alpha$ -decay energies and partial SF half-lives — allow for a crucial test of theoretical model predictions of the  $N = 162$  neutron-shell strength. The deduced  $Q_{\alpha}$ -value of 9.02 MeV for the doubly-magic  $^{270}\text{Hs}_{162}$  [19] is in good agreement with macroscopic-microscopic model predictions [21] showing clearly the shell stabilization at neutron number  $N = 162$ . In addition, also the decay properties of the Sg isotopes, observed as the Hs  $\alpha$ -decay daughter nuclides, provide important data for this comparison. The SF half-life of about 360 ms for  $^{266}\text{Sg}$  provides additional evidence for a shell stabilization around  $N = 162$ . Most recently, a  $^{25}\text{Mg}$  beam was used in a search for the unknown isotope  $^{268}\text{Hs}$ . The sensitivity of this experiment was well below one picobarn for an assumed  $^{268}\text{Hs}$  half-life of one second or longer.

### 3 Chemical properties of superheavy elements

After the first chemical studies have shown that Rf and Db are transactinides and member of group 4 and 5 of the periodic table (see Fig. 1) more detailed chemical studies were performed over the last two decades; see [1–4] for a comprehensive summary. It soon became clear that simple extrapolations of chemical properties within a given group of elements in the periodic table are not trustworthy. Especially Rf and Db exhibit chemical properties which are often very much different from extrapolations in group 4 and 5. Studies in the liquid phase, which concentrated on the aspects of hydrolysis and complex formation, were mainly performed in pure and mixed mineral acid solutions. First

detailed studies of Db in mixed HCl/HF solutions yielded a surprisingly non-Ta-like behavior [22]. Only subsequent experiments [23], which were carried out in pure acid solutions, showed a Db behavior in agreement with theoretical predictions [24] including relativistic effects. In recent years, the behavior of Rf was investigated in greater detail in the aqueous phase [5, 6, 25, 26] using first the Automated Rapid Chemistry Apparatus (ARCA) [27] and, most recently, the much more advanced “Automated Ion-exchange separation apparatus coupled with the Detection system for Alpha spectroscopy” (AIDA) [25, 28]. A number of surprising results were obtained; e.g., the fluoride complexation of Rf revealed stunning differences between Rf and both Zr and Hf [6]. These kinds of experimental findings still remain a challenge for theoretical model descriptions.

Aqueous chemistry with ARCA has reached Sg and two very basic chromatographic studies on cation-exchange resins were performed [7, 8, 29]. The one carried out with 0.1 M  $\text{HNO}_3$ /5  $\times 10^{-4}$  M HF showed that Sg elutes within 10 s from a 1.6  $\times$  8 mm cation-exchange column (Aminex A6, 17.5  $\pm$  2  $\mu\text{m}$ ). Therefore, under these conditions Sg behaves similarly to Mo and W in group 6 of the periodic table and distinctively different from hexavalent U which forms uranyl-ions [29]. This return to an expected behavior — after all the surprises with Rf and Db chemistry — was termed occasionally “oddly ordinary seaborgium”. However, this does NOT allow a statement like “seaborgium reaffirms periodic table’s predictive ability”. As not only relativistic effects but also other effects, like shell effects, on the atomic and molecular electrons can cancel each other with the result of surprisingly similar properties — or, in other cases, they can all act in such a direction that strong deviations from expected properties can be observed. Already the second Sg experiment [8], which was performed in pure 0.1 M  $\text{HNO}_3$ , indicated a non-W-like behavior of Sg. Contrary to W, Sg was not eluted from the cation-exchange column. From this, it was concluded that neutral or anionic oxofluoride complexes like  $\text{SgO}_2\text{F}_2$  or  $\text{SgO}_2\text{F}_3^-$  were likely to be formed and were eluted in the first experiment rather than  $\text{SgO}_4^{2-}$ . Without the presence of any complexing fluoride anions in the second experiment Sg was retained on the resin in cationic form, presumably, as  $\text{Sg}(\text{OH})_4(\text{H}_2\text{O})_2^{2+}$  or  $\text{SgO}(\text{OH})_3(\text{H}_2\text{O})_2^+$ . These species appear in dilute  $\text{HNO}_3$  due to the lower tendency of Sg to hydrolyze as compared with W [8].

The recent success in SHE chemistry in the liquid phase and in the gas phase was only feasible because of an enormous progress in many fields to meet a large number of physical, chemical, technological and metrological challenges. Intense and stable heavy ion beams became available and technologies were developed to stand high heat loads on windows, actinide targets and backings. In recent experiments at the GSI, e.g.,  $^{25}\text{Mg}$  and  $^{26}\text{Mg}$  beams were produced from the ECR-source with very low consumption of about 0.5 mg/h or even less of isotopically enriched material. This made stable beam intensities of up to 4  $\times 10^{13}$  particles per second available

at the experiment. Given is the “pulse” beam intensity in a 5-ms long macropuls with a 50 Hz repetition rate from the UNILAC which corresponds to about  $1.5 \mu A_{part}$  DC beam. With increasing beam intensities in the range of microamperes traditional He/cluster-jet transport systems reach their limits and become inefficient as transport yields sharply drop. Not only to circumvent these problems but to exploit a number of advantages, the formation of a volatile oxide directly in the recoil chamber and its transport to a combined chemistry and detector device [30,31] was applied in all Hs [9,10,19] experiments. In the past, gas-adsorption chromatographic studies of the lighter transactinides were mainly performed with volatile halide or oxohalide complexes. Apart from very few exceptions, after a cluster-jet transport these compounds were formed usually in a remote reaction chamber at the inlet of a chromatography oven; see [32] for a compilation.

Two kinds of gas-adsorption experiments have been performed with HsO<sub>4</sub> [9,10] both showing a typical group 8 behavior of Hs. Firstly and typical for its lighter homologue Os, all experiments demonstrated, as expected [33], the formation of a stable and at room temperature volatile tetroxide of Hs in an oxygen containing gaseous environment. However, in the thermochromatographic studies — in which deposition temperatures of Os and Hs compounds were measured by registering characteristic  $\alpha$ -decays in detector arrays mounted along a temperature gradient — a theoretically unexplained [33] higher adsorption temperature — or lower volatility — was measured for HsO<sub>4</sub> in comparison with OsO<sub>4</sub> [9]. An adsorption temperature of  $(-44 \pm 6)^\circ\text{C}$  was determined from the observation of seven HsO<sub>4</sub> molecules. From a comparison with the adsorption of OsO<sub>4</sub> at  $(-82 \pm 7)^\circ\text{C}$ , the following adsorption enthalpies ( $\Delta H_a^\circ$ ) on silicon nitride were deduced from the first Hs experiment [9]:  $-\Delta H_a^{\circ(T)}(\text{HsO}_4) = (46 \pm 2)$  kJ/mol for HsO<sub>4</sub> and  $-\Delta H_a^{\circ(T)}(\text{OsO}_4) = (39 \pm 1)$  kJ/mol for OsO<sub>4</sub>. Although the difference between theoretically predicted [33] and experimentally observed HsO<sub>4</sub> volatility is small on an absolute scale of adsorption temperatures or adsorption enthalpies it remains puzzling from a theoretical point of view why the inverse order of volatilities  $\text{HsO}_4 < \text{OsO}_4$  is observed experimentally as compared with the theoretical prediction  $\text{OsO}_4 \leq \text{HsO}_4$ . For the first time, Hs-chemistry experiment showed that a cross section limit below one picobarn can be reached in superheavy element chemistry — a crucial prerequisite to explore the chemistry of SHE in a wider region around element 114.

As in the preceding experiment, also in the second Hs experiment [10] tetroxides were formed in a recoil chamber and in its hot ( $600^\circ\text{C}$ ) outlet section. Contrary to the preceding experiment, which had to meet the challenge of using very dry gases to avoid early ice formation in the cold part of the detector, in this second experiment water was added (2 g H<sub>2</sub>O per kg gas) to the gas jet which contained a mixture of 1.1 L/min He and 0.1 L/min O<sub>2</sub>. These gases transported volatile products over a distance of about 13 m in PTFE capillaries within 3–4 s to a set of four detector boxes kept at room temperature. Each detector

box contained a linear array of four detectors (PIN-diodes) facing at a distance of 1 mm a stainless steel plate coated with a thin film of NaOH. Computer controlled, always three detector boxes in a row, i.e. 12 detectors, were measuring while the fourth box was refurbished and freshly prepared NaOH was mounted. The water in the transport gas maintained the chemical reactivity of the NaOH over the measuring period. The simultaneously produced and transported OsO<sub>4</sub> was used as a monitor. An osmate (VIII) is formed in the interaction of OsO<sub>4</sub> with the chemically reactive NaOH surface. More than 50% of the Os is found in front of detector one and the rest exhibits a significant tailing. Six decay chains of Hs were detected in the first five detectors. They were centered at detector number three [10]. The low statistics of the six Hs events does not allow drawing any conclusion about a possible lower reactivity of the HsO<sub>4</sub> as compared to OsO<sub>4</sub>. However, the observation (i) confirms the formation and stability of the volatile HsO<sub>4</sub> compound and (ii) shows the similarity in chemical reactivity between HsO<sub>4</sub> and OsO<sub>4</sub>. Presumably, the deposition of Hs is, in analogy to a well-known behavior of Os, the result of the formation of a hassate(VIII) according to:  $2\text{NaOH} + \text{HsO}_4 \rightarrow \text{Na}_2[\text{HsO}_4(\text{OH})_2]$ . In this experiment, for the first time, an acid-base chemical reaction was performed with the tetroxide of Hs.

Recent attempts to get a first glimpse of the chemistry of element 112 are most exciting and most challenging. Since the mid-70’s possible chemical properties of element 112 have attracted a lot of attention, mainly because of strong relativistic effects and the  $6d^{10}7s^2$  closed-shell configuration. Very early on, especially the question how closely element 112 would resemble the chemistry of Hg — its lighter homologue in group 12 — was in the focus (see [34] for a summary of early theoretical works) and a wide span of volatilities has been predicted for element 112. One group of predictions is based on theoretical model calculations including relativistic effects, see e.g. [34–36], while others rest on empirical extrapolations of chemical properties in the periodic table [37,38]. While earlier predictions didn’t exclude a highly volatile and chemically very inert element 112 which would very much resemble the behavior of an inert gas [35] more recent calculations are discussing a more Hg-like behavior but still with a higher volatility for element 112 [36]. A very recent fully-relativistic treatment of the interaction of element 112 with metallic surfaces of Au and Pd predicts for 112 a weaker adsorption as compared to Hg [36]. These quantitative, most advanced calculations predict that the adsorption temperature of element 112 on (ideal) Au surfaces will be 93 K below the one for Hg. In addition it is pointed out that element 112 will form some metal-metal bonding with Au and, therefore, element 112 will adsorb at much higher temperatures than Rn which is adsorbed by van der Waals forces only. Adsorption enthalpies of element 112 on metal surfaces obtained from an empirical model also indicate a weak chemical bond formed on Au surfaces and a “volatile noble metal” character of element 112 was predicted [37–39]. The element 112 volatility was expected to be much higher than that of Hg.

**Table 1.** Parameters and results of experiments to synthesize  $^{283}112$  in the  $^{48}\text{Ca} + ^{238}\text{U}$  reaction.

$E_{\text{proj}} / \text{MeV}$ (middle of target)	No. of events assigned to $^{283}112 / \alpha$ ; SF	$E_{\alpha} / \text{MeV}$ or SF	Technique <sup>i</sup>	$T_{1/2}$	$\sigma / \text{pb}$ ( <sup>a</sup> )	Ref.
231	0 ; 2	SF	VASSILISSA	(1.4 + 2.5 – 0.5) min	5.0 + 6.3 – 3.2	40
238	0 ; 0	–			<7.3	
231	0 ; 0	–	VASSILISSA		<2.2	41
234	0 ; 2	SF		(5.1 + 3.5 – 1.5) <sup>e</sup> min	3.0 + 4.0 – 2.0	
233	0 ; 7	SF	“Rn-like” chemistry	3 min assumed <sup>f</sup>	2.0 + 0.9 – 0.7	11
230	0 ; (1) <sup>g</sup>		DGFRS	3.4 s	0.5 + 1.2 – 0.4	17
234	4 ; (2) <sup>g</sup>	9.54		(4.0 + 1.3 – 0.7) s	2.5 + 1.8 – 1.1	
230	0 ; 0	–	BGS		<0.8 <sup>b</sup> , <1.6 <sup>c</sup>	43
236	0 ; 0	–			<0.96 <sup>b</sup> , <2.0 <sup>c</sup>	
233	0 ; 0	–			<0.8 <sup>b</sup>	42
236	0 ; (1) <sup>h</sup>	SF	SHIP	(5.2 + 25.1 – 2.4) <sup>h</sup> s	(0.7 + 1.6 – 0.6) <sup>h</sup>	
239	0 ; 0	–			<0.6 <sup>b</sup>	
231	– ; 0	–	“Hg-like”	5 min (SF) assumed <sup>f</sup>	<0.8 <sup>b</sup>	42
	– ; 0	–	“Rn-like”	5 min (SF) assumed <sup>f</sup>	<1.9 <sup>d</sup>	
235	0 ; 0	–	“Hg-like”	4 s ( $\alpha$ ) <sup>f</sup> , 5 min (SF) <sup>f</sup>	<2.3 <sup>d</sup> ; <1.7 <sup>d</sup>	
	0 ; 0	–	“Rn-like” chemistry	4 s ( $\alpha$ ) <sup>f</sup> , 5 min (SF) <sup>f</sup>	<2.7 <sup>d</sup> ; <7.6 <sup>d</sup>	

<sup>a</sup> Error bars and upper limit sensitivities at 68% c.i. if not otherwise specified. <sup>b</sup> “One-event” limits without statistical fluctuation.

<sup>c</sup> Upper limit sensitivity at 84% c.i. <sup>d</sup> Upper limit sensitivity at 95.45% c.i. <sup>e</sup> Mean value including previously observed events.

<sup>f</sup>  $T_{1/2}$  was not measured in this experiment; for cross section calculation the given  $T_{1/2}$  was assumed. <sup>g</sup> Interpreted as SF from the daughter nucleus after missing the  $\alpha$ -event from the mother decay. <sup>h</sup> No assignment was made to a specific isotope.

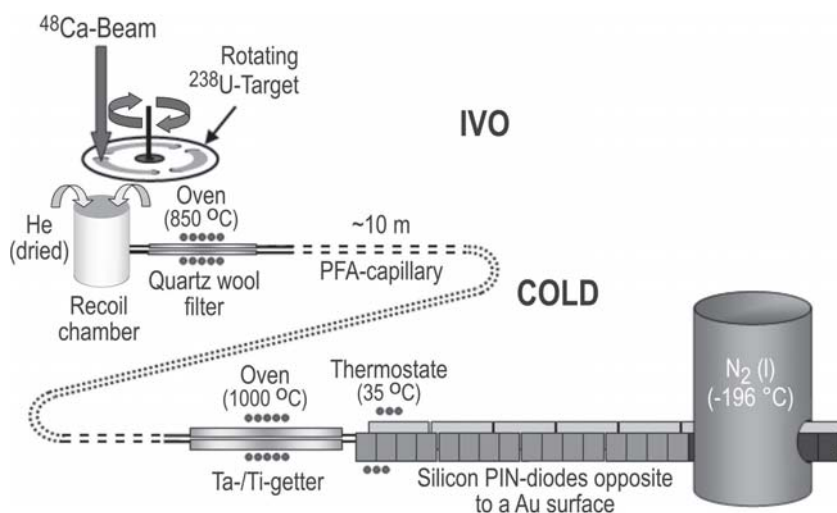
<sup>i</sup> The applied techniques comprise two vacuum recoil separators (the electrostatic separator VASSILISSA and the velocity filter SHIP), two gas-filled separators (DGFRS and BGS) and chemical techniques. For the latter ones, sensitivities were different for an assumed “Hg-like” and “Rn-like” behavior of element 112.

Recent experiments on element 112 [11, 12] constitute a transition from a more traditional approach in SHE chemistry — to form a chemical compound and chemically investigate it — to studies of an element in its atomic state. However, several experiments to study the volatility of element 112 in its atomic state gave partially controversial and inconclusive results [11, 12]. Still unpublished, a most recent press release [13] claims a successful chemical separation and identification of element 112. Unfortunately, a number of controversial reports [12, 17, 40–43] from different experiments (i) about the cross section to synthesize  $^{283}112$  in the  $^{48}\text{Ca}$  on  $^{238}\text{U}$  reaction and (ii) about the nuclear decay properties of  $^{283}112$  leave large parts of the scientific community in a state of confusion; see Table 1 for a summary.

After first chemistry experiments performed at the FLNR in Dubna [11] a large international collaboration carried out a second series of experiments started at the GSI [12]. Again, the reaction of  $^{48}\text{Ca}$  with  $^{238}\text{U}$  was selected to produce  $^{283}112$ . Simultaneously,  $^{220}\text{Rn}$  was produced as a transfer product from U and  $^{184-186}\text{Hg}$  from small amounts of Nd in the target. These experiments were aiming at measuring the adsorption behavior of element 112 in comparison with Hg and Rn on Au. Along the Au surface, a temperature gradient from +35 °C to

about –185 °C was established in a modified version of the Cryo On-line Detector (COLD), which was so successful in the Hs experiment [9]. A schematic view of this setup is shown in Figure 2. In the first somewhat slow GSI experiment, concentrating on a then believed 5-min SF decay for  $^{283}112$ , a Au-catcher was facing an array of 32 silicon PIN-diodes. Although some events were observed which could have been interpreted as SF from  $^{283}112$  small imperfections and open questions led the international collaboration agree to first repeat this experiment under improved conditions to substantiate the findings. An improved version was developed and was applied in the second experiment giving rise to much better measurements in (almost)  $4\pi$ -geometry [12]. Further improvements made the second experiment also much more sensitive to shorter half-lives in the region of a few seconds.

Also the second experiment, which, in addition to longer-lived SF decays, concentrated on shorter-lived  $\alpha$ -SF-decay sequences, did not detect any decay pattern which could be attributed unambiguously to a superheavy element. A broad range of adsorption properties between Hg and the noble gas Rn was covered. The upper limit sensitivity levels reached in both GSI experiments are listed in Table 1 for two different assumed decay modes and half-lives and for two different assumed volatilities. The levels



**Fig. 2.** Schematic view of the experimental setup used in the first experiment on element 112 at the GSI. Adapted from [1]. The target and recoil chamber arrangement to synthesize element 112 and to collect it in He gas is depicted in the upper left part followed by an oven section which serves as a filter to prevent cluster or aerosol material from passing through. This part is sometimes called IVO (In-situ Volatilization and On-line detection). At the end of the transport capillary a second oven further reduces the amount of oxygen and water in the transport gas before it enters the COLD at the warm end. At the other end liquid nitrogen is used as a coolant to create a negative temperature gradient along the segmented detector.

reached in these experiments are not low enough to clarify contradictions between previously published results on the cross section and on the decay of  $^{283}112$ .

#### 4 Perspectives

Many of the very successful experiments on the chemistry of SHE at the same time clearly demonstrated the limits of the applied techniques. A totally new quality of chemical studies is on the horizon when combining chemical techniques with gas-filled recoil separators as an ideal front-end tool not only to overcome the unwanted interaction of the primary heavy-ion beam with anything present in the recoil chamber but also to preseparate unwanted nuclear reaction products. Ground-breaking experiments began at the BGS [14,15]. The first Rf separation with the fast and continuously operating liquid-liquid extraction technique dubbed SISAK [14,44] clearly demonstrated this when SISAK was coupled to the BGS. Preseparated 4.7-s  $^{257}\text{Rf}$  was stopped in the gas of a Recoil Transfer Chamber (RTC) [15,31] — the key interface with a thin window between the gas filled separator and any chemistry setup — and was transported via a He/KCl-jet to the SISAK-system. The  $\alpha$ -decay of  $^{257}\text{Rf}$  was registered on-line with low resolution in a rather unspecific liquid-scintillator being part of the flowing organic phase. This is why only pre-separation enabled a clear Rf identification. The SISAK technique in combination with preseparation allows further improvements to study the extraction behavior of even heavier elements in aqueous solution. Under development are also alternative approaches like the use of crown ethers [45] to widen the scope of chemical investigations or to extend the studies of SHE in aqueous solution beyond Sg. To study these heavier elements electrochemical deposition techniques are in preparation. In addition, in the field of SHE chemistry previously unknown, completely new techniques and compounds will become accessible in gas-phase experiments; e.g. the use of organometallic compounds, when performing chemical reactions with pre-separated SHE to produce and investigate volatile species [46].



**Fig. 3.** (Color online) Photo of the gas-filled separator TASCA at the GSI showing (from the left) the differential pumping section which consists of two turbo pumps (below the two diagnosis chambers) and one roots pump (already installed but temporarily removed when this photo was taken), the target chamber, the dipole magnet and two quadrupole magnets.

Most recently, the TransActinide Separator and Chemistry Apparatus (TASCA) [47] was built at the GSI, see Figure 3, and a large international community is presently commissioning the separator and its peripheral components [47]. The goal of building TASCA was to provide the community with a gas-filled separator with maximized transmission (efficiency) for transactinide — or super-heavy element — research making use of hot-fusion reactions and actinide targets to produce the most neutron-rich, longest-lived nuclides. While chemical investigations of pre-separated SHE are in the focus and shall play a major role in the upcoming program a rich nuclear reaction and nuclear structure research is envisaged making use of TASCA. As it can be and will be operated with different gas fillings like, e.g.,  $\text{H}_2$  or  $\text{N}_2$  as an alternative to He as the standard gas, some basic research on the interaction of slow moving ions of the heaviest elements in different

gases and the determination of their charge states may become a most welcome side effect.

As the central components for the 3.5-m long gas-filled separator TASCAs we were making at least partially use of existing components from a former separator [48]. To optimize all separator components, new and old ones, ion-optical calculations were performed [49] based on the model fusion reaction of 5–6 MeV/u  $^{48}\text{Ca}$  on 0.5 mg/cm<sup>2</sup> actinide targets ( $^{238}\text{U}$ ,  $^{244}\text{Pu}$ ). The DQQ configuration with a 30°-degree dipole magnet (D) and a quadrupole doublet (QQ), which was selected as the best choice, allows operation in two possible modes. While the  $\text{DQ}_h\text{Q}_v$  mode gives the highest possible transmission, the smallest image size results from the  $\text{DQ}_v\text{Q}_h$  mode ( $\text{Q}_h$  and  $\text{Q}_v$  denote horizontally and vertically focusing quadrupoles). New ducts were designed, built and installed for the dipole and the quadrupoles to maximize the transmission in both modes. According to first model calculations [47,49], a horizontal and vertical acceptance of  $\pm 110$  and  $\pm 40$  mrad was achieved for the “high transmission”  $\text{DQ}_h\text{Q}_v$  mode. This corresponds to a solid angle of  $\approx 14$  msr which is at least 50% higher compared with the solid angle of the Dubna Gas-Filled Recoil Separator (DGFRS) which has a  $\approx 40\%$  transmission for the Ca on Pu reaction [17,50]. Based on this one can estimate that the TASCAs transmission should be about 50% higher than the DGFRS one; i.e. in the “high transmission” mode of TASCAs we are expecting about 60% transmission for evaporation residues produced in reactions like Ca on U or Pu. This is very similar to the best values given for the BGS [43]. TASCAs will combine two advantages of these two separators, i.e., the large transmission of the BGS and the small dispersion of the DGFRS. In the  $\text{DQ}_v\text{Q}_h$  mode, a reduced horizontal acceptance of  $\pm 34$  mrad will presumably reduce the transmission by a factor of  $\approx 0.6$ . However, an extraordinarily small image area should be achievable — a key issue to build small recoil transfer chambers for a fast transport of products into any chemistry setup and something which is unique for TASCAs [47,49].

To make use of the highest presently obtainable beam intensities at the UNILAC and the even higher intensities which will become available with the implementation of a new 28 GHz SC-ECR source in the near future, a windowless differential pumping section was installed and is successfully in operation [51]. A new target chamber was designed and built which accommodates (i) the rotating actinide target wheel in an easily removable cassette taking care of the safety aspects while working with highly radioactive material, (ii) the newly built drive, (iii) collimators, and (iv) beam diagnostic components. A beam current transformer upstream of the target shall allow for continuous non-destructive monitoring of the beam current. Equipped with a separator like TASCAs and combining it with all the tools which were developed over the last decade to study superheavy elements promises a big leap ahead and gives hope for a new quality in SHE research.

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