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Superheavy element studies with preseparated isotopes

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Summary. In recent years, significant progress in the field of superheavy element research has been achieved thanks to a novel combination of techniques from different fields. This "physical preseparation" approach includes the coupling of an ancillary setup – typically a chemistry apparatus or a counting setup – to a physical recoil separator. This latter preseparator removes unwanted nuclear reaction products as well as the intense heavy-ion beam associated with superheavy element experiments and thus isolates the evaporation residues of the nuclear fusion reactions. These are guided to the separators's focal plane, where they are extracted and available for further transport to external setups, *e.g.*, by a gas-jet. In this overview, the development of physical preseparation is described, and experimental results from nuclear chemistry and physics that were achieved with "preseparated" isotopes are summarized, with an emphasis on results relevant for superheavy element research. The covered topics range from chemical studies in the liquid as well as in the gas phase, the measurement of nuclear decay properties and of atomic masses. Preseparation was already shown to be a very powerful approach in these studies and promises to allow further progress in superheavy element research.

1. Chemical studies of superheavy elements – a brief introduction

Chemical experiments with the TransActiNide (TAN) elements, *i.e.*, the elements with $Z \geq 104$, often also referred to as the SuperHeavy Elements (SHEs) are an exciting topic within nuclear chemistry [1, 2]. One of the main aims of these experiments is to probe the structure of the periodic table in the region of the heaviest elements. Their chemical properties are most pronouncedly affected by ever stronger relativistic effects [3], which may give rise to properties that are differing from expectations based on simple extrapolations down the groups of the periodic table.

All elements up to hassium (Hs, $Z = 108$) [4] have been chemically studied, and recent reviews on this topic can be found in [1, 2]. The TANs from rutherfordium (Rf, $Z = 104$) to Hs are members of groups 4 through 8 of the periodic table. While they generally exhibit a behavior in general accordance with that of their nearest homologs, noteworthy exceptions have been found. These can only be reproduced by quantum chemical calculations that take relativistic effects into full account. In contrast, calculations that do not include relativity are not able to satisfactorily reproduce all obtained experimental results. Beyond Hs, the only element that had its chemical properties reproducibly studied is copernicium (Cn, $Z = 112$) [5, 6]. The interaction strength of Cn with Au was determined from the measured deposition temperature in thermochromatography experiments on an Au surface. Based on the obtained results, it is accepted as a noble metal, in line with its position in group 12 below Hg.

The heaviest element whose chemical behavior was experimentally investigated is element 114 (yet unnamed) [7, 8]. A first experiment [7] reported element 114 to be highly volatile and rather inert, somewhat in contrast to most of the contemporary predictions obtained at various levels of theory (see, *e.g.*, [9–14]). The results of this experiment, however, were discussed controversially [15].

Over the course of the past decades, a considerable amount of knowledge on the chemical properties of the TANs was acquired. All of these experiments face tremendous challenges such as small production rates – of the order of a few atoms per hour, day, or even week – and short lifetimes of even the longest-lived currently known isotopes, which are – with few exceptions – of the order of a few tens of seconds at most. Aside from these inherent difficulties, also technical reasons are responsible for the limited number of chemical systems studied to date. To understand this, two common techniques that were used in many past TAN experiments will briefly be described before preseparation, a new method, is introduced.

1.1 Thermalization and jet-transport directly behind the target

One commonly applied tool is to use an aerosol particle gas-jet [16, 17]. In this approach, nuclear reaction products are thermalized in a gas-filled volume located directly behind the target, the so-called recoil-chamber. However, not only the SHE of interest but also the majority of unwanted products (such as i) transfer products or ii) products of the interaction of the beam with the target assembly or impurities in the target) is thermalized in this volume. Due to the

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intense heavy-ion beam, a plasma is produced in the recoil chamber already at moderate beam intensities. The recoil chamber is flushed with an aerosol (usually He gas seeded with 10^6–10^7 aerosol particles/cm^3 of, *e.g.*, KCl, MoO_3, or C). Non-volatile reaction products attach to these particles, which are transported with the gas flow at high velocities, thus enabling efficient transport of the elements of interest (but also of unwanted reaction products) to chemistry setups often located outside the irradiation cave. An alternative approach [18] was successfully used, *e.g.*, in the chemical investigation of Hs in the form of HsO_4 [4] and subsequent nuclear chemical studies of the production and decay of Hs isotopes around deformed doubly-magic ^{270}Hs [19–22]. This exploited the formation of a highly volatile species in-situ in the recoil chamber and its transport to the chemistry setup in this form. This technique was shown to also be suitable and highly efficient for the very volatile elements Cn [5, 6] and most recently element 114 [7, 8], which were transported in their atomic state.

Both approaches have inherent disadvantages towards the chemical investigation of TAN elements: in the first approach, there is no selectivity between desired and unwanted reaction products, and a chemical system with a high separation factor for the element of interest is needed for its unambiguous identification. This identification is always based on the observation of its radioactive decay. This selectivity has to be favored over any other chemical property, and consequently, only relatively few suitable chemical systems are at hand. It is also well known that the yield of aerosol gas-jets drops significantly as the beam intensity reaches high levels (see, *e.g.*, Fig. 4 in [23]) due to the destruction of the aerosol particles in the plasma present in the recoil chamber. Moreover, both approaches involve very violent conditions with respect to, *e.g.*, the investigation of non-thermally stable compounds such as organometallic ones: in the first approach, high temperatures ($\sim 1000\,^{\circ}$C) are required for the destruction of the aerosol particles that then release the transported atoms, which allows their conversion into volatile species. In the second approach, a plasma is present in the recoil chamber as described earlier. Thus, neither of these techniques are particularly well suited to study such chemical systems.

These difficulties can be overcome by a novel combination of well-known methods: the coupling of (i) techniques well established in the field of SHE chemistry, such as the use of a gas-jet as discussed above, to (ii) a physical recoil separator, as exploited in a multitude of applications in many areas of low-energy nuclear physics, including SHE research. Setups comprising these two aspects have become well-known under the term "physical preseparation". Though still relatively new, short overview articles on preseparation can be found, see, *e.g.* [24, 25].

In Sect. 2, the pioneering work on preseparation at LBNL, Berkeley, CA, will be highlighted, and in Sect. 3, the TASCA project, which was conceived with the goal of building a highly efficient preseparator for studies of SHEs in the region of elements 112–118 produced in actinide target-based hot fusion reactions, will be discussed. Sect. 4 gives an overview on other recoil separators that were used as preseparators and the obtained results. A summary is contained in Sect. 5, together with a brief outlook.

2. Physical preseparation: the pioneering BGS work at LBNL

In the late 90s of the last century, the need for ever better background separation to advance to heavier and heavier elements became more and more apparent, and during the planning phase for setting up the Berkeley Gas-filled Separator (BGS) at the Lawrence Berkeley National Laboratory, the idea to extract EVaporation Residues (EVRs) of a nuclear fusion reaction from the physical recoil separator to make them available for further utilization was advocated [26]. With this, the idea of the coupling of external counting- or chemistry-setups to a physical recoil separator was born. The method can briefly be described as follows: the desired nuclear species are produced in a heavy-ion-induced fusion reaction and separated from the beam and the majority of the unwanted nuclear reaction products in a physical recoil separator. At its exit, they are extracted from the separator through a thin window and thermalized, *e.g.*, in a gas-filled volume which is referred to as the Recoil Transfer Chamber (RTC). A schematic of the BGS in the preseparator configuration is shown in Fig. 1.

As the BGS came online, this approach was pursued and an initial experiment [27] exploiting physical preseparation focused on the already well-established [18] oxidation of Os EVRs to the highly volatile tetroxide, its transport with pure He/O_2 gas, and, as a new technique, the deposition inside the "Cryo-Thermochromatographic Separator" (CTS). This is a detector array suitable for registering α-particles and fission fragments emitted from species deposited inside the channel.

2.1 Liquid–liquid extraction studies with SISAK

The very successful proof-of-principle study with the CTS led to the first application of physical preseparation to TAN elements very soon thereafter in liquid-liquid extraction experiments of rutherfordium (Rf, $Z = 104$) [28] exploiting the automated SISAK-system [29]. SISAK uses Liquid-Scintillation Counting (LSC) based detectors, and consequently, there is no need to produce dry samples suitable for measurements with solid-state detectors, which makes this system very fast and hence suitable for working with the relatively short-lived ^{257}Rf ($T_{1/2} \sim 5$ s [30–32]). This was produced in the ^{208}Pb(^{50}Ti, n) reaction, which yields energetic EVRs due to the rather heavy projectile, typical for cold fusion reactions. Such EVRs can easily penetrate also relatively thick RTC windows, like the 6-μm thick Mylar foils used in these experiments. On the other hand, the energy resolution of LSC systems is much poorer than that of solid-state detectors, which translates to the requirement that background suppression has to be relatively good to still allow the unambiguous identification of a TAN element produced at a small rate. The coupling of SISAK to the BGS resulted in very clean nuclear spectra and laid the ground for several further chemical studies of the first two TANs with SISAK [33–35]. The main interest in these experiments was to study the behavior of Rf in comparison with its lighter homologs Zr and Hf. These two elements exhibit similar chemical properties, which renders Rf experiments with the goal of differentiating between a more Zr-like *vs.* a more Hf-like behavior notoriously difficult. After the pilot experiment [28], which

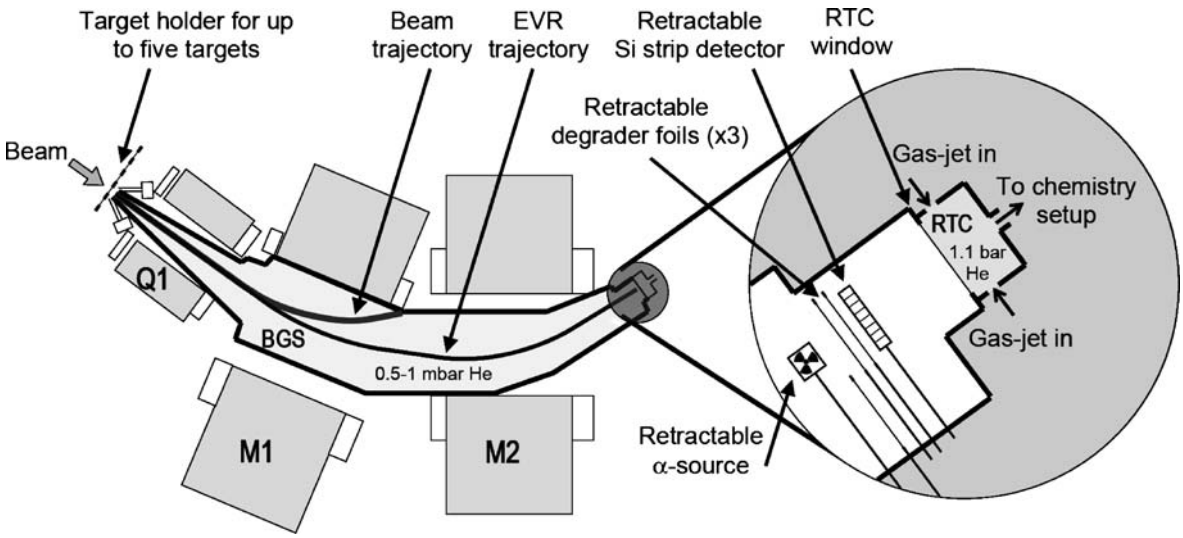

Fig. 1. Schematic of the BGS in its preseparator configuration. Reprinted figure with permission from Ch. E. Düllmann: Eur. Phys. J. D **75**, 76 (2007). © (2007) by EDP Sciences.

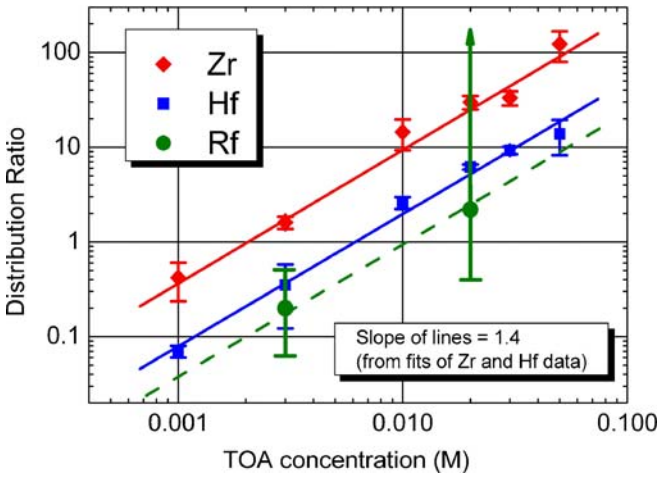

Fig. 2. Extraction of group-4 elements from sulphuric acid using tri-octylamine in toluene with SISAK. Reprinted figure with permission from J. P. Omtvedt *et al.*: Eur. Phys. J. D **75**, 95 (2007). © (2007) by EDP Sciences.

exploited the extraction of Rf from 6 M HNO_3 into 0.25 M dibutyl-phosphoric acid (HDBP) to demonstrate the capabilities of SISAK to unambiguously identify ^{257}Rf after preseparation in the BGS, an extraction system had been developed, in which Zr and Hf showed somewhat different behavior. It was based on the extraction of Zr/Hf into tri-iso-octyl amine (TIOA) from H_2SO_4. Rf was shown to extract to a lesser extent than Zr, and overall, the results, which are shown in Fig. 2, were in agreement with theoretical calculations, which predicted the extraction sequence Zr > Hf > Rf [36] under the given conditions. A final analysis of the experimental data will be given in [35].

2.2 Extraction of group 4 elements with crown ethers

A second set of experiments performed at the BGS was also dedicated to address the usually very similar chemical behavior of Zr and Hf in many chemical systems, and used preseparation to remove unwanted background in the BGS. Among the few chemical systems in which Zr and Hf behave differently, the extraction with crown-ethers, such as dibenzo-18-crown-6 (DB18C6), or dicyclohexano-18-crown-6 (DC18C6) is noteworthy. Extraction from hydrochloric acid into the crown ether diluted in dichloromethane was studied using short-lived preseparated Zr and Hf isotopes [37]. Optimum conditions for a future experiment with Rf were found using 8.5 M HCl as the aqueous phase and 0.025 M DC18C6 as the organic phase. The corresponding extraction yields were ∼ 28% for Hf and ∼ 84% for Zr. The extracted species consisted most likely of an ion-association complex formed between a Zr or Hf chloro-complex and a hydronium crown ether complex. Kinetic studies showed the complex formation and extraction to be completed after just 10 s, which makes this system suitable for studies with ^{261}Rf ($T_{1/2}$ ∼ 1 min).

2.3 Gas-phase chemical studies of group 4 *β*-diketonates

Besides the removal of undesired background activities, physical preseparation holds the promise to give access to studying SHE compound classes that were not previously accessible, such as organometallic ones. These were prohibited so far by the strong plasma in a recoil chamber directly behind the target, which leads to the instantaneous destruction of any organic compound being introduced into this volume (see, *e.g.*, [38]). Basic studies to verify that preseparation is an ideal solution to this problem were conducted with the relatively stable and volatile hexafluoroacetylacetonate (hfa) complexes using short-lived isotopes of Zr and Hf, the lighter homologs of Rf [39]. Their structure is shown in Fig. 3.

Suitable Zr and Hf isotopes were produced in ^{18}O- and ^{50}Ti-induced reactions. By exploiting a "cocktail beam", near-simultaneous studies of Zr and Hf were possible even though the used projectile was different. Besides switching of the beam, the target as well as the magnet settings of the BGS had to be changed, and the number of degrader foils was adjusted to match the kinetic energy and hence the recoil range of Hf to that of Zr EVRs. All of

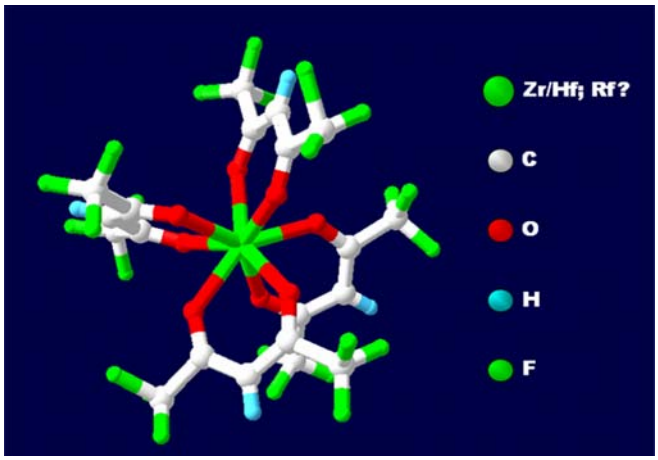

Fig. 3. Structure of $M(hfa)_4$ complexes (M = Zr, Hf). Figure courtesy, J. Faessler, UC Berkeley.

these changes were fast, which guaranteed the study of the two homologs under identical conditions. Nuclear reaction products were thermalized in the RTC, which was flushed with He enriched with the ligand, hfa. The reaction products exited the RTC through a quartz tube which contained a quartz wool plug that could be heated to temperatures above 600 °C. Volatile Zr and Hf species, most likely the well-known $M(hfa)_4$ (M = Zr, Hf) complexes, were synthesized in-situ, and their synthesis and decomposition was measured as a function of the temperature of the quartz wool plug, which served as the chemical reactor. From there, volatile species were transported with the carrier gas to various setups installed outside of the irradiation cave. The simplest setup consisted of activated charcoal filters that retained all transported Zr and Hf isotopes, the decay of which was measured with a γ detector. By comparing the activity with that implanted in an Al foil mounted directly behind the RTC window, absolute values of the transport yield were obtained.

The synthesis and decomposition of the volatile species was measured by varying the oven temperature. By using isotopes with different half-lives, the total time the EVRs spent between entering the RTC and their arrival in the filter – *i.e.*, the sum of the chemical reaction time and the transport time – was measured, and was found to be of the order of 1 min at temperatures above ∼ 150 °C, which is fast enough for experiments with ^{261}Rf. Above ∼ 450 °C, no transport of the metal complexes was observed, most likely due to decomposition of the free hfa ligand.

The separation of Hf from all other nuclear byproducts (i) in the BGS, and (ii) in the second step where only chemical species that were volatile under the given experimental conditions were transported to the charcoal filter, provided very clean samples that were suitable for measuring the half-life of ^{165}Hf. Conflicting values were reported in the literature (see discussion in [39]), and in the BGS/hfa-work, a value of (73.9 ± 0.85) s was obtained.

By varying the temperature of a 4-m long section of the perfluoroalkoxy (PFA) transport capillary from the RTC to the charcoal filter, isothermal chromatography experiments were performed and the adsorption enthalpy of $Hf(hfa)_4$ on PFA was evaluated to (−57 ± 3) kJ/mol. The measured breakthrough curve is displayed in Fig. 4.

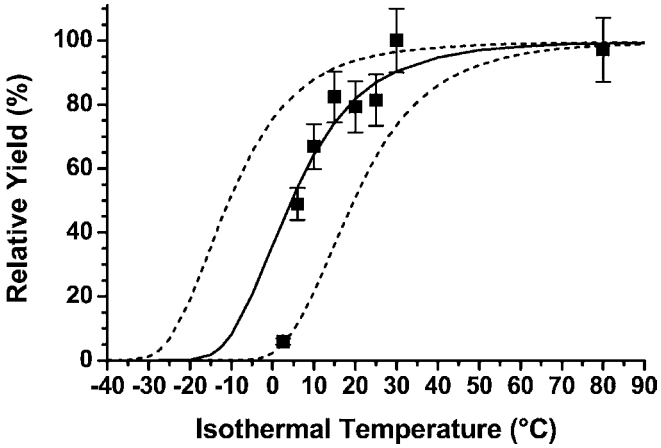

Fig. 4. Isothermal chromatogram of $^{162}Hf(hfa)_4$. A 4-m long isothermal section of a PFA Teflon tube served as isothermal chromatography column. The gas flow rate was 2100 mL/min. The solid line represents the result of a Monte Carlo simulation of the transport process with an adsorption enthalpy of −57 kJ/mol, the dashed lines indicate the 1σ limits of ±3 kJ/mol [39]. Reprinted with permission from Ch. E. Düllmann *et al.*: Radiochim. Acta **97**, 403–418 (2009); © 2009 Oldenbourg Wissenschaftsverlag GmbH.

These studies clearly showed that studies of new SHE compound classes are feasible using preseparated isotopes.

3. The TASCA project at GSI

3.1 Formation of the community, building and commissioning of TASCA

In 2002, scientists from various related fields gathered at the first "Workshop on Recoil Separator for Superheavy Element Chemistry" [40], which was mainly motivated by the realization that then current SHE chemistry techniques were likely close to be exhausted and threatened to prevent experimentalists from reaching elements heavier than bohrium (Bh, $Z = 107$) [41] or Hs [4], the heaviest ones chmically studied at that time. The requirements for the envisioned separator were a maximized efficiency for superheavy element chemistry and physics research using hot fusion reactions with actinide targets, thus giving access to the most neutron-rich and hence most long-lived isotopes of these elements. The separator should accept highest beam intensities, provide a relatively high background reduction, and a high suppression of the primary heavy-ion beam. A last requirement was that the RTC should be as small as possible, to allow for rapid flushing, which is necessary in light of the short half-lives of the isotopes of interest. Accordingly, the EVR image size in the focal plane of the separator had to be as small as possible. The model reaction chosen was 5–6 MeV/u ^{48}Ca on 0.5 mg/cm^2 actinide targets (mainly ^{238}U and ^{244}Pu).

The main workshop goals were to discuss and find the design of a recoil separator which fits best the needs for a preseparator coupled to superheavy element chemistry, and to establish an international community among interested experts in the fields of separator design, atomic and nuclear physics, and nuclear chemistry to design, build, test and operate such a recoil separator in combination with superheavy element chemistry. This was the birth of what became to be

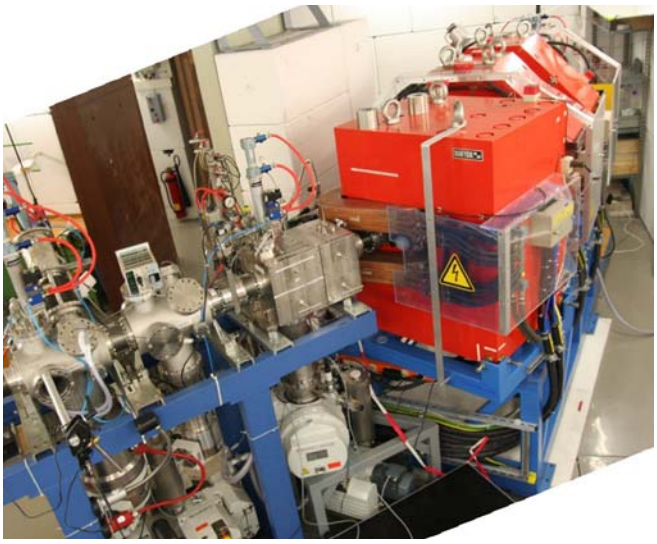

Fig. 5. Photograph of the TASCA gas-filled recoil separator at GSI.

known as the "TransActinide Separator and Chemistry Apparatus" (TASCA) and the TASCA collaboration. Further workshops were held and are nowadays held on an annual basis. In 2004, the collaboration decided to build a gas-filled separator, after analyzing also other possible designs (see, *e.g.*, [42]). Some parts were reused from the "NAchSEparator" (NASE), a DQQ-based separator with a 30° bending angle, which had been installed previously as a postseparator behind the "Separator for Heavy-Ion reaction Products" SHIP [43, 44]; sometimes, this device was referred to as the "HElium Charge-exchange Kaleidoscope" (HECK) [45]. The reused parts included mainly the three magents and two of the power supplies as well as the vacuum chamber housing the focal plane detector. However, despite the similar appearance of TASCA with the NASE (dominated by the three red magnets), changes and upgrades that were implemented are so substantial that the 3.5-m long TASCA separator is to be considered a new device. More details, partially given below, can be found in [46]. A photograph of TASCA is shown in Fig. 5.

To accept the high beam intensities available from the UNILAC, a windowless differential pumping section was installed, and a new target chamber was designed and built [47]. Recently, the target wheel was modernized and a four-segment wheel with 100 mm diameter, which will allow accepting even higher beam intensities, has been implemented [48]. The one key property that dominated the modifications was the maximization of the transmission. The magnetic fields were modeled, and a variety of magnet configurations using the existing dipole and two quadrupole magnets, but also configurations with additional elements were studied [49]. These design studies showed the classical DQQ-configuration to be the optimum choice, given that the originally implemented shims in the dipole magnet be removed, which led to a vertical gap about 10% larger than in NASE. This allowed for a vacuum chamber with a larger vertical opening and hence higher efficiency. The ducts and the vacuum chamber inside the magnets were redesigned: in the dipole magnet mainly to make use of the larger space available after removal of the shims and to extend the beam stop to a region outside of the magnet to provide better background suppression; in the quadrupole chamber, an innovative approach was taken: the chamber walls smoothly followed the pole shoes of the magnets, which led to a "butterfly-like" cross section of this chamber. These two modifications, together with optimized and slightly different positions of the target and of the three magnets relative to each other increased the transmission of TASCA compared to NASE significantly.

A unique feature of TASCA among all currently operated DQQ-separators is the possibility to revert the polarity of the quadrupole magnets, from the classical DQ_hQ_v, where the indices refer to vertical (v) and horizontal (h) focusing, to the DQ_vQ_h configuration. The DQ_hQ_v configuration provides maximum efficiency and is therefore referred to as the "High Transmission Mode" (HTM). The solid angle is 13.3 msr [49], which is significantly larger than, *e.g.*, the 10 msr of the Dubna Gas-Filled Recoil Separator (DGFRS). The efficiency of TASCA for actinide-target based ^{48}Ca-induced fusion reactions [50, 51] is roughly 50% larger than that of the DGFRS and approaches the best values reported from the BGS [52]. TASCA thus combines the advantages of these two separators, *i.e.*, the large transmission of the BGS and the small dispersion of the DGFRS. In the DQ_vQ_h configuration, EVRs are focused into a very small area of $\sim (30 \times 40)$ mm^2 in the focal plane, key to building small recoil transfer chambers for a fast transport of products into any chemistry setup. This comes at the cost of a somewhat reduced transmission. The solid angle in this "Small Image Mode" (SIM) is 4.3 msr, still comparable, *e.g.*, to the 3.3 msr of SHIP [53]. A recent overview of recoil separators used for SHE studies can be found in [54].

In 2006, an extensive commissioning program [55] started. In the course of about two years, the characteristics of TASCA were measured extensively and compared to ion-optical simulations [56]. A large variety of nuclear reactions, from very asymmetric ones such as $^{22}\mathrm{Ne} + ^{238}\mathrm{U}$ to more symmetric ones such as $^{54}\mathrm{Cr} + ^{\mathrm{nat}}\mathrm{Gd}$ leading to EVRs in the range of $Z = 76$–102 were used. A multitude of properties were studied extensively, among these the influence of the nature of the filling gas on the efficiency and suppression of different background components [57] or the dependence of the efficiency on target thickness and gas pressure. The studies were carried out in both, the HTM as well as the SIM. The efficiency of TASCA was also determined in a direct way by comparing the amount of EVRs implanted in a catcher foil mounted directly behind the target to that in a catcher foil located in the focal plane. The measurements were in very good agreement with the predictions according to [56] and fully confirmed the high efficiency of TASCA. In the final commissioning experiment, the "superheavies" that start with Rf [2] were reached when $^{260-262}$Rf were produced and studied in the reaction $^{238}\mathrm{U}(^{22}\mathrm{Ne}, 4\text{–}6n)$ [58].

Very much in the spirit of the first workshop, the building and commissioning of TASCA and its peripheral components was carried out by a large international community, led by the GSI nuclear chemistry department, and also the scientific program is executed by similar collaborations. This program [15] currently includes chemistry experiments exploiting TASCA as a preseparator, but also a rich nuclear physics program, with a current focus on the search for new

elements, the direct identification of the atomic number of nuclides produced in ^{48}Ca-induced fusion reactions, and nuclear structure studies towards ^{270}Hs [59]. Through studies of the atomic charges of ions moving through different fill gases, even atomic physics is touched upon [60].

3.2 Chemistry studies at TASCA

From the very start of the commissioning phase on, a working group was active to design, construct, and optimize two RTCs for both, the HTM as well as the SIM. Initial designs were based on the experience gained in the studies at the BGS [27, 39, 61]. To allow studying the influence of various parameters such as the depth of the RTC or the gas flow pattern (across the RTC window or across the depth of the RTC), modular designs were chosen [62, 63]. The first chemistry experiment performed behind TASCA focused on studying the underpotential deposition [64] of Os on a variety of metal surfaces [62, 65]. Inititally, it was planned to perform these experiments without preseparation, using a gas-jet transport of Os EVRs from a recoil chamber directly behind a target, and to identify the isotopes of interest with γ spectroscopy. However, only projectile-like fragments could be identified (see Fig. 6A in [62]). The chemistry setup was thus installed behind TASCA, and through removal of unwanted reaction products and the corresponding background reduction, the studies could successfully be performed. Critical potentials as well as the deposition kinetics were studied. The results showed, *e.g.*, that the adsorption of Os on Pd was significantly stronger than the adsorption of Os on Ni.

3.2.1 First transactinide chemistry behind TASCA: Rf fluoro complex studies with ARCA

The proof-of-principle experiment that demonstrated the readiness of TASCA for performing chemistry experiments with single atoms of TANs exploited a coupling of the Automated Rapid Chemistry Apparatus (ARCA) [66] to TASCA. The $\sim$ 1-min ^{261a}Rf was produced in the ^{244}Pu(^{22}Ne, $5n$) reaction with a cross section of several nb [67], preseparated in TASCA in the HTM – where the efficiency of TASCA to guide ^{261a}Rf produced in the asymmetric ^{22}Ne + ^{244}Pu reaction to the HTM RTC is slightly larger than 10% –, thermalized in the HTM RTC [62], and transported with a He/KCl gas-jet to ARCA. The formation of anionic fluoride complexes of Rf in dilute (7×10^{-4} and 1×10^{-3} M) HF was studied using anion-exchange chromatography [62]. Two elution fractions were collected and assayed for ^{261}Rf: a first one, eluted with the HF solution, and a second one where Rf adsorbed on the column was stripped with 5 M HNO_3. The α spectra showed seven events that were attributed to ^{261a}Rf or its daughter, ^{257}No. All of them were registered in fraction 2, from which lower limits (68% confidence interval) on the fraction of Rf adsorbed on the resin of 67.1% (7×10^{-4} M HF) and 74.5% (1×10^{-3} M HF) were deduced. While the chemical implications of this experiment were limited, the obtained α spectra were very clean (see Fig. 8 in [62]), and the experiment unambiguously demonstrated TAN chemistry studies with TASCA to be feasible.

3.2.2 Os liquid–liquid extraction studies with SISAK

Another set of experiments that has preparatory character towards a potential future experiment with Hs was recently performed [68]. The automated liquid-liquid extraction system SISAK was coupled to TASCA, and the distribution of Os between a NaOH and a toluene phase was studied. The chemical system was developed [69] using γ-ray emitting Os isotopes. The TASCA experiments served to proof that the combination of SISAK and TASCA, along with an improved liquid scintillation double-detector system (as given in Fig. 1C in [34]), is able to fulfil the requirements of a Hs experiment, *i.e.*, working with preseparated isotopes with short half-lives, and detection of α-decaying and spontaneously fissioning isotopes.

3.2.3 Gas phase chemical studies of element 114

The recent highlight chemistry experiment at TASCA was dedicated to the topic that stood at the very beginning of the TASCA project: the investigation of the most neutron-rich and hence most long-lived among the currently known isotopes of element 114, produced in a hot-fusion reaction with actinide targets. Studying the chemical properties of element 114 is among the hottest topics in current superheavy element chemistry. As detailed in Sect. 1, the heaviest element that had its chemical properties reproducibly studied is copernicium (Cn, element 112) [5, 6]. The importance of reproducibility of such studies cannot be overestimated. Results of previous experiments on the chemical properties of Cn, *e.g.*, could not be confirmed in follow-up studies (see, *e.g.*, [70–72]), in part due to insufficiently well-known (or wrong, as is clear nowadays) nuclear decay properties of the investigated isotopes. The chemical identification of element 114 was recently reported from an international collaboration working at the Flerov Laboratory of Nuclear Reactions (FLNR) in Dubna, Russia. Element 114 was reported to be much more volatile than its nearest lighter homolog in group 14, Pb. However, the main deduced chemical property, the adsorption enthalpy on a Au surface, was reported with an associated error large enough to render strong conclusions on the chemical behavior of element 114 impossible. Nevertheless, the data were interpreted to point to a noble gas-like behavior of element 114, which binds to a Au surface through a weak physisorption bond, rather than to a noble-metal like one, as it is predicted by most current theoretical works (see, *e.g.*, [9–14], but also [73] which contains predictions for a noble gas-like behavior). The experiment was not unanimously accepted by the scientific community [15]. As some of the raised criticism is connected to the background in the nuclear spectra of this experiment, which was performed without preseparation, an improved follow-up study was carried out under significantly improved conditions, exploiting the advantages of the TASCA facility.

As a first step towards a chemistry experiment with element 114 at TASCA, production and decay of element 114 in the ^{244}Pu(^{48}Ca, $3-4n$)^{288,289}114 reaction was studied [50, 51]. The obtained results confirmed the nuclear properties as reported from experiments at the DGFRS [74], and the experiment also verified the superior performance of TASCA for the subsequent chemical study of element 114. A fraction of the physics run was performed in the SIM [51],

because only this mode provides the fast flushing-time of the small RTC, which was a key requirement for experiments with ^{288,289}114 with their short half-lives (of the order of 1 s [50]). Two decay chains were observed [51] and proved the TASCA transmission even in the SIM to be high enough for a chemistry experiment.

The experiment was successful, and in total, two decay chains were observed that are attributed to element 114 and its daughters. The final analysis of the data is currently being carried out, and the results will be reported elsewhere [8].

3.2.4 Toward new compound classes of SHE

Recent experiments at TASCA explored the possibility to synthesize metal-organic compounds of *d*-elements, very much in the spirit of the BGS work with group 4 hfa-complexes [39]. The data are currently under analysis and will be published elsewhere [75].

4. Upgrades of existing recoil separators

Inspired by the prospects that preseparation holds for superheavy element research and intrigued by the simplicity of the approach, SHE groups working at laboratories where gas-filled recoil separators are installed and where an active chemistry program is pursued quickly adopted the novel combination of well-known methods. To this end, the GAs-filled Recoil Ion Separator (GARIS) [76, 77] installed at RIKEN, Wako, Japan as well as the DGFRS [78] were equipped with an exit window through which EVRs exit the separator and are available for further studies.

4.1 GARIS

The SHE group at RIKEN pursued a very systematic approach implementing preseparation at GARIS. Careful parameter studies were performed to explore optimum conditions for transporting preseparated EVRs with a KCl-based gas-jet to a rotating wheel-based "Measurement system for Alpha-particle and spontaneous fissioN events ONline" (MANON) [79] using various suitable isotopes such as ^{206}Fr, which can be produced with large cross sections, or ^{245}Fm, produced in the ^{40}Ar + ^{208}Pb reaction [23, 80]. The influence of, *e.g.*, the gas-flow rate or the temperature of the KCl-oven was studied, and the magnetic rigidity, $B \cdot \rho$, of EVRs was measured. The independence of the gas-jet yield from the beam intensity was demonstrated as well as the suppression of background in the measured spectra.

As a next step, the much more asymmetric ^{238}U(^{22}Ne, 5*n*)^{255}No reaction was exploited, which produces relatively slow EVRs with energies of only about 10 MeV, and hence a small range of only 2.2 µm in Mylar (extrapolated with SRIM [81] using the procedure outlined in [51]), which is traditionally used as the RTC window. A 1.1 ± 0.1 µm-thick Mylar window that is (i) thin enough for these EVRs to penetrate, (ii) able to withstand the pressure difference between the GARIS regime (∼ 1 mbar) and the RTC regime (up to ∼ 1 bar), (iii) tight enough for the pressure in GARIS to remain stable at the preset value was built. As done at the BGS [27] or at TASCA [62, 63], supporting grids with maximized geometrical transmission were used. The $B \cdot \rho$ of ^{255}No in dilute He was measured [82], which would have been difficult to achieve in a focal plane detector because in reactions induced by ^{22}Ne, the magnetic rigidities of the EVRs and of elastically scattered Target-Like Fragments (TLFs), which exit the target with twice the momentum of the beam, happen to be almost the same, which leads to a tremendous counting rate (due to the TLF) in the focal plane. Only a different choice of the filling gas remedies this problem, but overall, the use of preseparation – which is not sensitive towards high TLF count rates in the focal plane – is advantageous. While different parameterizations to estimate magnetic rigidities of EVRs in dilute He exist [83–85], the prediction of these for slow heavy ions such as ^{255}No produced in a ^{22}Ne-induced reaction is difficult, because of the observed non-linearity of the charge state trends towards lower ion velocities (see, *e.g.*, Fig. 5.13 in [86] or Fig. 1 in [87], where it becomes most apparent when the data shown in the figure are combined with higher-velocity data) in He. Based on both, the measured α spectrum as well as the half-life, the unambiguous identification of ^{255}No was demonstrated [82].

In following experiments, the asymmetric ^{248}Cm target-based reactions ^{248}Cm(^{18}O, 5*n*)^{261}Rf [88] and ^{248}Cm(^{22}Ne, 5*n*)^{265}Sg [89] were exploited to produce the isotopes of Rf and seaborgium (Sg, $Z = 106$) that are most suitable for experiments with preseparated isotopes. The experiments on ^{261}Rf focused on measurements on nuclear properties of this isotope. Originally discovered more than 40 years ago, evidence for the existence of a second, more short-lived state in ^{261}Rf was accumulated in the last about 10–15 years [19, 20, 90–92]. It was observed as the α-decay daughter of ^{265}Sg in decay chains passing through these nuclides. Only recently, evidence for the formation of this second state as an EVR in a heavy-ion induced fusion reaction was obtained [67], and the most complete and most precise values for its production and decay properties were obtained in an experiment at GARIS [93] where EVRs of the ^{248}Cm(^{18}O, 5*n*)^{261}Rf reaction were preseparated in GARIS and transported by a gas-jet to MANON. This second state (often referred to as ^{261b}Rf) decays by α decay (27 ± 6%) with an α-particle energy of 8.52 ± 0.05 MeV and by SF (73 ± 6%) with a half-life of 1.9 ± 0.4 s [93]. The production of either of the two states in ^{261}Rf is equally probable in the ^{248}Cm(^{18}O, 5*n*) reaction, the total cross section (for both states) was measured to be 23 ± 4 nb [93]. The most recent work at GARIS focused on ^{265}Sg [89], where it will be interesting to compare the directly measured decay data with the results of a meta-analysis [94, 95] of a large number of (different) experiments where only few events were observed in each single experiment.

4.2 DGFRS

As outlined in Sect. 3.2, chemical properties of element 114 were reported from a PSI-led collaboration who performed the experiment by applying a gas-jet directly behind ^{242,244}Pu targets, *i.e.*, without preseparation. Some aspects of this element 114 chemistry experiment were viewed critically [15], not least because of substantial background in the nuclear spectra [96, 97]. To overcome this problem, the PSI-led collaboration performed an experiment aiming at

studying element 114 behind the DGFRS [98]. As expected, the background was strongly reduced (see Fig. 7 in [98]). One α-SF chain was observed, which fits well with the known data of ^{285}Cn and ^{281}Ds. The observed chemical behavior could be explained by the transport and deposition of ^{285}Cn, and accordingly, the chain is assigned to most likely originate from Cn and not from element 114, the decay of which was missed.

It is noteworthy that among all chemistry experiments devoted to element 114 [7, 8, 98], those exploiting preseparation were able to conclusively observed decay-chains passing through long-lived ^{285}Cn ($T_{1/2} \sim 29$ s [50, 51, 99]) and its daughter, ^{281}Ds ($T_{1/2} \sim 13$ s [50, 51, 99]), while this was impossible in other experiments [7] because the chance for observing randomly correlated background-events over the long time period necessitated by the long half-lives of ^{285}Cn and ^{281}Ds was so large that a clear distinction between such "random correlations" and true correlations originating from ^{285}Cn and its daughter was impossible. Apparently, the most long-lived among the currently known isotopes of element 114, ^{289}114, with a half-life of $2.1^{+0.8}_{-0.4}$ s [50] (which was derived using data from [50, 99]) can only be used in studies with preseparation.

4.3 SHIPTRAP at SHIP

A slightly different system that uses a recoil separator to deliver clean EVR beams is the double Penning-trap system SHIPTRAP [100] installed behind the velocity filter SHIP [101] at GSI Darmstadt. SHIPTRAP is ideally suited to directly measure atomic masses of the heaviest elements with unprecedented precision. EVRs from the SHIP are thermalized in a buffer gas cell and guided to an extraction nozzle by electric fields. The concept is thus slightly different from RTC-based setups, the main idea behind the approach, however, is very similar. SHIP is a vacuum separator in which high electric fields are applied. Consequently, the vacuum window that separates the 10^{-6} mbar vacuum region in SHIP from the buffer gas cell regime has to fulfill very stringent requirements. These are much more relaxed in the case of He filled separators behind which He-filled RTCs are operated, and which accordingly tolerate some leaking. At SHIPTRAP, the window consists of a 2 mg/cm^2 thick Ti foil. EVRs extracted from the cell are bunched and injected into a first Penning trap, which provides isobaric separation, and then into a second Penning trap, in which the mass is determined with a precision of the order of 10^{-7} by measuring the ion's cyclotron frequency, which is directly related to its mass. The recent measurements of $^{252-254}$No [102, 103] represent the first direct mass measurements beyond uranium and pave the way for studies into the region of the SHE. A cyclotron resonance as obtained in these studies for ^{254}No is displayed in Fig. 6.

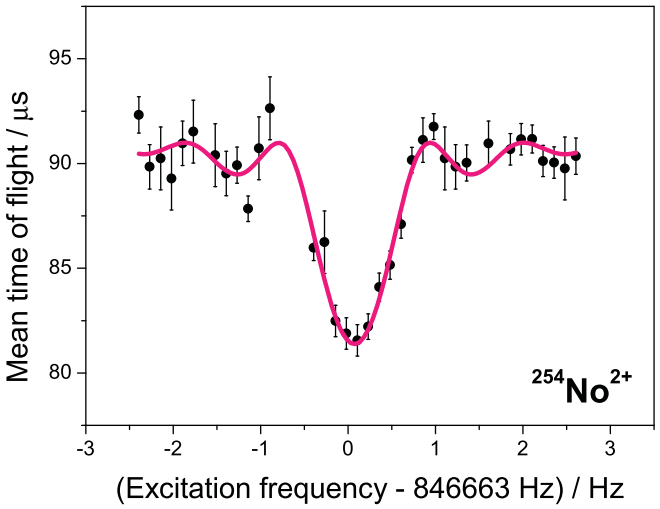

Fig. 6. Cyclotron resonance curve of $^{254}No^{2+}$. The solid line is a fit of the theoretical line shape to the experimental data (filled black circles). Error bars are one standard deviation.

Heavy element research is one of the strong pillars of the SHIPTRAP program, however, different topics from other fields like astrophysical nucleosynthesis (see, *e.g.*, [104]) are addressed as well using preseparated isotopes in a lighter mass region.

5. Results achieved thanks to preseparation and outlook

5.1 Results achieved with preseparated isotopes

As has been laid out in this overview, the idea to use a physical recoil separator as a preseparator was first discussed in 1997 [26] and the first report of an experiment performed with preseparation appeared in 2002 [27]. In the few years since then, preseparation has been implemented in most of the laboratories active in SHE research that feature a gas-filled recoil separator (LBNL, RIKEN, FLNR) and also at the vacuum device SHIP, and it was a strong aspect in the design of a new device, TASCA at the GSI Darmstadt. Experimental results cover a variety of topics, mainly from the field of nuclear chemistry but also from nuclear physics. They are summarized in Table 1.

Some of these studies are of methodical character exploring and verifying the potential that preseparation studies promise. The success of these clearly demonstrates that many exciting results can be expected to precipitate from studies that apply this technique.

5.2 Outlook

It is well conceivable that preseparated isotopes will play an important role in a variety of studies from different fields. Improvements of the method itself are still desirable, especially concerning the fundamental understanding of the ionic charge states of EVRs moving through dilute gases, which is still missing and forces experimenters to rely on semi-empirical systematics [60, 83].

As follows from what is given in the introduction, the elements meitnerium (Mt, $Z = 109$), darmstadtium (Ds, $Z = 110$), and roentgenium (Rg, $Z = 111$) have never been studied chemically to date. On the one hand, this is connected to the fact that suitable isotopes, *i.e.*, long-lived ones with $T_{1/2}$ of the order of at least 1 s, cannot be produced directly with cross sections promising for chemical studies. However, there are also reasons connected to the chemistry of the elements in the respective groups 9–11. This is more complex than that of the earlier TAN groups, and

Table 1. Summary of results obtained using physically preseparated isotopes. For a description and explanation of the used abbreviations for the preseparators and the chemistry and counting setups, see text.

	Element/isotope	Topic	Preseparator	Chemistry/ counting apparatus	Ref.
Physics	^{165}Hf	Measurement of $T_{1/2}$	BGS	γ-spectroscopy	[93]
	$^{252-254}$No	Direct mass measurements	SHIP	SHIPTRAP	[102]
	^{261b}Rf	Identification as EVR	TASCA	ROMA	[67]
	^{261b}Rf	Identification as EVR	GARIS		[93]
Chemistry	Os	Gas phase chemical study of OsO_4	BGS	CTS	[27]
	Os	Underpotential deposition	TASCA	Manual	[62, 65]
	Os	Oxo/hydroxo ion formation in liquid phase	TASCA	SISAK	[68]
	Various *d*-elements	Studies of new compound classes	TASCA	various	[75]
	Rf	Liquid–liquid extraction	BGS	SISAK	[28, 34]
	Rf	Fluoride complexation	TASCA	ARCA	[62]
	Cn	Gas phase chemical study	DGFRS	COLD	[98]
	Element 114	Gas phase chemical study	TASCA	COMPACT^2	[8]

suitable systems that are comparable, *e.g.*, to the tetroxide system in group 8, which was exploited extensively in chemical as well as nuclear physics studies of Hs, have not yet been identified. With the prospect of using preseparated isotopes, at least some of the constraints present in earlier approaches are now lifted and it remains to be seen if Mt, Ds, and Rg will be studied using preseparated isotopes.

The next heavier elements, Cn to element 118, are generally assumed, based on the systematics of the periodic table but also confirmed by theoretical calculations, to be rather volatile in their elemental state. This has been confirmed experimentally for Cn [5, 6] and element 114 [7, 8] using gas phase chromatographic techniques. Such techniques are likely applicable also to element 113, and potentially also to element 115, even though the half-life of the longest-lived known isotope of this element, $^{289}115$, is only $0.22^{+0.26}_{-0.08}$ s, based on the observation of five decays [105]. An alternative approach to gas phase chemical approaches is the use of vacuum chemistry techniques (see [73] and references therein) which are known to be much faster. The conversion of energetic EVRs (as they are available in the focal plane of the preseparator) into an atomic beam suitable for chemical adsorption studies remains one of the challenges of this approach. The implantation into a hot catcher and subsequent release appear most promising, and preliminary experiments have been performed at the BGS [106], but significant improvements will be necessary before this technique can be applied to SHE research.

Finally, the ground-breaking experiments at SHIPTRAP open up the perspective of direct mass measurements of single ions of SHE and laser spectroscopic studies of their atomic properties [107].

Presparated isotopes will thus play a key role in many future chemistry experiments with SHE, but also in other fields.

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