The Chemistry of Transactinide Elements — Experimental Achievements and Perspectives

M. Schädel[*]

Gesellschaft für Schwerionenforschung mbH, Planckstr. 1, D-64291 Darmstadt, Germany

The chemistry of transactinides and superheavy elements has reached element 108. Preparations are under way to leap to element 112 and beyond. This development, its current status and future perspectives are reviewed from an experimental point of view. The atom-at-a-time situation of transactinide chemistry is briefly outlined. Experimental techniques and important results enlightening the chemical properties of elements 104 through 108 are presented in an exemplary way with emphasis on the aqueous chemistry of the lighter ones. From the results of these experiments it is justified to place these elements in the Periodic Table of the Elements into groups 4 through 8, respectively. However, strongly due to the influence of relativistic effects, it is no longer possible to deduce detailed chemical properties of these superheavy elements from this position. Perspectives for future research programs are given.

1. Introduction

Today we know of 112 chemical elements.[1] The discoveries of elements 114[2–4] and 116[5,6] are currently waiting to be confirmed. Element 104, rutherfordium, Rf, marks the beginning of a remarkable series of chemical elements: From a nuclear point of view they can be called superheavy elements — as they only live because of their microscopic shell stabilization, see e.g. References 1,7, — and from a chemical point of view they are transactinide elements — as the series of actinides ends with element 103.[8] One of the most important and most interesting questions for a chemist is the one about the position of the transactinides in the Periodic Table of the Elements, PTE, see Figure 1.

From atomic calculations, see e.g. References 9–11, one expects the filling of the *6d* electron shell for the transactinides, and, consequently, a chemical behavior similar to the lighter transition metals in the fifth and sixth periods. However, it is by no means trivial to put rutherfordium into group 4 of the Periodic Table — and the heavier ones into successive groups — if one accepts that the Periodic Table as an ordering scheme also reflects the chemical property of an element at a given position.

Modern relativistic atomic and molecular calculations[10–12] clearly show the very large influence of direct and indirect relativistic effects on the energetic position and the sequence of electronic orbitals. This is also associated with changes in their radial distributions. All these changes are so pronounced that, if compared with non-relativistic calculations, significantly different chemical properties for the transactinides would not be surprising. Therefore, it is of great interest to study chemical properties of the transactinides in detail and to compare these with properties deduced from extrapolations and from modern relativistic molecular calculations in combination with empirical models. First generation experiments on rutherfordium[13–15] and element 105, dubnium, Db,[16,17] rendered enough justification to place Rf and Db into group 4 and 5 of the Periodic Table.

The chemistry of transactinide elements has now reached element 108. Preparations are under way to reach out for element 112 and beyond. These experiments have concentrated on the question how the chemical properties of the transactinides compare with the ones of their respective lighter homologs and how well the Periodic Table accommodates the transactinide elements as transition metals in the seventh period.

This contribution is focusing on recent key experiments (*i*) to unravel detailed chemical properties of elements 104 and 105 in aqueous solution, (*ii*) first survey experiments on the aqueous chemistry of element 106, seaborgium, Sg, and (*iii*) the first, successful experiments on element 107, bohrium, Bh, and

element 108, hassium, Hs, performed in the gas phase. It includes aspects of nuclear reactions, in an exemplary way experimental techniques, important experimental results enlightening the chemical properties of these elements, and perspectives. For comprehensive reviews of earlier and additional results, especially on the Rf, Db, and Sg chemistry, see, e.g., References 18–21.

2. Experimental Techniques

2.1. Nuclear Synthesis and Decay. Actinide target based heavy-ion reactions, frequently termed "hot-fusion" reactions, are used to synthesize the most neutron-rich and relatively long-lived isotopes of transactinides; see Table 1 for a compilation. Cross sections are in the range from a few nanobarns to about 10 nb for 78-s ^{261}Rf, 34-s ^{262}Db, and 27-s ^{263}Db, about 240 pb for 7.4-s ^{265}Sg, 70 pb for 17-s ^{267}Bh, and 6 pb for 9-s ^{269}Hs (Ref. 22–27). See Reference 28 for a recent, comprehensive discussion of ^{261}Rf and ^{262}Db cross sections. Except for the most recent Hs experiment, see below, setups were used, that tolerate beam intensities of approximately 3×10^{12} heavy ions per second on targets of about 0.8 mg/cm^2 thickness. This yields maximum production rates from about two or three atoms per minute for Rf and Db isotopes to five atoms per hour for ^{265}Sg and even less for ^{267}Bh and heavier nuclides. Therefore, all chemical separations on transactinides are performed

Figure 1. Periodic Table of the Elements. The known transactinide elements 104 through 112 shall take the positions of the seventh period transition metals below Hf in group 4 and Hg in group 12. While chemical studies have justified placing the elements Rf through Hs into the Periodic Table, the chemically "unknown" heavier elements 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: Th below group 4 elements Zr and Hf, Pa below Nb and Ta, and U below the group 6 elements Mo and W.

*Corresponding author. E-mail: m.schaedel@gsi.de. FAX: +49-6159-71-2903.

TABLE 1: Nuclides from "hot-fusion" nuclear reactions used in transactinide chemistry.

Nuclide	Half-life	Target	Projectile	Evap.	σ	Prod. rate[d]	Comment	Reference
^{261m}Rf	78 s	^{248}Cm	^{18}O	$5n$	$\approx$10 nb	2 min^{-1}	a	22, 28
		^{244}Pu	^{22}Ne	$5n$	4 nb	1 min^{-1}	c	29
^{262}Db	34 s	^{249}Bk	^{18}O	$5n$	6 nb	2 min^{-1}	a	23
		^{248}Cm	^{19}F	$5n$	1 nb	0.5 min^{-1}	b	28, 30
^{263}Db	27 s	^{249}Bk	^{18}O	$4n$	10 nb	3 min^{-1}	b	23
^{265}Sg	7.4 s	^{248}Cm	^{22}Ne	$5n$	$\approx$240 pb	5 h^{-1}	a	24
^{266}Sg	21 s	^{248}Cm	^{22}Ne	$4n$	$\approx$25 pb	0.5 h^{-1}	b	24
^{266}Bh	$\approx$1 s	^{249}Bk	^{22}Ne	$5n$	25–250 pb		b	25, 26
^{266}Bh	17 s	^{249}Bk	^{22}Ne	$4n$	$\approx$70 pb	1.5 h^{-1}	a	25, 26
^{269}Hs	9 s	^{248}Cm	^{26}Mg	$5n$	$\approx$6 pb	3 d^{-1}	a	27
^{270}Hs	2–7 s	^{248}Cm	^{26}Mg	$4n$	$\approx$4 pb	2 d^{-1}	b	27

[a]Reaction normally used in chemistry experiments.

[b]Reaction occasionally used or nuclide observed as a "by-product" or discovered in a radiochemical experiment.

[c]Measured with recoil separator.

[d]Assuming typical values of 0.8 mg/cm^2 target thickness and 3×10^{12} s^{-1} beam particles.

with single atoms on an "atom-at-a-time"[31] scale. Characteristic α decays, and, more significantly, time correlated α(mother)-α(daughter)-decay chains, are used to identify these isotopes in specific fractions or at characteristic positions after chemical separation.

2.2. Schematic for On-line Experiments

2.2.1. Production and Transport.   The atom-at-a-time situation of chemical separations with transactinides requires an optimization of the following parameters:

(*i*) Choice of the nuclear reaction — normally the one with the highest cross section.

(*ii*) High production rates — ion-source, accelerator, and target technology are cornerstones.

(*iii*) Fast transport and fast chemical separation — to avoid severe nuclear decay losses.

(*iv*) High yields in transport and in chemical separation.

(*v*) Efficient detection and identification.

(*vi*) A chemical separation procedure which fulfills these needs and, at the same time, provides the desired information about the chemical behavior of the transactinide element.

Many details of various experimental setups and detectors are described in References 18–20, 32.

A schematic presentation of components for automated on-line experiments with transactinides is shown in Figure 2. Heavy-ion beams are passing through a vacuum isolation window and target-backing before interacting with the actinide target material. Characteristic to all recent experiments on Rf through Bh was the use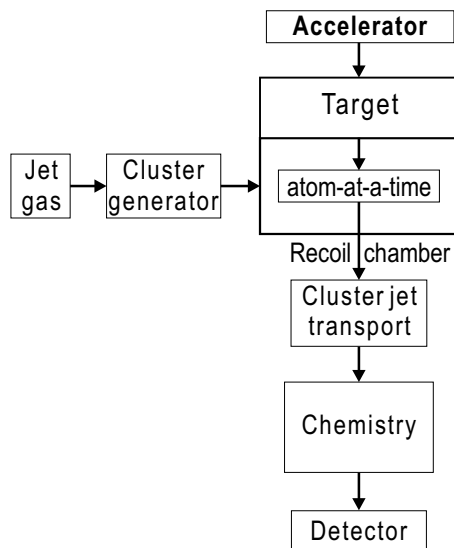 of aerosols ("clusters") as the carrier material for nuclear reaction products to be transported in the He-gas jet from the recoil chamber — where they are stopped in the gas — to the chemistry apparatus. Routinely, KCl is used as an aerosol material for separations carried out from the aqueous phase and carbon for gas chromatographic separations. Transport times of typically two to five seconds were achieved.

2.2.2. Liquid Phase Chemistry Setups.   So far, all liquid-phase separations to study the chemical behavior of transactinides were performed in a discontinuous, batch-wise operation with large numbers of cyclic repetitions. While in several experiments on Rf and Db manual procedures were used,[33] most transactinide separations were carried out with automated instruments as schematically shown in the upper part of Figure 3. The implementation of the **A**utomated **R**apid **C**hemistry **A**pparatus, **ARCA**,[34] yielded the predominant share of today's knowledge about the chemical behavior of the transactinide elements Rf through Sg in aqueous solution. ARCA II is micro-computer controlled and allows fast, repetitive, and reproducible chromatographic separations in a miniaturized and chemically inert liquid-chromatography system (8 mm long micro-columns of 1.6 mm i.d.) with typical cycle times between 45 s and 90 s. Depending on the chemistry, the columns were filled with cation or anion exchange resin or an organic extractant on an inert support material.

Common to all batch-wise separations are time consuming evaporation steps to prepare one or more samples for α spectroscopy and to measure spontaneous-fission fragments. Samples are dried by intense IR light and a stream of hot He gas. While separation times are normally between 5 s and 10 s, evaporating the aqueous phase requires about 20 s.

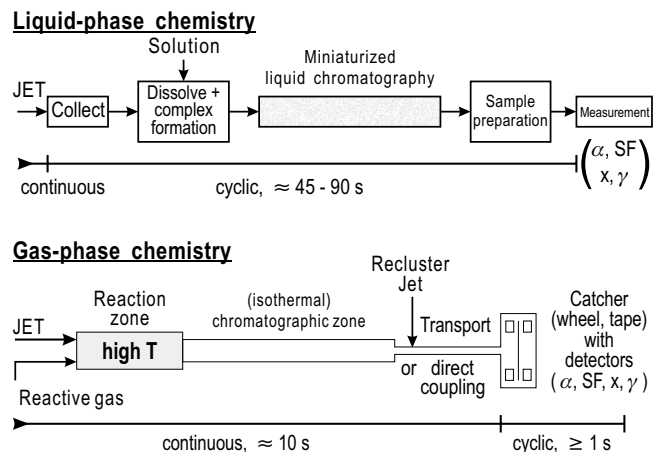A breakthrough concerning the automatization of the sam-

Figure 2.   Schematic presentation of components of recent on-line experiments with the transactinides Rf to Bh.

Figure 3.   Schematic flow chart of components for an automated on-line chemistry from liquid phase (upper part) and from gas phase (lower part).

ple preparation was achieved with the **A**utomated **I**on Exchange Apparatus Coupled with **D**etection System for **A**lpha Spectroscopy, **AIDA**.[35,36] In addition, AIDA includes the automated transport of samples to and from the vacuum chamber for nuclear spectroscopy. AIDA, very similar to ARCA in its part as a chemical separation apparatus — with improvements in some details —, has recently been applied to detailed studies of the Rf chemistry.

Perspectives of continuously working liquid-phase separation systems are discussed in the last section of this contribution.

2.2.3. Gas Phase Chemistry Setups.   Continuously operating gas-phase separations,[37] like the **On-Line Gas chromatographic Apparatus**, **OLGA**,[38] were extremely instrumental to study the halide- and oxide-compound formation of the transactinides Rf through Bh and to investigate their characteristic retention time — a measure very often expressed as a "volatility". The lower part of Figure 3 shows a flow scheme of such type of chromatographic separation. Common to all of these experiments is the use of the known nuclide half-life to determine a "retention-time-equivalent" in gas-chromatographic experiments on an atom-at-a-time scale — its the 50% yield value of a break-through curve measured as a function of the isothermal temperature. The temperature corresponding to the 50% yield at the exit of the chromatography column is equal to the temperature at which, in classical gas-chromatographic separations, the retention time would be equal to the half-life of the investigated nuclide. Products leaving the chromatography column are usually attached to new aerosols in a so-called recluster process and are transported in a gas-jet to a detector system like the **RO**tating wheel **M**ultidetector **A**nalyzer, **ROMA**.[39] Here samples are assayed for time correlated, characteristic α decays and for spontaneous-fission fragments.

More details are discussed in the context of the bohrium chemistry. The very unique experimental technique used for the first chemical identification and investigation of hassium is outlined in the next section.

3. Experimental Results and Discussion

Experimental results presented in this section yield important information on the chemical behavior of these elusive elements — for a chemist a value in itself. Discussing these properties in the context of other elements properties, the structure of the Periodic Table or even the manifestation of relativistic effects is an increasingly difficult task. Here one should keep in mind that the measured quantities — deposition temperatures or retention times in thermo- or gas-chromatography and elution positions or distribution coefficients in liquid chromatography or extraction experiments — can straight forward only be compared with
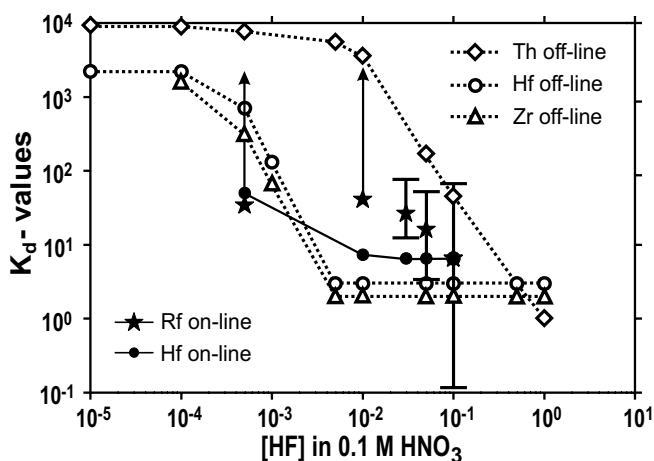
the behavior of other elements investigated in the same experiments, and only if all investigated nuclides have about the same half-life. In the interpretation of these results, e.g., beyond the pure analogy to the lighter homologs, assumptions about the oxidation state or the type of compound formed enter. Empirical model assumptions are needed, e.g., to calculate physicochemical quantities like adsorption — or sublimation enthalpies. The step towards the interpretation of these results in terms of relativistic effects establishes an even more sophisticated task complicated by other effects in the atoms and molecules like spin-orbit coupling, configuration mixing, and shell effects.[10,11]

In this section, exemplary results of the chemical behavior of Rf, Db, and Sg in aqueous solution are presented and discussed. First experiments successfully performed for Bh and Hs in the gas phase are briefly outlined. A more comprehensive compilation of recent results from chemical separations in the gas-phase can be found in References 19, 20.

3.1. Element 104, Rutherfordium.   Already the pioneering experiments of the Rf behavior in acidic, aqueous solutions[14,15] demonstrated that Rf behaves different from trivalent actinides and as expected for a member of group 4 of the PTE with Zr and Hf as the lighter homologs. Many attempts have been made to study the behavior of Rf in comparison with group-4 elements and with tetravalent Th and Pu ions as pseudohomologs, see, e.g., References 18, 19, 21, 33, 40 for summaries. However, some of these experiments yielded conflicting results and some were plagued with adsorption problems, see discussion in Reference 19.

The key issue of most of these experiments concentrated on the competing strength of hydrolysis and complex formation. To shed more light on this question additional data were determined with ARCA to study the behavior of Rf in comparison with lighter homologs and pseudo-homologs. Differences in the F^--
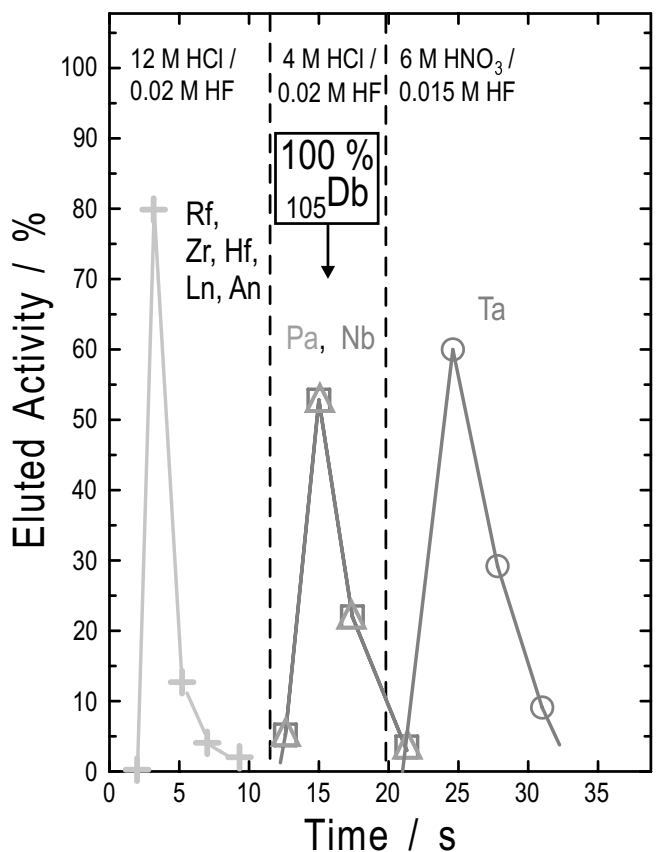

Figure 5.   Elution curve for pentavalent Nb- and Pa-ion tracers with 4 M HCl/0.02 M HF from TIOA/Voltalef columns with ARCA.[47] Db was eluted together with Nb and Pa and did not appear in the Ta strip fraction (6 M HNO₃/0.015 M HF). The first fraction represents loading and washing steps with 12 M HCl/0.02 M HF to separate unwanted trivalent (Ln, An) and tetravalent (Zr, Hf, Rf) species.

Figure 4.   Sorption of Zr, Hf, Th, and Rf on the cation exchange resin Aminex A6 (CIX) from 0.1 M HNO₃ at various HF concentrations. As indicated, some data were obtained in off-line and some in on-line experiments. Revised version of data from Reference 41.

complexation of the group-4 elements Zr, Hf, and Rf, and the pseudo-homolog Th were obtained in mixed 0.1 M HNO$_3$/HF solutions on cation exchange resin (CIX; see Figure 4) and on anion exchange resins (AIX). Data shown in Figure 4 are re-evaluated data from Reference 41. Lower limit values and large error bars for some of the Rf distribution coefficients, K_d, mainly result from low statistics of observed Rf α-decays in one of the two fractions assayed.

K_d values drop for Zr and Hf on CIX between 10^{-4} M and 10^{-2} M HF. For Rf this decrease is observed at about one order of magnitude higher HF concentrations, and even higher for Th, i.e., the transition from cationic to neutral and anionic species requires higher HF concentrations for Rf compared with Zr and Hf but lower than the ones needed for Th. This established the following sequence of F$^-$-complex formation strength at low HF concentrations: Zr ≥ Hf > Rf > Th.

This result is in agreement with data obtained with AIDA[35] and with theoretical expectations.[12] A complete and quantitative understanding of the Rf behavior requires further quantumchemical calculations and a theoretical treatment of complex formation and hydrolysis of these tetravalent ions. However, using the ionic radii of Zr (0.072 nm), Hf (0.071 nm), Rf (0.078 nm), and Th (0.094 nm)[11,42] it is suggestive to apply the HSAB (Hard Soft Acid Base) concept[43] for an explanation of the differences observed on the CIX. In the HSAB concept one assumes that the hard F$^-$-anion interacts stronger with small (hard) cations. From this, one would expect, in agreement with the observation, a weaker F$^-$-complexation of Rf as compared with Zr and Hf.

On AIX, a rise in K_d values from about 10 to more than 100 is observed between 10^{-3} M and 10^{-1} M HF for Zr and Hf measured off-line in batch extraction experiments. This is consistent with the continuation of the trend observed on CIX. For Th off-line data, and for the Hf and Rf on-line data, no significant rise of the K_d values was observed on AIX between 10^{-3} M HF and concentrations of up to 1 M HF. While this is expected for Th, which does not form fluoride complexes, it comes as surprise for Hf and Rf. How much this is affected by the 0.1 M HNO$_3$
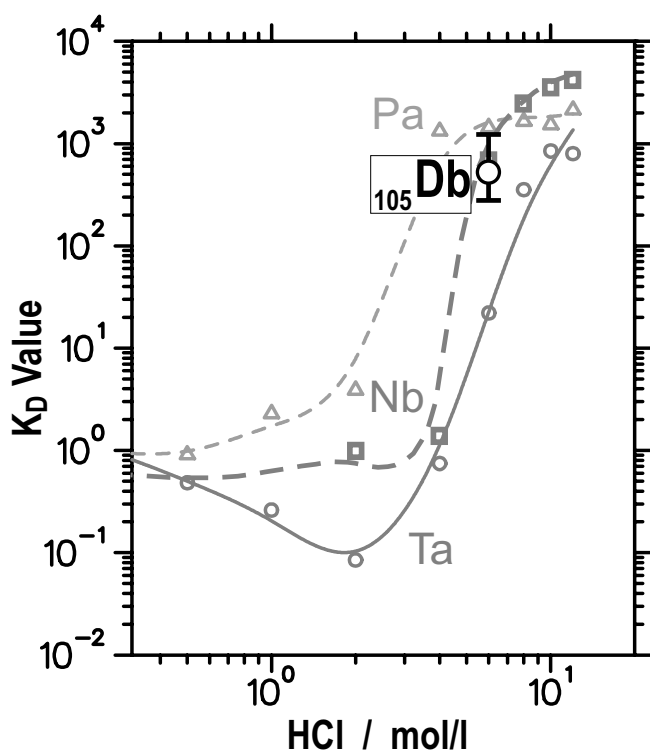
in solution remains unclear. Presently, these data reflect the best knowledge from these experiments. However, it is obvious that there are still unresolved problems with the consistency between off-line and on-line data. To solve this puzzle, kinetic effects are presently considered. Therefore, one shall view the AIX data as under discussion.

Earlier experimental results suggest that Rf forms anionic F$^-$-complexes in pure 0.2 M HF[44] and in mixed 0.27 M HF/0.1 M and 0.2 M HNO$_3$ solutions.[45] It is interesting to note that Rf shows an intermediate behavior between Zr and Hf, with some tendency to be closer to Hf, in the formation of neutral chloride complexes probed with tributylphosphate at 8 M HCl.[46] The formation of anionic chloride complexes of Zr, Hf, and Rf above 8 M HCl was shown in experiments with AIDA.[36] This clearly indicates the group-4 properties of Rf. A hypothetically Th-like or Pu-like behavior of Rf was probed with an anion exchange resin and 8 M HNO$_3$ in experiments with AIDA. While Th and Pu form anionic complexes and are adsorbed Rf remains in solution[36] as expected for a typical group-4 element with non Th-like — and non Pu-like properties.

3.2. Element 105, Dubnium. In its first aqueous chemistry element 105, dubnium, Db, (called hahnium, Ha, in many of the early publications) was adsorbed onto glass surfaces from HCl and HNO$_3$ solutions,[17] a behavior very characteristic for group-5 elements. An attempt to extract Db fluoride complexes failed under conditions in which Ta extracts but Nb does not.[17]

The first detailed comparison between Db, its lighter homologs Nb and Ta, and the pseudo-homolog Pa was carried out from solutions at different HCl concentrations with small amounts of HF added. They were performed as liquid-liquid extraction chromatography experiments with triisooctyl amine, TIOA, as a stationary phase on an inert support in ARCA. In these experiments Db showed a striking non Ta-like behavior, e.g., see Figure 5. At 0.5 M HCl/0.01 M HF, Db even showed Pa-like properties.[47,48]

The interpretation of these results is hampered by the use of the mixed HCl/HF solution that did not allow clearly distinguishing which complex was formed. In contrast to the experimentally observed extraction sequence from HCl solutions with small amounts of HF added, the inverse order Pa ≫ Nb ≥ Db > Ta was theoretically predicted[49] for the extraction from pure HCl solutions. This work considered the competition between hydrolysis[50] and chloride complex formation. Recent experimental studies performed in the pure F$^-$, Cl$^-$, and Br$^-$ system[51] are in excellent agreement with the theoretical predictions which include relativistic effects.[49,52]

For the system Aliquat 336(Cl$^-$) and (pure) 6 M HCl an extraction sequence of Pa > Nb ≥ Db > Ta was determined, see
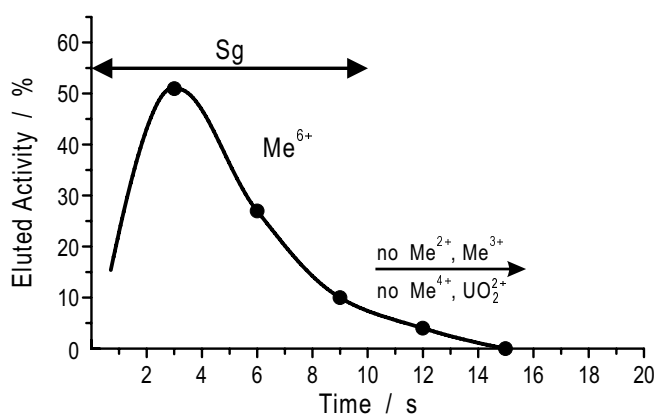

Figure 6. Coefficients for the extraction of Ta (small circles, solid line), Nb (squares, long-dashed line), and Pa (triangles, short-dashed line) tracers from pure HCl solutions at various concentrations into Aliquat 336(Cl$^-$). The K_d of Db at 6 M HCl (large circle) is plotted with error bars encompassing 68% confidence limits. Data from Reference 51.

Figure 7. Elution curve for W-tracer (Me^{6+}), modeling the Sg separation on ARCA in 0.1 M HNO$_3$/5 × 10^{-4} M HF, together with "lower limit" (lower right arrow) for the elution of di-, tri-, tetravalent ions, and UO$_2^{2+}$. Elutions were performed at room temperature with a flow rate of 1 mL/min from 1.6 × 8 mm columns filled with the 17.5 ± 2 μm particle size cation exchange resin Aminex A6. From Reference 54.

Figure 6, which — in agreement of the theoretical prediction[49,52] — is inverse to that in HF containing HCl solution. K_d values of 1440, 683, 438^{+532}_{-166}, and 22 were measured for the Pa, Nb, Db, and Ta, respectively, in series of off-line and on-line experiments. More information on other, previously performed experiments is given in References 18, 19, 21, 33.

3.3. Element 106, Seaborgium. The first chemical separations of element 106, seaborgium, Sg, in an aqueous solution were performed from 0.1 M HNO_3/5 ×10^{-4} M HF on a cation exchange resin in ARCA.[53,54] Seaborgium, detected through three correlated α-decays of the Rf- and No-daughter nuclei, elutes together with the hexavalent Mo- and W-ions; see Figure 7 for a chromatogram of the W-tracer studies. Hexavalent U-ions, present as UO_2^{2+} cations, are strongly retained on the column as di-, tri-, and tetravalent ions are. This result showed that the most stable oxidation state of Sg in aqueous solution is +6, and that, like its homologs Mo and W, Sg forms neutral or anionic oxo- or oxohalide-compounds. Under the given condition, Sg exhibits properties very characteristic of group 6 elements, and does not show U-like properties.

A second experiment was performed with pure 0.1 M HNO_3 to probe the influence of the F^--anions on the complex formation.[55] Contrary to the lighter homologous elements Mo and W, seaborgium was not eluted from the CIX in pure 0.1 M HNO_3. From this it can be concluded that F^--anions significantly contributed to the complex formation in the first experiment. This rules out that Sg was eluted as $[SgO_4]^{2-}$ in the first experiment while the formation of the neutral complex SgO_2F_2 or the anionic complex $[SgO_2F_3]^-$ seems to be likely. The non-tungsten like behavior of Sg in pure HNO_3 may be attributed to its weaker tendency to hydrolyse.[55,56] While Mo and W can reach the neutral species $MO_2(OH)_2$ (M = Mo, W) for Sg hydrolysis presumably stops at $[Sg(OH)_5(H_2O)]^+$ (sometimes characterized as $[SgO(OH)_3]^+$)[56] or already even at $[Sg(OH)_4(H_2O)_2]^{2+}$.

Results and discussions from studies of halide-oxide compounds and oxide-hydroxide compounds of Sg in gas chromatographic experiments can be found in References 57–59.

3.4. Element 107, Bohrium. So far, the only information on the chemical properties of bohrium was obtained in gas chromatographic experiments[26] with an experimental setup schematically shown in Figure 8. The investigation of volatile Me(VII) halides-oxides was an obvious way to study Bh in comparison with the lighter group-7 homologous elements Tc and Re according to their intermediate position between the lighter transactinides, which form volatile halides, and the highly volatile group-8 tetroxides.

Nuclear reaction products were carried with C-aerosols as a cluster material from the recoil chamber to the **On-Line Gas** chromatographic **Apparatus, OLGA,**[38] see Figure 8. The reactive gases HCl and O_2 were added in front of the high temperature zone of the reaction oven where C-aerosols were burned and where the compounds MeO_3Cl (Me = Tc, Re, Bh) were formed. Relative yields of the breaking-through compounds were measured as a function of the isothermal temperature in the quartz chromatography column in the detection syste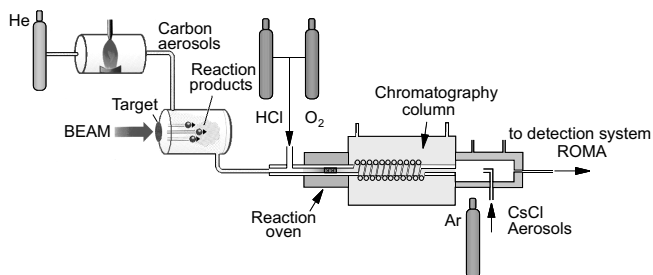m **RO**tating wheel **M**ultidetector **A**nalyzer, **ROMA.**[39] CsCl was used as a re-cluster material in the Bh experiments to transport those compounds, which leave the chromatography column, to the ROMA.

From the observation of four ^{267}Bh nuclear-decay chains at 180 °C (isothermal chromatography temperature), two at 150 °C, and zero at 75 °C a BhO_3Cl break-through curve was modeled and was compared with known curves from tracer studies with lighter homologous elements.[26] The characteristic 50% yield of the BhO_3Cl curve is located at a higher temperature compared with the ReO_3Cl and the TcO_3Cl behavior.

Qualitatively, this results shows that Bh behaves like a member of group 7 of the PTE and forms a volatile halide-oxide — presumably BhO_3Cl as inferred from its properties — which is less volatile than the compound of the lighter homologous elements.[26] More quantitatively, the deduced BhO_3Cl adsorption enthalpy is in good agreement with a theoretical prediction including relativistic effects.[60] This result coincides with the value expected from empirical correlations of thermochemical properties and Bh positioned in group 7 of the periodic table.[26]

3.5. Element 108, Hassium. Very recently, the first chemical separation and identification of a Hs compound was achieved.[27,61] From its expected chemical properties as a member of group 8 of the Periodic Table it is most attractive to investigate the highly volatile tetroxide — a unique property only known for group 8 transition metals. The experiment was performed at the UNILAC accelerator at GSI with a setup schematically shown in Figure 9. This experiment was unique, in a number of aspects, and different from recent gas-chromatographic experiments:

(*i*) A rotating target wheel, ARTESIA, see Figure 9, for ^{248}Cm targets in combination with a gas-jet transport system was applied for the first time in transactinide chemistry to accept higher beam intensities.

(*ii*) The chemical reaction with the reactive gas O_2 was performed "in-situ" in the recoil chamber IVO, **I**n situ **V**olatilization and **O**n-line **D**etection.[62] An oven attached to the recoil chamber provided a fast and efficient oxidation of the stopped recoils. This allowed transporting highly volatile compounds without any cluster material in a dry He/O_2 gas mixture over 10 m in a Teflon capillary to the detection system. It should be noted that already early work on the halide volatility of Rf used a chemical reaction in or immediately behind the recoil chamber directly attached to a thermo-chromatography column, see Reference 63 and references therein.

(*iii*) An on-line thermo-chromatography separator and detector setup, COLD, **C**ryo **O**n-line **D**etector,[61] operated at low temperatures was mainly used. This is an improved version of the previously developed Cryo-Thermochromatography Separator, CTS,[64] which was also used during this beam time. COLD consists of 36 pairs of silicon PIN-photodiodes coupled to a support which provides the negative temperature gradient between about room temperature at the entrance side and about 100 K at the gas exit.

Nuclear decays of the known isotope ^{269}Hs and of the isotope ^{270}Hs, which was discovered in the course of this experiment, were observed[27] in a narrow peak[61] along the temperature gradient. From the observation of HsO_4 and its adsorption position, in comparison with the known one of OsO_4, one concludes that

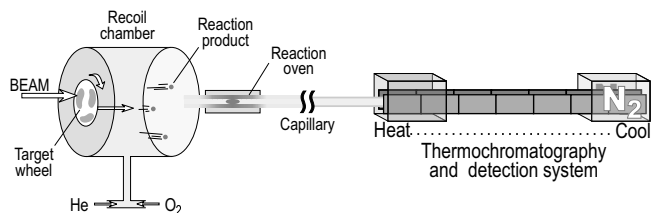**Figure 8.** Schematic view of the Bh gas-chromatography experiment. Adapted from Reference 26.

Figure 9. Schematic view of the low temperature thermo-chromatography experiment to investigate HsO_4. Adapted from Reference 61.

Hs forms a relatively stable, volatile tetroxide — as expected for a typical member of group 8.[12,65] However, the exact adsorption position is at a surprisingly high temperature,[61] i.e., HsO_4 exhibits an unexpected low volatility or, in other terms, a high, negative adsorption enthalpy.

4. Summary

As has already been observed in earlier experiments, the results from these more recent studies also justify positioning of the transactinides — or superheavy elements —, beginning with element 104, into the seventh period of the Periodic Table, see Figure 1. So far chemical studies on their behavior have been performed with Rf, Db, and Sg in aqueous phase. Studies in the gas phase — also performed for Rf, Db, and Sg — have recently been extended to Bh and Hs. All experimental results yield properties which place these elements into their respective group of the Periodic Table — Rf, Db, Sg, Bh, Hs into group 4, 5, 6, 7, and 8, respectively. This demonstrates that the Periodic Table still remains an appropriate ordering scheme.

However, a closer and more subtle look reveals that all the more detailed chemical properties of these elements — in comparison with their lighter homologs — are no longer reliably predictable by simple extrapolations in the Periodic Table. But modern relativistic molecular calculations in combination with empirical models allow for quantitative, or semi-quantitative, comparisons of experimental and theoretical results, and they show excellent agreement in a number of cases. From this one can deduce that relativistic effects strongly influence the chemical properties of the transactinides and they can give indications what facets of the chemical properties are predominately due to relativistic effects.

5. Perspectives

The prospects to study chemical properties of transactinides beyond element 106 in aqueous phase experiments mainly depend on the development of methods to cope with production rates of less than one atom per hour. This situation will remain even after further developments to increase production rates from higher heavy-ion beam intensities and developments of new target design which can tolerate these intensities. But for all of that, only continuously running separation processes with an on-line detection or with detection of long-lived daughter isotopes are applicable as long as the half-lives for these superheavy element nuclides will be in the range of minutes or shorter.

Fascinating and challenging prospects are ahead with half-lives of the order of about 10 s for isotopes of element 107, Bohrium, Bh, — for which a gas-chromatographic separation and characterization was already achieved[26] — and element 108, Hassium, Hs, located at or slightly below the $N = 162$ neutron-shell. Even more interesting and more challenging will be future attempts to extend the aqueous chemistry of superheavy elements all the way to element 114. The most recent results from the reaction of ^{48}Ca ions with ^{238}U and $^{242,244}Pu$ targets[2,3] indicate the existence of even longer-lived nuclides of elements 108 to 114 between neutron number $N = 169$ and $N = 174$.

In addition to the fascinating chemistry aspects of these superheavy elements, nuclear chemistry experiments are vital tools for a clear identification of the atomic number (element identification) of relatively long-lived spontaneously-fissioning nuclides at the end of α-decay chains.

Two types of continuously operating liquid-chemical separation processes are presently emerging for transactinides. The one with a truly continuous separation and detection technique is the fast, automated, and miniaturized centrifuge system **SISAK III**[66–68] which allows separating nuclides with half-lives of the order of 1 s. Separations are based on liquid-liquid extractions with subsequent phase separation and detection of the α decay in the organic phase with an extractive liquid scintillator.[69] Most recent improvements of the detection, data acquisition, and pulse shape discrimination system were tested in Rf-chemistry experiments and yielded very promising aspects for the future application of the SISAK system in the transactinide chemistry with short-lived nuclides.

A different approach to continuously separate transactinide elements and to establish their chemical behavior from the observation of long-lived daughter isotopes in one or another fraction was introduced with the three- or multi-column technique.[44,45,70] First experiments have been performed with Rf to study the F^--complexation[44,45] and with Db for nuclear studies in the RACHEL device.[30] Rf and Db, transported with the He(KCl)-jet to the chemistry apparatus, were continuously dissolved, and this solution was passed through three consecutive ion-exchange columns. Primary produced di- and trivalent actinides were "filtered" out on the first cation-exchange column. In the next anion-exchange column anionic species were retained for some time, while the following cation-exchange column adsorbed cationic species — the long-lived α-decay products of Rf. These were eluted after the end of irradiation and were detected by off-line spectroscopy. So far, the Rf experiments showed only a group-4 behavior of Rf and were not able to determine subtle differences within the chemical properties of group-4 elements.

One of the disadvantages of this technique is its limited range of half-lives and K_d values. Its big advantage over batch-wise techniques like ARCA is its potential to study short-lived isotopes of, e.g., a few seconds half-life. In addition, because of its continuous and not manpower consuming operation, it may allow extending these studies to nuclides with cross sections well below the nanobarn level. Preparations are under way to perform such studies to determine differences in the hydrolysis and complex formation of Mo, W, and Sg.

Existing separation techniques, like ARCA II,[34] will remain essential tools to shed more light on the diverse and often unexpected behavior of the lightest transactinide elements. With the relatively long half-lives and rather high cross sections one has a chance to reveal properties in more complex chemical systems which may not be applicable for heavier elements.

Well developed gas-phase chemistry techniques are at hand to deepen our insights into many unresolved questions of compound formation, volatility, and adsorption behavior of group 4 through 7 halides, oxides, and mixed compounds. A challenge for the future will be the gas-phase studies of the metallic transactinides beyond group 8. Element 112 has a very unique position in this row with Hg as its lighter homolog in group 12. Here first experiments are under way[71] to look into the fascinating question whether element 112 will exhibit a Hg-like behavior or, possibly, a much more inert Rn-like one.

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