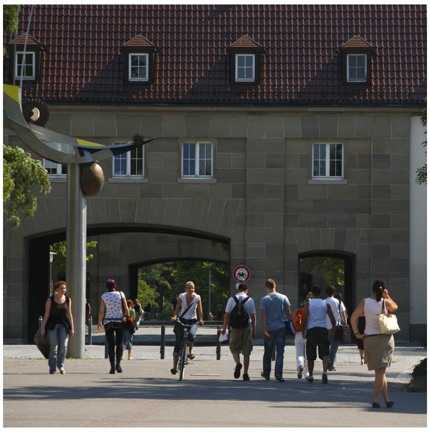
SEVENTH WORKSHOP ON THE CHEMISTRY OF THE HEAVIEST ELEMENTS

JOHANNES GUTENBERG UNIVERSITÄT MAINZ

October 11 – 13, 2009

Workshop Program



SEVENTH WORKSHOP ON THE CHEMISTRY OF THE HEAVIEST ELEMENTS

Program

Monday, October 12, 2009

9:00 Welcome

Theoretical Chemistry **Chair: N. Trautmann**

09:10 V. Pershina
Theoretical Predictions of the experimental behaviour of the heaviest elements
09:35 A. Borschevsky
Fully relativistic ab initio studies of properties of the heaviest elements
10:00 J. Anton
Adsorption behaviour of elements 112, 114 and their homologues studied by fully-relativistic DFT cluster calculations

10:25 Coffee break

Physics of SHE **Chair: A. Türler**

10:45 K. E. Gregorich
Hot fusion in recoil separators
11:10 A. Gorshkov
Measurements of $^{260\text{-}262}$Rf produced in ^{22}Ne + ^{244}Pu fusion reaction at TASCA
11:35 K. Morimoto
Production and decay properties of ^{266}Bh and its daughter nuclei by using the ^{248}Cm(^{23}Na, 5n) reaction
12:00 N. Sato
Production and decay properties of $^{263,\,264,\,265}$Hs

12:25 Lunch break

Chair: K. E. Gregorich

13:30 R. Graeger
Reaction studies about the Q-value influence on the production of superheavy elements
13:55 Ch. E. Düllmann
Decay properties of “chemistry isotopes” of light even-Z transactinides
14:20 Ch. E. Düllmann
Studies of the reaction ^{244}Pu(^{48}Ca, 3-4n) ^{288,289}114 at TASCA

14:45 Coffee break

Gas phase chemistry Chair: J. V. Kratz

15:30 A. Serov
Adsorption interaction of ^{113m}In and ^{212}Pb isotopes with quartz
15:55 R. Eichler
Chemical investigations of element 114
16:20 A. Yakushev
Study of 114 chemistry

Aqueous Chemistry Chair: H. Nitsche

16:45 Y. Nagame
Aqueous chemistry of Rf and Db
17:10 Y. Kasamatsu
Anion-exchange experiment of Db with AIDA-II
17:35 A. Toyoshima
Electrochemical studies of the heaviest actinides
18:00 K. Ooe
Solvent extraction studies of molybdenum and tungsten as homologues of seaborgium (element 106)

18:30 Dinner
19:30 Wine tasting

Tuesday, October 13, 2009

Chair: Y. Nagame

09:00 J. Even
Underpotential deposition – a method to study the chemistry of hassium
09:25 F. Samadani
A SISAK extraction system for chemical studies of hassium
09:50 D. Hild
MicroSISAK – Improvements of a device for continuous liquid-liquid extraction on a microliter scale
10:15 R. A. Hendersson
Automated chemistry efforts at LLNL using Eichrom´s DGA resin

10:40 Coffee break

11:00 M. E. Bennett
Extraction chromatographic studies of Rf and Db homologs

Technical aspects Chair: H. Gäggeler

11:25 M. Schädel
TASCA – A new tool in the quest for superheavy element studies
11:50 A. Yakushev
A new TASCA focal plane detector and data acquisition system
12:15 H. Nitsche
From new actinide target technology to heavy element chemistry

12:40 Lunch break

Chair: M.Schädel

13:40 H. Haba
RIKEN GARIS as a promising interface for superheavy element chemistry – Production of ^{261}Rf using the GARIS/gas-jet system
14:05 R. Dressler
PureCOLD for ß-α-pile-up suppression – a status report
14:30 D. Wittwer
Stopping force estimations for element 114 in mylar and argon gas
14:55 J. Runke
Preparation of ^{244}Pu targets by electrodeposition
15:20 K. Eberhardt
Coupling of TASCA with SHIPTRAP
15:45 J.V. Kratz
The Helmholtz-Institute Mainz: Stucture, Symmetry, and Stability

16:10 End of the Workshop

Theoretical Predictions of the Experimental Behaviour of the Heaviest Elements

V. Pershina [a,*], *J. Anton* [b], *T. Jacob* [b], *A. Borschevsky*[c], *E. Eliav*[c], *U. Kaldor*[c]

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The heaviest elements which are presently of interest for the gas-phase chemical studies are 112, 113, 114 and possibly heavier. Their adsorption is to be studied using the gas-phase thermochromatography technique, whereby the elements are adsorbed on gold covered silicon detectors of the chromatography column [1,2]. The electrochemical deposition of Pb/114 and Os/Hs on some noble metal electrodes from aqueous acidic solutions is also a way to study their reactivity [3]. Aqueous chemistry experiments are concentrating on the studies of the complex formation of Rf through Hs and their lighter homologs in the chemical groups under new experimental conditions [4].

For these sophisticated experiments with single atoms, the assistance of the chemical theory is required. Predictions of the experimental behaviour based on relativistic electronic structure calculations are indispensable for the modern chemical studies. They help design the experiments, interpret the observed behaviour and understand the nature of chemical interactions in a systematic way.

With this aim in view, our groups have recently been concentrating on the calculations of the electronic structures of various systems of Hs, elements 112-118 and their lighter homologs using the best available fully relativistic quantum chemical codes [5-10].

Results of the novel 4*c*-DFT and *ab initio* DC studies on these elements, as well as a critical analysis of the data and systematics in this area will be presented. One of those interesting cases is shown in Fig. 1. There, results of various studies for group 12 elements Hg and element 112 and group 14 elements Pb and element 114 are compared: the calculated binding energies of the gold containing dimers D_e(MAu) [5], of the ad-atom-gold cluster systems D_e(M-Au$_n$) [6] and adsorption enthalpies ΔH_{ads} of these elements on gold (measured [1,2] and predicted in the present studies).

From this picture, one can see that element 114 is stronger reacting with gold than element 112 in all the cases, which is lying on its different electronic structure: the 7p$_{1/2}$ (114)AO fits energetically better to the valence orbitals of gold than the 7s(112) AO, which is more stabilized than the 7p$_{1/2}$(114) AO. One can also see that the point for the measured ΔH_{ads} of element 114 on gold [2] is far down, in the area of very low energies, thus evidencing that this should not come from the interaction with gold.

Some other interesting cases are discussed in the presentation.

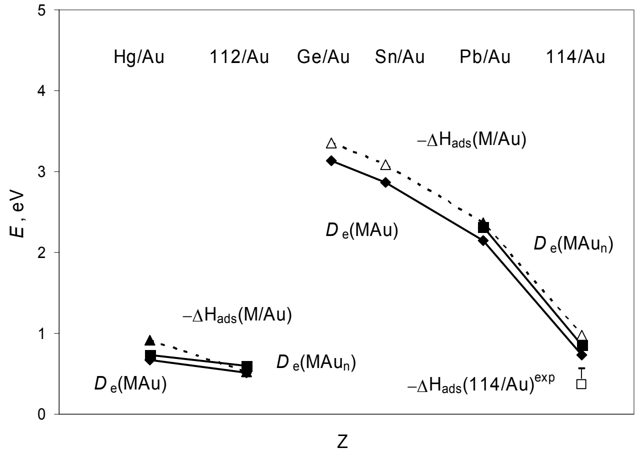

Figure 1: Binding energies of group 12 elements Hg and element 112 and group 14 elements Pb and element 114 in various systems: the calculated binding energies D_e(MAu) of the gold dimers (filled rhomboids); binding energies D_e(M-Au$_n$) of large ad-atom-gold cluster systems (filled squares), and adsorption enthalpies ΔH_{ads} on gold (filled triangles- - measured and open ones – predicted). The measured ΔH_{ads}(114) is shown with an open square [2]

References

[1] R. Eichler *et al.* Nature, Letters, **2007**, *447*, 72.
[2] R. Eichler, *et al.* Abstract of the NRC7 Conference, Budapest, 2008.
[3] H. Hummrich *et al.* Radiochim. Acta **2008**, *96,* 73.
[4] A. Toyoshima *et al.* Radiochim. Acta **2008**, *96,* 125.
[5] V. Pershina, J. Anton and B. Fricke, J. Chem. Phys. **2007**, *127*, 134310.
[6] C. Sarpe-Tudoran, B. Fricke, J. Anton, V. Pershina. J. Chem. Phys. **2007**, *126*, 174702.
[7] J. Anton, T. Jacob, V. Pershina, GSI Annual Report 2008.
[8] V. Pershina, A. Borschevsky, E. Eliav and U. Kaldor, J. Chem. Phys **2008**, *128*, 024707.
[9] V. Pershina, A. Borschevsky, E. Eliav and U. Kaldor, J. Phys. Chem.. **2008**, *112,* 13712.
[10] V. Pershina, A. Borschevsky, E. Eliav and U. Kaldor, J. Chem. Phys **2008**, *129*, 144106.

Fully relativistic *ab initio* studies of properties of the heaviest elements

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Predictions of the interaction of superheavy elements with various surfaces are essential for their identification and separation. Accordingly, our theoretical chemical studies have been aiming at predictions of their adsorption behavior on various surfaces, using the state of the art fully relativistic *ab initio* calculations of their atomic and molecular properties.

Adsorption on inert surfaces

Recently, we have predicted the adsorption properties of the superheavy elements 112, 114, 113 and 118 on inert surfaces [1-3]. Relativistic effects are of crucial importance for heavy and superheavy elements, hence all the calculations were performed using the fully relativistic 4-component Dirac Hamiltonian. The electronic correlation was taken into account by using the state of the art relativistic coupled cluster method, with single, double and perturbative triple excitations (CCSD(T)). Very large basis sets, consisting of 26*s*24*p*18*d*13*f*5*g*2*h* functions, were used [4]. In order to assess the accuracy of the predictions, the same calculations were performed for the lighter homologues of the 7*p* elements, mercury to radon, and the results are compared to the experimental values, where available.

The calculated atomic properties, including polarizabilities and adsorption enthalpies on inert surfaces of the entire 7*p* row will be presented. Using the properties of the lighter homologues for comparison, the influence of relativistic effects on adsorption properties of the superheavy elements will be analyzed.

Adsorption on gold

The adsorption of elements 112 and 114 on gold surfaces is investigated through the fully relativistic *ab initio* calculations of the electronic structure of the 112Au and 114Au dimers. The 2-component relativistic Hamiltonian, obtained by the infinite order 2-component transformation of the Dirac Hamiltonian in the finite basis set [5], was used. This is one of the most economical and accurate approximations to the full Dirac-Coulomb Hamiltonian. Correlation was taken into account using the Fock space coupled cluster method, one of the most powerful tools in quantum chemistry. It also allows us to obtain, in one calculation, the energy of MAu (M=112, 114), MAu^+, and MAu^-. Thus we can look at the potential energy curve of the neutral molecule and both its ionized states in order to obtain their equilibrium bond lengths, dissociation energies, ionization potentials, and vibrational constants. For comparison, the same calculations are performed on their lighter homologues, HgAu and PbAu. All the calculations are performed using the Dirac program package [6].

References

[1] V. Pershina, A. Borschevsky, E. Eliav and U. Kaldor, J. Chem. Phys **128**, 024707 (2008)

[2] V. Pershina, A. Borschevsky, E. Eliav and U. Kaldor, J. Phys. Chem. . **112**, 13712 (2008)

[3] V. Pershina, A. Borschevsky, E. Eliav and U. Kaldor, J. Chem. Phys **129**, 144106 (2008)

[4] K. Faegri, Theor. Chim. Acta 105 (2001) 252

[5] M. Barysz, A.J. Sadlej and J.G. Snijders, Int. J. Quantum Chem. **65,** 225 (1997),

[6] DIRAC04, written by H.J. Ja. Jensen *et al* (2004)

Adsorption Behaviour of Elements 112, 114 and their Homologues Studied by Fully-Relativistic DFT Cluster Calculations

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Investigations of chemical and physical properties of the heaviest elements (those beyond Lr) is a hot topic since several decades. In this time period, many new elements were discovered and after a proper characterization were added to the Periodic Table of the elements, thus being of fundamental interest. The main problem of such investigations, however, is the rather short half-live of these elements, which requires the development of innovative experimental techniques. Similarly, for theoretical studies, standard quantum-mechanical packages that treat a system mostly non- or only scalar-relativistically are not satisfactory: Due to rather significant relativistic effects on the electron shells of the heaviest elements, a fully-relativistic, four-component (4*c*) description is required.

Recently, we have studied the adsorption behaviour of HsO_4 and its homologues, RuO_4 and OsO_4, on a quartz substrate [1]. While adsorption itself was treated via a physisorption model, the molecular properties required by this model were calculated with the use of our fully-relativistic density functional theory (4*c*-DFT) approach [2]. Here, we present direct calculations of the adsorption energies of the heaviest elements on a metal surface now, without a physisorption model, by using a cluster-approach [2]. According to this approach, the metal surface is simulated by metal clusters of various size onto which the adatoms of interest are placed at various positions.

In the present studies, we have calculated binding energies, E_b, for elements 112 and 114 and their lighter homologues Hg and Pb, respectively, on very large gold cluster (up to 120 atoms), simulating both the Au(100) and Au(111) surfaces. We found that with Au(111) our binding energies for Hg, Pb, and element 112 much better fit to the experimental adsorption enthalphies [3,4] than those obtained with the Au(100) surface [5]. Thus, we expect the same to be the case for element 114.

We found that Hg/112 and Pb/114 adsorb preferentially at different positions. Also, the adsorption energies of elements 112 and 114 are related to those of their lighter homologues in a different way. Thus, $E_b(112)$ is only slightly (0.1–0.2 eV) lower than $E_b(\text{Hg})$, while $E_b(114)$ is much lower (1.4 eV) than $E_b(\text{Pb})$. This is due to the fact, that in element 112 both the relativistically stabilized 7*s* and destabilized 6d AOs take part in the binding, while for element 114 binding is mostly determined by the relativistically stabilized $7p_{1/2}$ orbital. In contrast to Pb, where $6p_{3/2}$ strongly contributes to the surface bond, the $7p_{3/2}$ does not participate due to its large relativistic destabilization (the spin–orbit splitting between $7p_{1/2}$ and $7p_{3/2}$ is about 3.5 eV). Furthermore, binding of element 114 to gold is similar to that of element 112, however, about 0.2 eV stronger.

Overall, our predicted sequence in the E_b values is $E112 < Hg < E114 \ll Pb$. Thus, we expect that in the thermochromatography experiments, element 114 will adsorb right at the beginning of the chromatography colum with the hot end of 35 °C, at the position of Hg.

Detailed results - binding energies, bond lengths, and other properties of the calculated systems - will be given in the presentation.

References

[1] V. Pershina, J. Anton, T. Jacob, *Phys. Rev. A* **2008**, *78*, 032518.

[2] J. Anton, B. Fricke, E. Engel, *Phys. Rev. A* **2004**, *69*, 012505.

[3] J. P. Niklaus, *et al.* PSI Annual Report **2000**, p. 8.

[4] R. Eichler *et al.*, *Nature, Letters*, **2007**, *447*, 72.

[5] C. Sarpe-Tudoran, B. Fricke, J. Anton, V. Pershina, *J. Chem. Phys.* **2007**, *126*, 174702.

Hot Fusion in Recoil Separators

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Relatively long-lived transactinide elements (i.e., elements with atomic number $Z \geq 104$) up to $Z = 108$ have been produced in nuclear reactions between low Z projectiles (C to Al) and actinide targets. Cross sections have been observed to decrease steeply with increasing Z and reach the level of a few picobarns for element 108 [1]. Production cross sections of several picobarns have been reported [2-4] for comparatively neutron-rich nuclides of 112 through 118 produced via hot fusion reactions with ^{48}Ca and actinide targets. There are now reports of two independent confirmations [3, 5] of SHE production in such reactions. Some of those heavy nuclides are reported to have lifetimes on the order of seconds or longer. The relatively high cross sections in these hot fusion reactions are not fully understood and this has renewed interest in systematic studies of heavy-ion reactions with actinide targets.

Oganessian has indicated that the large cross sections for SHE production in ^{48}Ca reactions with actinide targets result from enhanced survivability of the compound nucleus during de-excitation by neutron evaporation in competition with fission [2]. Spherical shell effects expected in the region approaching Z=114 and N=184 should lead to larger fission barriers, which could be expected to result in enhanced survival of the heavy element products. However, compared to the trend of exponentially decreasing cross sections with increasing Z (see fig. 11 of [2]), up to six orders of magnitude of cross section enhancement during evaporation of four neutrons is required to explain the picobarn-level SHE cross sections. It should be noted that a similar enhancement of the compound nucleus evaporation residue (EVR) cross sections was not observed [6] in the vicinity of the strong spherical N=126 shell.

A systematic study of heavy element formation by compound nucleus – evaporation reactions using ^{238}U targets with neutron-rich projectiles from ^{22}Ne through ^{37}Cl to provide data for understanding and modeling these reactions was undertaken at Lawrence Berkeley National Laboratory. Data is shown in Figure 1. Together with data from similar reactions with ^{18}O [7], ^{19}F [7], ^{34}S [8], ^{40}Ar [9], and ^{48}Ca [3, 10-14], cross section trends were analyzed to arrive at conclusions about critical angular momentum for fusion (l_{crit}), survival during deexcitation by neutron evaporation in competition with fission and other de-excitation modes (Γ_n/Γ_{tot}), comparisons with the standard fusion-evaporation model HIVAP [15], and extrapolation to SHE cross sections.

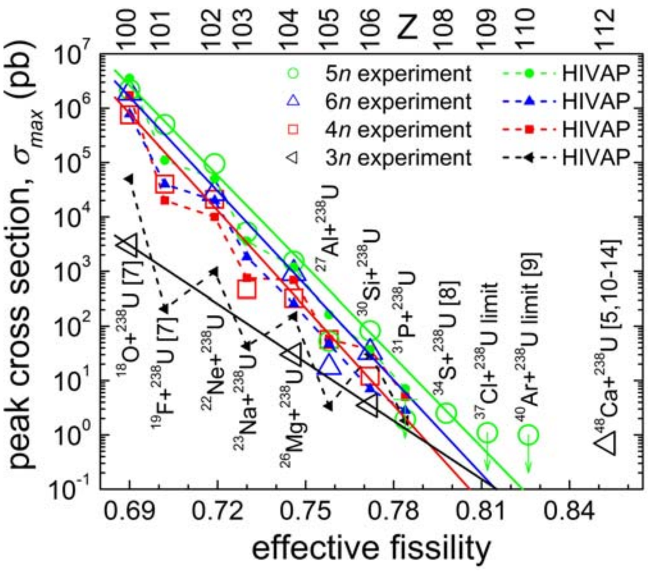

Figure 1: σ_{max} (open symbols) as a function of effective fissility [16]. Heavy straight lines (top to bottom) are exponential fits to even-Z σ_{max} for the 5*n*, 6*n*, 4*n*, and 3*n* exit channels. Filled symbols connected by dahsed lines are results of HIVAP calculations [15].References

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[1] J. Dvorak, *et al.*, Phys. Rev. Lett. **97**, 242501 (2006).
[2] Y. T. Oganessian, J. Phys. G **34**, R165 (2007).
[3] S. Hofmann, *et al.*, Eur. Phys. J. A **32**, 251 (2007).
[4] R. Eichler, *et al.*, Nature **447**, 72 (2007).
[5] R. Eichler, *et al.*, Nucl. Phys. **A787**, 373c (2007).
[6] D. Vermeulen, H. G. Clerc, C. C. Sahm, K. H. Schmidt, J. G. Keller, G. Münzenberg, and W. Reisdorf, Z. Phys. A **318**, 157 (1984).
[7] E. D. Donets, V. A. Shchegolev, and V. A. Ermakov, Sov. J. Nucl. Phys. **2**, 723 (1966).
[8] Y. A. Lazarev, *et al.*, Phys. Rev. Lett. **75**, 1903 (1995).
[9] S. Hofmann (private communication).
[10] Y. T. Oganessian, *et al.*, Eur. Phys. J. A **5**, 63 (1999).
[11] Y. T. Oganessian, *et al.*, The European Physical Journal A - Hadrons and Nuclei **V19**, 3 (2004).
[12] W. Loveland, K. E. Gregorich, J. B. Patin, D. Peterson, C. Rouki, P. M. Zielinski, and K. Aleklett, Phys. Rev. C **66**, 044617 (2002).
[13] K. E. Gregorich, *et al.*, Phys. Rev. C **72**, 014605 (2005).
[14] Y. T. Oganessian, *et al.*, Phys. Rev. C **70**, 064609 (2004).
[15] W. Reisdorf and M. Schädel, Z. Phys. A **343**, 47 (1992).
[16] P. Armbruster, Acta Phys. Pol. B **34**, 1825 (2003).

Measurements of $^{260\text{-}262}$Rf produced in ^{22}Ne + ^{244}Pu fusion reaction at TASCA[*]

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Production and decay of ^{260}Rf, ^{261a,b}Rf and ^{262}Rf [1,2,3], produced in the asymmetric nuclear fusion reaction ^{244}Pu(^{22}Ne,xn) was studied in the final experiment of the commissioning phase of TASCA. TASCA was operated in the High Transmission Mode (HTM) [4]. The ^{22}Ne ion beam (average intensity: 0.8 μA_{part}) impinged on a rotating target wheel with 0.4 mg/cm^2 ^{244}PuO2 targets on 2.2 µm Ti backings. Three beam energies in the center of the target, $E_{c.o.t.}$, of 109 MeV, 116 MeV and 125 MeV, were used for the production of ^{262}Rf, ^{261}Rf and ^{260}Rf, respectively. The transmission of Rf has been optimized in He filling gas. The optimal pressure was 0.4 mbar. The magnetic rigidity, Bρ, was determined to be 1.99 T·m. To increase suppression of unwanted products, a He/H_2 (2:1) filling gas at a pressure of 1.5 mbar was used in experiments with the FPDs. Separated reaction products were implanted into a Focal Plane Detector (FPD). Two types of the FPD were used: a (80 x 36) mm^2 16-strip Position-Sensitive silicon Detector (PSD) or a (58 x 58) mm^2 Double-Sided Silicon Strip Detector (DSSSD). In other experiments, evaporation residues were guided to a Recoil Transfer Chamber (RTC), where they passed a 1.2 µm thick (140 x 40) mm^2 Mylar window and were thermalized in 1.2 bar He in the RTC (depth: 17 mm) and transported to the Rotating wheel On-line Multidetector Analyzer (ROMA) by an He/KCl jet (gas flow rate: 3.45 L/min) through a 4 m long polyethylene capillary (inner diameter: 2 mm).

The measurement of ^{260}Rf, produced in the 6n evaporation channel at $E_{c.o.t.}$= 125 MeV yielded 15 time ($\Delta t \leq 200$ ms) correlated EVR-SF events in the PSD. The correlation time analysis yielded a half-life of $21^{+7.3}_{-4.3}$ ms (errors are within the 68% confidence interval). A search for ^{262}Rf decays at $E_{c.o.t.}$ = 109 MeV 10 position and time correlated EVR-SF events observed in the DSSSD, with EVR energies of 0.8 to 3.5 MeV and SF fragment energies of > 80 MeV. The measured $T_{1/2}$ for ^{262}Rf is 190^{+85}_{-45} ms (Fig. 1a), in contradiction with values from [1,2]. In addition, 15 short EVR-SF correlations were registered with $\Delta t \leq 4$ ms and EVR energies of 7.5 ± 5.0 MeV. They were attributed to the decay of ^{244mf}Am ($T_{1/2}$ = 0.9 ms) and ^{246mf}Am ($T_{1/2}$ = 76 µs). Because of a relatively high counting rate of EVR-like events in the DSSSD a random event analysis was performed for EVR-SF correlations within a Δt of 1 s. The random event number, n_b, was calculated individually for each detector pixel because of very non-uniformly EVR distribution in the FPD.
^{261}Rf was produced in the 5n channel at $E_{c.o.t}$ = 116 MeV and was detected in ROMA. Stepping time of 35 s (for ^{261a}Rf) were used. 149 single α-particles (E_α = 7.8 – 8.5 MeV) from ^{261a}Rf and ^{257}No were registered; among these 28 α-α correlations. Also, 11 SF-events were registered and are attributed to ^{261b}Rf based on the measured $T_{1/2}$ of $2.2^{+0.9}_{-0.5}$ s (Fig. 1b). The SF activity assigned in [2] to ^{262}Rf likely originated from then unknown ^{261b}Rf.

From our results and cross section of 4.4 nb [5], a transmission of Rf through TASCA to a 140 x 40 mm^2 large area in the focal plane of 10% follows. For ^{261b}Rf, a cross section of $1.8^{+0.8}_{-0.4}$ nb was calculated, respecting decay during transport. With an estimated transmission of 6% to the area of the FPDs, preliminary cross sections for ^{260}Rf and ^{262}Rf are ≈ 1.2 nb and ≈ 450 pb, respectively. ^{261b}Rf was observed for the first time as an EVR. The production ratio of ^{261a}Rf to ^{261b}Rf is about 2.5:1.

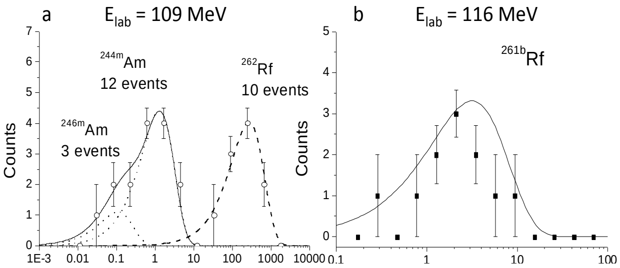

Figure 1: Time distributions of a) EVR-SF correlations from the DSSSD and b) SF decays from ROMA.

References

[1] L. P. Somerville *et al.*, Phys. Rev. C **31**, 1801 (1985). [2] M. R. Lane *et al.*, Phys. Rev. C **53**, 2893 (1996).
[3] Ch. E. Düllmann, A. Türler., Phys. Rev. C **77**, 064320 (2008).
[4] A. Semchenkov *et al.*, NIMB **266**, 4153 (2008).
[5] Yu. Lazarev *et al.*, Phys. Rev. C **62**, 064307 (2000).

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[*] Work supported by BMBF project 06MT248.
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Production and Decay Properties of ^{266}Bh and its daughter nuclei by using the ^{248}Cm(^{23}Na,5n) Reaction

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A nuclide, ^{266}Bh, is the great-grand-daughter of ^{278}113 that is produced in the ^{209}Bi + ^{70}Zn reaction [1, 2]. The identification was based on a genetic link to the known daughter nucleus ^{262}Db by alpha-decays. The identification of the heaviest nuclides is very difficult because of their extremely small production cross sections. Only five atoms have been assigned to ^{266}Bh by direct production [3, 4]. The main purpose of this work is to provide further confirmation of the production and identification of the isotope ^{278}113.

The experiment was performed at the RIKEN Linear Accelerator (RILAC) Facility. A ^{248}Cm$_2$O$_3$ target was prepared by electro-deposition onto a titanium backing foil. Six pieces of the target were mounted on a rotating wheel of a diameter of 10 cm. A ^{23}Na beam was extracted from RILAC. Beam energies of 126, 130, and 132 MeV were used. The ^{23}Na beam was pulsed with micro and macro pulse structures. The micro-pulse structure was used to prevent the irradiation of the target frame and the two vacant target positions of the wheel. In the macro-pulse structure, the beam was ON for 3 s and then OFF for 3 s independently of the micro-pulse structure. The typical beam intensity was 1 particle micro A on average, and the 1.9 x 10^{19} of beam dose was accumulated in about two months machine time. A gas filled recoil ion separator GARIS was used to collect evaporation residues and separate them from the beam particles. The evaporation residues ware transported to the focal plane of the GARIS. Silicon detector box was installed in the focal plane of GARIS. The total counting rate of the focal plane detector was approximately 3 x 10^4 /s in the beam ON period, while that was 5-10 /s in the beam OFF period.

We have observed 32 correlations in total. 14 correlation events were clearly assigned to the decay chain of ^{266}Bh, and 10 correlations events were tentatively assigned to the decay chain of ^{266}Bh. And the rest correlations were assigned to decay chain of ^{267}Bh or not assigned.

As a present result [5], a state in ^{266}Bh, which decays by an alpha emission with the energies ranging from 9.05 to 9.23 MeV, feeds a state in ^{262}Db, which decays by alpha emission and by SF with a previously known half-life. The result provided a further confirmation of the production and identification of the isotope of the 113th element, ^{278}113, studied by a research group at RIKEN. The isotope ^{267}Bh, which is a reaction product of the 4n evaporation channel, was also produced and identified.

In this presentation, we will report the result of decay properties of ^{266}Bh and its daughter nuclei in detail.

References

[1] K. Morita *et al.*, J. Phys. Soc. Jpn. 73 (2004) 2593.
[2] K. Morita *et al.*, J. Phys. Soc. Jpn. 76 (2007) 045001.
[3] P. A. Wilk *et al.*, Phys. Rev. Lett. 85 (2000) 2697.
[4] Z. Qin *et al.*, Nucl. Phys. Review 23 (2006) 400 (Chinese journal in English)
[5] K. Morita *et al.*, J. Phys. Soc. Jpn. 78(2009) in press

Productions and Decay properties of ^{263,264,265}Hs

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Decay properties of hassium nuclides attract some physical interest because they have deformed magic proton number, $Z = 108$. The deformed neutron shell gap at $N = 152$ and 162 have also been found from systematics of α-decay energy and half-life values. Dvorak *et al.* studied $N = 162$ region of Hs nuclei [1]. However, neutron-deficient Hs isotopes had not been studied in detail. Oganessian *et al.* [2] and Ghiorso *et al.* [3] suggested the α-decay of ^{263}Hs, but the decay property of ^{263}Hs, such as α-particle energy and lifetime was not reported. On the other hand, productions and direct measurements of lifetime of only four atoms of ^{264}Hs were reported [4, 5].

In this work, the production and decay properties of ^{263,264,265}Hs were investigated by using the ^{208}Pb(^{58}Fe,n)^{265}Hs, ^{208}Pb(^{58}Fe,2n)^{264}Hs, ^{207}Pb(^{58}Fe,n)^{264}Hs, ^{206}Pb(^{58}Fe,n)^{263}Hs and ^{208}Pb(^{56}Fe,n)^{263}Hs reactions.

The experiment was performed at the linear accelerator facility in RIKEN (RILAC) from June 13 to 25, 2008. Ions of ^{58}Fe^{13+} and ^{56}Fe^{13+} were extracted from the 18-GHz ECR ion source and accelerated by RILAC. Beam energies of 284.1, 285.5, 287.0, 290.0 and 294.1 MeV for ^{58}Fe and 280.4 MeV for ^{56}Fe were used. Typical beam intensities were 4.5×10^{12} /s for ^{58}Fe and 2.3×10^{12} /s for ^{56}Fe.

The targets were prepared by vacuum evaporation of metallic lead on a 30 μg/cm^2 carbon backing foil. The target thicknesses were 400 μg/cm^2 for ^{206}Pb (Enrichment 99.3%), 430 μg/cm^2 for ^{207}Pb (Enrichment 98.3%) and 440, 480 and 500 μg/cm^2 for ^{208}Pb (Enrichment 98.4%). The targets were covered by a 10 μg/cm^2 thick carbon to protect the target from sputtering. Sixteen targets were mounted on a rotating wheel with a diameter of 30 cm. The wheel was rotated during the irradiation by 3000 rpm.

Reaction products were separated in flight from the beam by a gas-filled recoil ion separator, GARIS and were guided into a detection system at the focal plane of GARIS. GARIS was filled with helium gas at a pressure of 86 Pa. The magnetic rigidity of GARIS was set at 2.05 Tm. The transport efficiency of GARIS was estimated to be 80%.

Evaporation residues (ERs) are implanted into the silicon position-sensitive detector (PSD) after passing through the two timing counters. The PSD has an effective area of 60 mm × 60 mm. An identification of the products was based on genetic correlation of the mother and daughter nuclei. Four silicon detectors (SSDs) of the same size to the PSD but no position sensitivity were set in box shape in the backward direction of the PSD in order to detect the escaping α-particles from the PSD. The energy resolution for decay α–particles measured only by the PSD was 45 keV in FWHM while one for measured by sum of the PSD and SSD was 80 keV in FWHM. The counting rate of 'decay' signals (no signal in timing counters) was about 0.3 cps. The detection system was checked by detecting ERs from the $^{\mathrm{nat}}$Ce + ^{58}Fe reaction. The details about separator and detector are reported elsewhere [6, 7].

For ^{263}Hs, we have observed nine α-decay chains in total. The average property of the decay chains of ^{263}Hs is shown in Fig.1 [6]. If we assume that the highest α-particle energy group 10.82 MeV is from the ground state to ground state transition, the Q-value of the α-decay (Q_α) can be deduced to $Q_\alpha = 10.99$ MeV. This value is in agreement with some theoretical predictions, such as $Q_\alpha = 10.89$ MeV by Koura *et al.* [8] and $Q_\alpha = 11.20$ MeV by Goriely *et al.* [9]. For ^{264}Hs, we have observed new α-particle energy groups and enhanced statistical precision of the decay data. We have also measured production cross-section for the ^{58}Fe + ^{208}Pb reaction in five energy points.

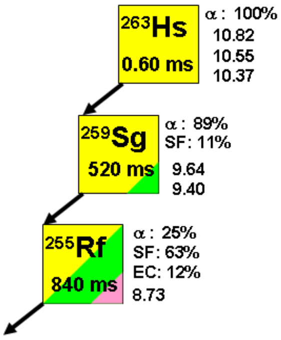

Figure 1: The average properties of the decay chains of ^{263}Hs. Energies are listed in MeV. The times given are half-lives [6].

References

[1] J. Dvorak *et al., Phys. Rev. Lett.*, **2008**, *100*, 132503.
[2] Yu. Ts. Oganessian *et al., Z. Phys. A*, **1984,** *319*, 215.
[3] A. Ghiorso *et al. Phys. Rev. C*, **1995**, *51*, R2293.
[4] G. Münzenberg *et al., Z. Phys. A*, **1987,** *328*, 49.
[5] Y. A. Akovali, *Nucl Data Sheets*, **1999,** *87*, 249 and references therein.
[6] D. Kaji *et al., J. Phys. Soc. Jpn.*, **2009**, *78*, 035003.
[7] K. Morita *et al., Eur. Phys. J. A*, **2004,** *21*, 257.
[8] H. Koura *et al., Prog. Theor. Phys.*, **2005,** *113*, 2073.
[9] S. Goriely *et al., At. Data and Nucl. Data Tables*, **2001,** *77*, 331.

"Reaction studies about the Q-value influence on the production of superheavy elements"

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Superheavy elements owe their existence exclusively to nuclear shell effects which stabilize them against spontaneous fission (SF). Theoretical predictions located the center of stability at the hypothetical doubly-magic spherical nucleus with Z=114 and N=184. This picture of an island of relatively long-lived nuclei far from the known quasistable ^{238}U was modified with the inclusion of higher orders of deformation. These calculations [1] suggested the ground state shell correction energy of deformed nuclei around Z=108 and N=162 to reach values as large as for ^{298}114. Relatively long partial SF half-lives were calculated for nuclei near ^{270}Hs, rendering α-decay the predominant decay mode. The doubly-magic nucleus ^{270}Hs has been observed for the first time by J. Dvorak, et al. in *4n* evaporation channel of the reaction ^{26}Mg + ^{248}Cm using highly efficient radiochemical on-line separation and detection system COMPACT [2]. In further experiments, the excitation function of this reaction has been measured at five different beam energies with the observation of a new isotope ^{271}Hs produced in 3n evaporation channel [3]. Recently, the formation of ^{270}Hs in a 4n evaporation channel in the fusion reactions with different asymmetry, ^{26}Mg + ^{248}Cm, ^{30}Si + ^{244}Pu, ^{36}S+ ^{238}U, and ^{48}Ca + ^{226}Ra has been studied by calculations using a two-parameter Smoluchowski equation by Liu and Bao [4]. Due to the same compound nucleus and hence to the same exit channel, the cross section of all these reactions can only be influenced by entrance channel effects, as the asymmetry for example. Although compound nucleus formation is favorable for systems with larger mass asymmetry, the reactions ^{48}Ca + ^{226}Ra and ^{36}S+ ^{238}U are predicted to result in a higher cross section for the formation of ^{270}Hs due to a lower reaction Q value. In this work we want to investigate the influence of the entrance channel on the production cross section of complete fusion reactions near the Bass barrier and to proof the validity of the aforementioned theoretical predictions. The produced Hs-isotopes are separated by chemical means as HsO_4 and measured using our cryo thermochromatography system COMPACT, which is more efficient for Hs-nuclei than any recoil separator. The measurement of the nuclear fusion reactions ^{36}S+ ^{238}U, using our chemistry setup at GSI, Darmstadt, and ^{48}Ca + ^{226}Ra using the Gas-Filled Recoil Separator at the FLNR, Dubna, have been started so far. During the first experiment on the former reaction, one possible ^{270}Hs-decay has been observed (Fig.1) while during the first two experiments on the latter reaction six ^{270}Hs decays could be identified. While the reaction cross section of the reaction ^{238}U(^{36}S, 4n)^{270}Hs is about one order of magnitude lower (Fig.2), than the aforementioned theoretical prediction the cross section for the reaction ^{226}Ra(^{48}Ca,4n)^{270}Hs agrees more or less with the predictions.

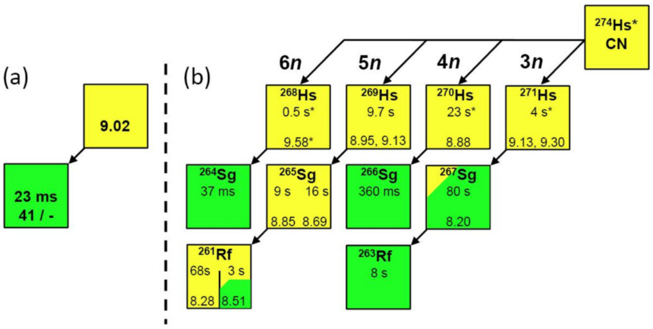

Figure 1: The observed decay chain (a) and decay properties of the $^{268\text{-}271}$Hs isotopes and their daughters (b)

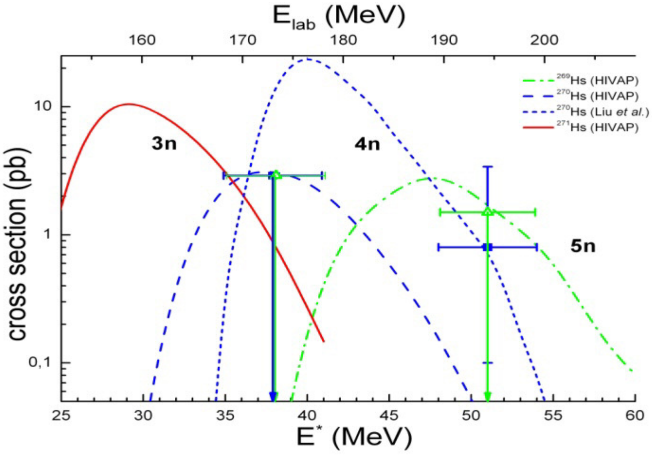

Figure 2: Production cross sections and limits measured for the reaction ^{238}U(^{36}S, 3-5n)$^{269\text{-}271}$Hs in comparison with HIVAP calculations and calculations from [4]. Quadrates correspond to ^{270}Hs (4n channel) and triangles correspond to ^{269}Hs (5n channel)

Due to the usage of a gas filled recoil separator during the ^{48}Ca experiments, the half life of ^{270}Hs could be measured for the first time and the previously published new decay properties for ^{270}Hs [2] could be confirmed.

References

[1] Z. Patyk, A.Sobiczewski, *Nucl. Phys. A* **533**,132 (1991).
[2] J. Dvorak *et al.*, *Phys. Rev. Lett.* **97**, 242501 (2006).
[3] J. Dvorak *et al.*, *Phys. Rev. Lett.* **100**, 132503 (2008).
[4] Z. H. Liu, J.-D. Bao, *Phys. Rev. C* **74**, 057602 (2006).

Decay properties of "chemistry isotopes" of light even-Z transactinides

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Chemical investigations of the light even-Z transactinides Rf, Sg, and Hs have traditionally been performed with only few, naturally the most long-lived, isotopes. Rf chemistry was mostly performed with ^{261a}Rf, occasionally with ^{257}Rf or ^{259}Rf. Sg experiments were reportedly conducted with ^{265,266}Sg according to the original papers, or with shorter-lived ^{263}Sg. Chemical studies of Hs used $^{269-271}$Hs. The heavier two of these isotopes were discovered in the course of these studies.
Improved data on the decay properties of all these isotopes – a clear benefit to chemistry experiments – were obtained in recent years and are presented here. Isomeric states in ^{261}Rf and ^{265}Sg were found. Here, the notation "g" for ground state and "m" for isomeric state is used when the states were fixed while "a" and "b" denote states where it is unclear, which one (if any) is the ground state.

Rutherfordium: ^{257g,m}Rf, ^{259}Rf, and ^{261a,b}Rf

An isomeric state in ^{257}Rf was known for some time [1], and improved data on its half-life were obtained recently [2].
Historically, the question concerning a potential SF branch in ^{259}Rf has been important. New studies found an electron-capture (∈) branch leading to ^{259}Lr that partially decays by SF [3]. This explains apparent SF from ^{259}Rf well.
About 30 years after the discovery of ^{261}Rf [4], an isomer was found in this isotope [5]. First it was observed exclusively following α-decay of ^{265}Sg [6]. Recently, evidence was obtained for its production also as an EVR in the reactions ^{244}Pu(^{22}Ne,5n) [7] and potentially ^{238}U(^{26}Mg,3n) [3].

Seaborgium: ^{263a,b}Sg and ^{265a,b}Sg

Two states are known in ^{263}Sg [8]. Recently, an SF branch in one of the two known states was found [9].
The story of ^{265}Sg is much more interesting. At the time of the first Sg chemistry studies [10-13], the authors of these publications believed to work with ~7-s ^{265}Sg and ~21-s ^{266}Sg [14], both decaying predominantly by α-decay. Recent measurements of the ^{26}Mg+^{248}Cm reaction [5], where the α-decay precursors of these isotopes were produced in the 4n and 5n exit channels led to the surprising observation that ^{266}Sg decays by SF with a rather short half-life of ~360 ms. A reanalysis of all decay chains ascribed to ^{265,266}Sg showed that the data were consistent with the interpretation that all Sg decays originated from ^{265}Sg [6]. In addition, evidence for an isomeric state was found in ^{265}Sg [6]. Recent reports from RIKEN [15] seem to confirm the results of this analysis.

Hassium: ^{269}Hs, ^{270}Hs, and ^{271}Hs

All chemical studies of Hs used the isotopes $^{269-271}$Hs produced in the reaction ^{248}Cm(^{26}Mg,3n-5n). There is additional data available for ^{269}Hs from the decay chains starting at ^{277}112 [16,17], where the observed α-particle energies were slightly higher. This may be due to conversion electron summing, which was less pronounced in, e.g., [5].
^{270}Hs was recently also observed in the reactions ^{226}Ra(^{48}Ca,4n) and ^{238}U(^{36}S,4n) [18].

For many nuclei with isomeric states, improved data on the isomeric production ratio as well as on feeding patterns between connected isotopes was obtained.

A summary of the decay properties of these isotopes based on the data given in the text is displayed in Tab. 1.

Isotope	$T_{1/2}$ (s)		E_α (MeV)		branching (%)	
^{257g}Rf	7.2	[2]	8.3-8.9	[1,2]	∈:14; SF <1.4	[1,2]
^{257m}Rf	4.1	[2]	9.02, 8.97	[1,2]		
^{259}Rf	2.5	[3]	8.77, 8.87	[19]	∈: 15	[3]
^{261a}Rf	68	[20]	8.29	[5]	SF: <11	[21]
^{261b}Rf	3	[5]	8.51	[5]	SF: 91	[5]
^{263a}Sg	0.9	[8]	9.06	[8]	SF: 13	[9]
^{263b}Sg	0.3	[8]	9.25	[8]	α: 100?	
^{265a}Sg	9	[6]	8.85	[6]	α: 100?	
^{265b}Sg	16	[6]	8.70	[6]	α: 100?	
^{269}Hs	10	[16,17]	8.95, 9.13	[5]	α: 100?	
^{270}Hs	~23*	[5]	8.88	[5]	α: 100?	
^{271}Hs	~4*	[5]	9.13, 9.30	[5]	α: 100?	

* estimated from measured E_α

Table 1: Decay properties of "chemistry isotopes" of Rf, Sg, and Hs. Given are the half-life ($T_{1/2}$), the energy of the main α-lines (E_α), and the known branching into different decay modes.

References

[1] F.P. Heßberger et al., Z. Phys. A. **359** (1997) 415.
[2] I. Dragojević et al., PRC **78** (2008) 024605.
[3] J.M. Gates et al., PRC **77** (2008) 034603.
[4] A. Ghiorso et al. Phys. Lett. **32B** (1970) 95.
[5] J. Dvorak et al., PRL **97** (2006) 242501; **100** (2008) 132503.
[6] Ch.E. Düllmann et al., PRC **77** (2008) 064320.
[7] A. Gorshkov, contribution to this workshop.
[8] S. Hofmann, Rep. Prog. Phys. **61** (1998) 639.
[9] K.E. Gregorich et al., PRC **74** (2006) 044611.
[10] M. Schädel et al., Nature **388** (1997) 55.
[11] M. Schädel et al., RCA **77** (1997) 149; **83** (1998) 163.
[12] A. Türler et al., Angew. Chem. Int. Ed. **38** (1999) 2212.
[13] S. Hübener et al., RCA **89** (2001) 737.
[14] A. Türler et al. PRC **57** (1998) 1648.
[15] H. Haba, priv. comm.
[16] S. Hofmann et al., Eur. Phys. J. A **14** (2002) 147.
[17] K. Morita et al., J. Phys. Soc. Jpn. **76** (2007) 043201.
[18] R. Graeger et al., contribution to this workshop
[19] C.E. Bemis et al., PRC **23** (1981) 555.
[20] M. Asai, contribution to TAN07, 2007, Davos, Switzerland.
[21] B. Kadkhodayan et al., RCA **72** (1996) 169.

Studies of the reaction ^{244}Pu(^{48}Ca,3-4n)^{288,289}114 at TASCA

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In the past few years, the new gas-filled TransActinide Separator and Chemistry Apparatus TASCA was installed and commissioned at the GSI in Darmstadt [1]. The year 2008 marked the transition from commissioning towards a science-driven program. As a first highlight experiment in the region of the heaviest elements, the reaction ^{244}Pu(^{48}Ca,xn)$^{292-x}$114 was studied in the Summer of 2009.

The main aspects driving this experiment were the following:

-production of element 114 in the ^{48}Ca+^{244}Pu reaction with a rather large cross section of about 5 pb is claimed by the Dubna-Livermore collaboration [2] but has not been reproduced independently;

-the isotopes produced in the 3n and 4n channel of this reaction, ^{289}114 and ^{288}114, respectively, are reported to be long-lived enough for gas-phase chemical experiments [3]: $T_{1/2}(^{289}114) = 2.6^{+1.2}_{-0.7}$ s and $T_{1/2}(^{288}114) = 0.80^{+0.27}_{-0.16}$ s;

-all of the few experiments that verified at least part of the results from FLNR either reported cross sections that were lower than those reported from the Dubna-Livermore collaboration [4-6] or did not report any cross section at all [6,7];

-a number of experiments reported cross section limits lower than the FLNR cross sections [6,8,9];

-excitation functions have not been measured at labs other than FLNR, with the exception of [5], where, however, the statistics was rather poor;

-the chemistry of element 114 is unknown. An experiment to study chemical properties of this element is currently (September 2009) running at TASCA [10]. A preceding experiment showing that TASCA is ready for experiments with single short-lived atoms of superheavy elements produced with cross sections of the order of a few pb was a prerequisite for this experiment.

Measurements of an (at least partial) excitation function to reliably determine the maximum cross section of the ^{48}Ca+^{244}Pu reaction as well as of the magnetic rigidity of the produced element 114 isotopes in pure He with good statistics were thus most important in the design of the experiment.

At the workshop, a preliminary account of the experiment will be given.

References

[1] M. Schädel, contribution to this workshop.
[2] Yu. Ts. Oganessian et al., Phys. Rev. C **69** (2004) 054607.
[3] Yu. Ts. Oganessian, J. Phys. G **34** (2007) R165.
[4] S. Hofmann, Eur. Phys. **32** (2007) 251.
[5] L. Stavsetra, accepted for publication in Phys. Rev. Lett., and H. Nitsche, contribution to this workshop.
[6] R. Eichler et al., Nucl. Phys. A **787** (2007) 373c.
[7] R. Eichler et al., Angew. Chem. Int. Ed. **47** (2008) 3262.
[8] K.E. Gregorich et al., Phys. Rev. C **72** (2005) 014605.
[9] R. Eichler et al., Radiochim. Acta **94** (2006) 181.
[10] A. Yakushev, contribution to this workshop.

Adsorption Interaction of ^{113m}In and ^{212}Pb Isotopes with Quartz

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Investigation of chemical properties of super heavy elements (SHE) produced in ^{48}Ca induced nuclear fusion reactions with actinide targets [1] is a challenging task for many research groups in the world. One of the main approaches to perform such experiments is based on series of model experiments involving lighter homologues of SHE. Only a limited number of methods are available for studying the chemical behavior of SHE and their lighter homologues in liquid phase and gas-phase [2]. Recently, by using gas adsorption chromatography, based on the well-known Insitu-Volatilization and On-line detection setup (IVO) with the Cryo-On-Line Detector (COLD), the chemical interaction of element 112 was investigated [3]. In the present research an experimental determination of $\Delta H_{ads.}$ of carrier-free ^{113m}In and ^{212}Pb isotopes on quartz surface was performed in preparation of experiments with the elements 113 and 114.

Experimental

Lighter homologues of SHE elements ^{113m}In ($T_{1/2}$=99 min) and ^{212}Pb ($T_{1/2}$=10.65 hrs) were prepared from home-made generator systems. For that purposes 0.5g of ^{nat}Sn were irradiated at the SINQ-NAA facility at PSI for 2hrs. The irradiated sample was dissolved in $HCl_{conc.}$ with a subsequent deposition onto an anion-exchange column. Using 1M HCl as an eluent separation of ^{113m}In isotope from parent ^{113}Sn was achieved [4] (Fig. 1). ^{113m}In was transformed into oxide form and dried on a Ta surface. This sample was used as source for thermochromatographic investigations with quartz as stationary surface. ^{212}Pb was prepared from a ^{nat}Th generator. $^{nat}ThF_2$ was dissolved under refluxing conditions in $HNO_{3(conc.)}$ and deposited onto a cation-exchange column. Using 1M HCl/CH_3OH (90:10 %) as eluent ^{212}Pb isotope was quantitatively separated from all by-products of the ^{232}Th decay [5].

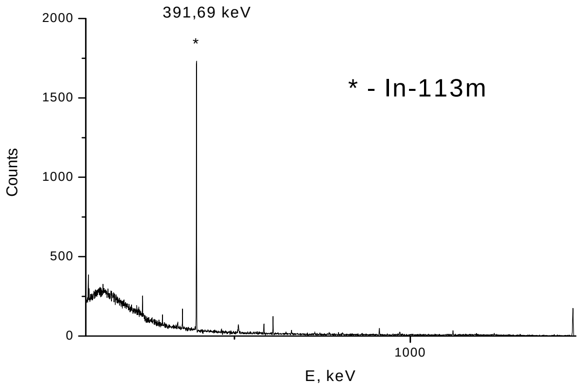

Figure 1: Gamma spectrum of ^{113m}In eluted from a Sn isotope generator

Results and Conclusions

The interaction of indium with quartz surface was investigated using a highly purified carrier gas to exclude trace amounts of water and oxygen. The entire thermochromatography column was encapsulated in a steel tube. The gas had to pass a Ta getter (1000°C) before hitting the In source deposited on Ta and heated up to 1000°C. Afterwards, the In was transported again over a hot Ta getter. This setup supposed to ensure the elemental state for the quite reactive indium. Reproducibility of obtained data was achieved by several repetitions of the experiment at the same conditions. The measurements of the distribution patterns have been carried out using a HPGE-γ-detector with a lead collimator having a window size of 1 cm. The microscopic Monte-Carlo simulation approach of gas adsorption chromatography was applied to evaluate the adsorption enthalpy of Indium on quartz as $\Delta H_{ads} = -140 \pm 10$ kJ/mol. (Fig. 2). The experiments with lead are recently under way.

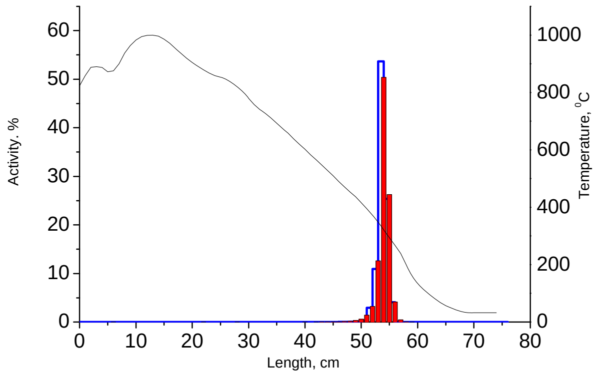

Figure 2: Thermochromatogram of ^{113m}In on quartz surface (red filled area – experimental results, blue line – results of Monte-Carlo simulations).

Acknowledgements

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References

[1] Yu. Ts. Oganessian et al., J. Phys. **2007,** G 34, 165-242
[2] M.Schädel Angew.Chem. Int. Ed. **2006,** 45, 368
[3] R. Eichler et al., Nature **2007**, 447, 72-75
[4] R. Eichler et al., Angew. Chem. Int. Ed. **2008**, 47, 3262–3266
[5] Z. Szeglowski et al., J. Radioanal. Nuclear Chem., **2005**, 265, 3, 367-370
[6] D.A. Goodwin et al., Nucleonics, **1966**, 24, 65-68

Chemical Investigation of Element 114

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The observation of long-lived isotopes of elements 112 and 114 at the Flerov Laboratory for Nuclear Reactions, Dubna, Russia (for review see [1]) inspired the investigation of their chemical behavior. Elements 112 and 114 are members of groups 12 and 14 of the periodic table, respectively. Relativistic effects in the electron structure of these superheavy elements (SHE) influence their chemical behavior and complicate the prediction of their chemical properties. With increasing atomic number Z the following typical trends are observed: 1) the first ionization potential and volatility are increasing along group 12; 2) the metallic character is enhancing in groups 13-16 [2-4]. On the other hand, relativistic calculations of the electronic structure of SHE predict closed shell electronic ground state configurations for elements 112($Rn{:}5f^{14}6d^{10}7s^2$) and 114($Rn{:}5f^{14}6d^{10}7s^27p_{1/2}{}^2$) leading possibly to a high volatility and a chemical inertness [4-11]. However, other relativistic calculation models predict an increased chemical inertness but still a chemical similarity to their lighter homologues [11-13] We present here the results of gas phase adsorption chromatographic experiments with ^{283}112 and $^{287-288}$114 revealing experimental evidence for an increased stabilization of the elemental state for both elements. Element 112 is shown to resemble a typical group 12 element forming a metallic adsorption bond with the chromatographic gold surface [14,15] (see Fig. 1) - a result which is recently well reproduced by theory. For element 114 a strong stabilization of its atomic state is most probably responsible for the observed evidence of a noble-gas like adsorption behavior revealing a more than 1000°C lower deposition temperature on gold compared to its homologue lead. New results from experiments performed in spring 2009 will be presented.

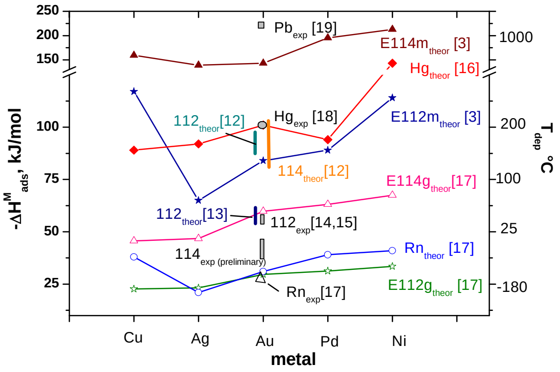

Figure 1: Experimental and theoretical adsorption enthalpies of the elements Hg, Pb, Rn, Element 112, and Element 114 (m – as metal, g - as noble gas) on the metals Cu, Ag, Au, Pd, and Ni (left hand side). Hypothetical (T_{dep}>40°C, not achievable in the used Cryo On-Line Detector (COLD)) and observed (T_{dep}<40°C) deposition temperatures under the experimental conditions of the transactinide experiments in the thermochromatography with COLD (right hand side).

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References

[1] Yu. Ts. Oganessian, *J. Phys.* G **2007**, 34, R165-R242 (2007).
[2] B. Eichler, *Kernenergie* **1976**, 19, 1976, 307-311.
[3] B. Eichler, PSI Report 00-09, Metal chemistry of transactinides. Villigen (2000) ISSN 1019-0643.
[4] B. Fricke, *Structure and bonding* **1975**, 21, 90-144.
[5] K.S. Pitzer, *J. Chem. Phys.* **1975**, 63(2), 1032.
[6] P. Pyykkö et al., *Acc. Chem. Res.* **1979**, 12, 276-281.
[7] P. Schwerdtfeger et al., in: Encyclopaedia of Computational Chemistry, Vol. 4, Wiley, New York, 1998, pp. 2480-2499.
[8] A. Landau et al., J. Chem. Phys. **2001**, 114, 2977-2980.
[9] W. Liu et al., Adv. Quant. Chem. **2001**, 39, 325-355.
[10] C. Nash, J. Phys. Chem. **2005**, A 109, 3493-3500 (2005).
[11] M. Seth et al. Angew. Chem. Int. Ed. **1998**, 37, 2493-2496 (1998).
[12] V. Pershina, et al., J. Chem. Phys. **2007**, 127, 134310 (2007).
[13] V. Pershina et al. 175 Years D.I. Mendeleev Anniversary Symposium FLNR, Dubna, Russia, January 2009
[14] R. Eichler et al., *Nature* **2007**, 447, 72.
[15] R. Eichler et al., *Angew. Chem. Int. Ed.* **2008**, 47(17), 3262.
[16] Eichler, B. et al. 1983 Radiochim. Acta 33, 121.
[17] Eichler, R., Schädel M.. 2002 J. Phys.Chem. B 106, 5413.
[18] Soverna, S. et al. 2005 Radiochim. Acta 93, 1.
[19] Eichler, B., et al. 1981 Preprint JINR P12-81-717, Dubna.

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Study of 114 chemistry

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Abstract not available at the time of print

Aqueous Chemistry of Rf and Db

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We have systematically investigated aqueous chemistry of the transactinide elements, rutherfordium (Rf) and dubnium (Db), at JAEA (Japan Atomic Energy Agency). Ion-exchange behavior of Rf and Db together with their lighter homologues in acidic solutions has been studied with the rapid ion-exchange separation apparatus AIDA [1]. It has been confirmed that Rf is a member of the group-4 elements and found that the fluoride complexation of Rf is significantly different from that of the homologues Zr and Hf. Different adsorption behavior of Db and the homologues Nb and Ta on the anion-exchange resin in HF solution has been also observed. In this report, we present fluoro complex formation of Rf and Db.

The isotopes of 78-s ^{261}Rf and 32-s ^{262}Db were produced via the ^{248}Cm(^{18}O, 5*n*) and ^{248}Cm(^{19}F, 5*n*) reactions, respectively, at the JAEA tandem accelerator. Ion-exchange chromatographic behavior of Rf and Db, and their homologues was investigated with AIDA.

Anion-exchange behavior of Rf was first studied in HF solution, and it was found that the adsorption behavior of Rf is quite different from that of the homologues [2, 3]. Then, the anion-exchange experiments with Rf was conducted in HF/HNO_3 solution to quantitatively understand the formation of anionic fluoride complexation of Rf. Distribution coefficients (K_d) of Rf and the homologues were measured as a function of the nitrate ion concentration $[NO_3^-]$ at the constant fluoride ion concentration $[F^-] = 3 \times 10^{-3}$ M. From the log K_d vs. log $[NO_3^-]$ plot, we unequivocally determined the species of Rf on the binding sites of the resin as the hexafluoro complex $[RfF_6]^{2-}$. The homologues are also to be present as $[MF_6]^{2-}$ (M = Zr and Hf), while the absolute K_d values of Rf are about two-orders of magnitude smaller than those of the homologues. In the log K_d against log $[F^-]$ relation, the result indicated that at $[NO_3^-] = 0.01$ M, the formation of $[MF_6]^{2-}$ for Zr and Hf occurs at $[F^-] \sim 10^{-5}$ M, while that of $[RfF_6]^{2-}$ starts at around 10^{-3} M. There was about two-orders of magnitude difference in the fluoride ion concentration between Rf and the homologues for the formation of the same hexafluoro complexes. This clearly demonstrates that the formation of the fluoro complexes of Rf is much weaker than those of the homologues Zr and Hf [4].

Cation-exchange behavior of Rf and the homologues in HF/0.1 M HNO_3 was also studied with the same manner as those in the anion-exchange experiments. The K_d values of Rf on the resin decreased with increasing $[F^-]$ in the range of 5×10^{-5} - 3×10^{-4} M due to the consecutive formation of fluoro complexes, while for the homologues the decreasing features of K_d were observed at about one-order of magnitude lower $[F^-]$ [5]. This means that the transition from cationic to neutral and then anionic fluoro complexes of the group-4 elements requires higher $[F^-]$ for Rf than for the homologues, establishing the following sequence of the strength of the fluoro complex formation, Zr ~ Hf > Rf.

The anion-exchange experiment with Db was conducted in HF, and it was found that the adsorption of Db on the anion-exchange resin was evidently smaller than those of the homologues Nb and Ta [6]. To obtain further information on the fluoride complexation of Db, the ion-exchange experiments are being performed in HF/HNO_3 solution with the improved AIDA system, AIDA-II, based on the model experiments with the homologues Nb, Ta and Pa [7, 8]. The result indicates that the adsorption of Db on the anion-exchange resin is quite different from that of the homologue Ta, while that is close to the adsorption of Nb and the pseudo-homologue Pa [9].

The present study at JAEA has been carried out in collaboration with RIKEN, Niigata University, Osaka University, Tokyo Metropolitan University, Kanazawa University, University of Tsukuba, GSI Helmholtzzentrum für Schwerionenforschung, and Mainz University.

References

[1] Y. Nagame *et al*., Radiochim. Acta **2005**, *93*, 519-526.
[2] H. Haba *et al*., J. Am. Chem. Soc. **2004**, *126*, 5219-5224.
[3] A. Toyoshima *et al*., J. Nucl. Radiochem. Sci. **2004**, *5*, 45-48.
[4] A. Toyoshima *et al*., Radiochim. Acta **2008**, *96*, 125-134.
[5] Y. Ishii *et al*., Chem. Lett. **2008**, *37*, 288-289.
[6] K. Tsukada *et al*., Radiochim. Acta **2008**, *97*, 83-89.
[7] Y. Kasamatsu *et al*., J. Nucl. Radiochem. Sci. **2007**, *8*, 69-72.
[8] Y. Kasamatsu *et al*., J. Radioanal. Nucl. Chem. **2009**, *279*, 371-376.
[9] Y. Kasamatsu *et al*., Presentation in this workshop.

Anion-exchange experiment of Db with AIDA-II

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Chemistry researches on element 105 (Db), the group-5 element in the 7th period, have been performed by comparative studies with its homologues Nb and Ta and pseudo homologue Pa [1]. There are, however, only few data and little is known about the chemical properties of Db. For a deeper understanding of the properties of Db, more detailed chemical investigations are required.

In dilute HF solution, it is reported that Nb forms oxofluoro complexes, while Ta and Pa are present as fluoro complexes, showing clearly different chemical behavior among the homologues. In our previous work, the anion-exchange behavior of Nb, Ta, and Pa in HF/HNO_3 solution was systematically investigated by a batch method [2], and significantly different behavior of the elements was observed. Here, we first report the results of on-line anion-exchange experiments with Nb and Ta in HF/HNO_3. Then, the anion-exchange behavior of Db studied by using a newly developed rapid ion-exchange and α-spectroscopy apparatus (AIDA-II) is presented.

Niobium-88 and ^{170}Ta were produced in the ^{nat}Ge(^{19}F, x*n*) and ^{nat}Gd(^{19}F, x*n*) reactions, respectively, at the JAEA tandem accelerator. Reaction products were continuously transported by a He/KF gas-jet system to the collection site of AIDA-II [3]. After the collection for 1-5 min, the products were dissolved in HF/HNO_3 solutions (0.010-5.0 M HF/0.040-0.60 M HNO_3) and were fed onto the column (ϕ1.0 × 3.5 mm) filled with the anion-exchange resin MCl GEL CA08Y at a flow rate of 1.2 mL/min. The effluent fractions were collected in 7 polyethylene tubes. The remaining Nb and Ta in the column were stripped with 0.015 M HF/6.0 M HNO_3 and collected in another 2 tubes. The samples were assayed by γ-ray spectrometry with a Ge detector to obtain elution curves Ta. Distribution coefficients, K_d, obtained from the elution curves agree well with those in the previous batch experiment [2] as shown in Fig. 1. This indicates that chemical reactions in the present system reach equilibrium.

Dubnium-262 was produced in the ^{248}Cm(^{19}F, 5*n*) reaction. The products deposited on the collection site of AIDA-II for 70–90 s were dissolved in a few hundred μL of HF/HNO_3 solutions (0.31 M HF/0.10 M HNO_3 and 0.89 M HF/0.30 M HNO_3; [F^-] = 0.0030 M) and were fed onto the column under the same experimental conditions as those with Nb and Ta. The eluate was collected on a 15 mm × 300 mm tantalum sheet which was continuously moving toward an α-particle detection chamber at 2.0 cm/s, as fraction 1. The sample on the sheet was automatically evaporated to dryness with a halogen heat lamp and then subjected to an α-particle measurement in the chamber equipped with an array of 12 silicon PIN detectors [3]. The remaining Db on the resin was stripped with 0.015 M HF/6.0 M HNO_3. The effluent was collected on another sheet as fraction 2 and was subjected to the evaporation and measurement in the same way. The anion-exchange procedure was repeated more than 1000 times under each condition. The number of α counts ascribed to the decay of 34-s ^{262}Db and its daughter 3.9-s ^{258}Lr was evaluated in each fraction. The percent adsorption (*%ads*) was obtained according to 100 × A_2 / (A_1 + A_2), where A_1 and A_2 are radioactivities in fractions 1 and 2, respectively. The K_d values of Db, being plotted in Fig. 1, were evaluated from the *%ads* values with the relationship between the *%ads* values and the K_d values of Nb and Ta. It is found that the adsorption of Db on the anion-exchange resin is considerably weaker than that of the closest homologue Ta in the periodic table and is similar to that of the lighter homologue Nb and also the pseudo homologue Pa under the present conditions.

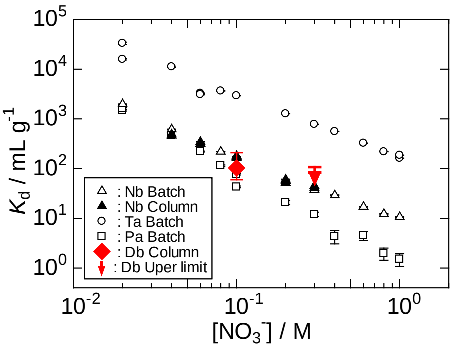

Figure 1: The K_d values of Nb, Ta, Pa, and Db as a function of [NO_3^-].

References

[1] M. Schädel, *Angew. Chem. Int. Ed.* **2006**, *45*, 368-401.

[2] Y. Kasamatsu, A. Toyoshima, H. Toume, K. Tsukada, H. Haba, K. Akiyama, T. Yoshimura, Y. Nagame, *J. Radioanal. Nucl. Chem.* **2009**, *279*, 371-376.

[3] K. Tsukada, Y. Kasamatsu, M. Asai, A. Toyoshima, Y. Ishii, H. Toume, Y. Nagame, *JAEA-Tokai TANDEM Ann. Rep. 2007, JAEA-Review 2008-054* **2008**, 67.

Electrochemical studies of the heaviest actinides

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It is interesting to study oxidation-reduction (redox) properties of the heaviest elements. Changes in electronic configurations of the heaviest elements appear in the stabilities of oxidation states (redox potentials) in aqueous solution. Thus, redox studies of the heaviest elements provide valuable information on the valence electrons influenced by relativistic effects. In this paper, our redox studies of the heaviest actinides using an electrochemical apparatus will be presented.

An electrochemistry apparatus combined with an ion-exchange chromatography technique available even for single atoms has been recently developed [1]; oxidation states of the single atoms can be identified based on chromatographic behavior. A working electrode is made by a bunch of glassy-carbon fibers (11 μmϕ) which are packed into a porous Vycor-glass tube (4.6 mm i.d. × 30 mm) as an electrolytic diaphragm to electrically connect to counter and reference electrodes. The surface of the carbon fibers is chemically modified with Nafion perfluorinated ion-exchange resin so that the working electrode is also employed as a cation-exchanger.

Electrochemical oxidation of nobelium ($_{102}$No) in 0.1 M α-hydroxyisobutyric acid (α-HIB) solution was performed using the apparatus [2]. The isotope ^{255}No (3.1 min) was produced in the ^{248}Cm(^{12}C,5*n*) reaction at the JAEA tandem accelerator. Reaction products transported by a He/KCl gas-jet method were deposited on a rapid chemistry assembly for 10 min. After collection, the products were dissolved with 0.1 M α-HIB (pH 3.9) and were subsequently fed into the column-type working electrode. The potential applied to the working electrode was adjusted to 0.2 to 1.2 V referred to an Ag/AgCl electrode in 1.0 M LiCl. The effluent from the column electrode was consecutively collected with a volume of 180 μL on 6 separate Ta discs. The remaining products in the column were stripped with 360 μL of 3.0 M HCl and collected on another 2 Ta discs. The 8 samples were evaporated to dryness using hot helium gas and halogen heat lamps and were then transferred to an α-spectrometry station equipped with eight 600 mm^2 passivated implanted planar silicon detectors. The ^{248}Cm target contained Gd to simultaneously produce ^{162}Yb (18.9 min) as the trivalent ion by the Gd(^{12}C,*x*n) reaction. After the α-particle measurement, the discs were assayed by γ-ray spectrometry to monitor elution behavior of ^{162}Yb^{3+}. The elution behavior of ^{81}Sr (22.2 min) and ^{162}Yb was investigated to establish clear separation between divalent (Sr^{2+}) and trivalent (Yb^{3+}) ions with the present system. The nuclides ^{81}Sr and ^{162}Yb were produced in the Ge(^{12}C,*x*n) and Gd(^{12}C,xn) reactions, respectively. The elution behavior was established by assaying effluent samples in plastic tubes with γ-ray spectrometry.

Figures 1(a) and 1(b) show the elution behavior of ^{255}No and ^{162}Yb under the applied potentials of (a) 0.2 V and (b) 1.2 V, respectively, while the elution of ^{81}Sr and ^{162}Yb in the separate experiment is depicted in Fig. 1(c). From the reference run with ^{81}Sr and ^{162}Yb, the clear separation between the trivalent and divalent ions is obvious; ^{162}Yb^{3+} is eluted with 0.1 M α-HIB, while ^{81}Sr^{2+} is present in the subsequent strip fraction of 3.0 M HCl. The elution positions of ^{81}Sr and ^{162}Yb are almost independent of the applied potentials in the studied range of 0.2 - 1.2 V. At 0.2 V, ^{255}No is detected in the 3.0 M HCl fraction while the monitored ^{162}Yb^{3+} is eluted earlier with α-HIB as shown in Fig. 1(a). The adsorption of ^{255}No is the same as that of ^{81}Sr^{2+}, clearly indicating that No is bound in its stable divalent state. On the other hand, the elution of ^{255}No in the 0.1 M α-HIB fraction is unambiguously observed at 1.2 V at the position of ^{162}Yb^{3+}. This behavior demonstrates that No^{2+} is successfully oxidized to No^{3+} and the trivalent state is complexed in the α-HIB solution with the present apparatus.

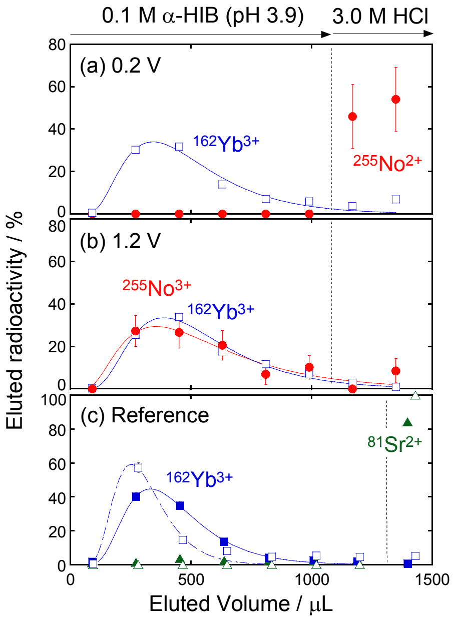

Figure 1: Elution behavior of ^{255}No and ^{162}Yb at the applied potentials of (a) 0.2 V and (b) 1.2 V. (c) Elution of the typical trivalent ^{162}Yb^{3+} and the divalent ^{81}Sr^{2+} in the reference experiment: close symbols at 1.2 V, open symbols at 1.2 V.

References

[1] A. Toyoshima *et al.*, *Radiochim. Acta*, **2008**, *96,* 323-326.
[2] A. Toyoshima *et al.*, to be submitted.

Solvent Extraction Studies of Molybdenum and Tungsten as Homologues of Seaborgium (Element 106)

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The pioneering cation-exchange experiments of seaborgium (element 106, Sg) were carried out by Schädel *et al.* in 1997 and 1998 [1,2]. There is, however, no report on aqueous chemistry of Sg following these works. Detailed chemical studies in various systems are required to clarify the chemical properties of Sg. In this work, solvent extraction behavior of molybdenum (Mo) and tungsten (W) which are lighter homologues of Sg was investigated. Carrier-free isotopes of Mo and W were produced in heavy-ion-induced nuclear reactions to make a reliable comparison of extraction behavior of mononuclear compounds of these elements with that of single Sg atoms because macro amounts of Mo and W form polyoxometalate compounds in aqueous solution below pH 8–9. Extractants used in this study were tetraphenylarsonium chloride (TPAC) or Aliquat 336 which extract anionic species in aqueous solution.

Molybdenum and tungsten isotopes, ^{90}Mo ($T_{1/2}$ = 5.7 h) and ^{173}W ($T_{1/2}$ = 7.6 min), were produced in the ^{nat}Ge(^{22}Ne,*x*n) and ^{nat}Gd(^{22}Ne,*x*n) reactions, respectively, using the RIKEN K70 AVF cyclotron. Tungsten isotopes, ^{173}W and ^{174}W ($T_{1/2}$ = 31 min) were also produced in the ^{nat}Dy(^{16}O,*x*n) reactions using the AVF cyclotron of Research Center for Nuclear Physics, Osaka University. Reaction products recoiling out of the target were transported by a He/KCl gas-jet system to a chemistry laboratory. The transported products were collected on a polyester or Naflon sheet and were then dissolved in 200 μL of 0.1–11 M hydrochloric acid (HCl). The concentrations of Mo and W isotopes in the aqueous phase were approximately 10^{-13} M. Equal volume of the 0.05 M TPAC-chloroform or the 0.05 M Aliquat 336-chloroform solution was mixed with the aqueous phase. The mixture was shaken for 15 min in the extractions of Mo with TPAC or for 3 min in the other experiments. After centrifuging, 160 μL aliquots of the aqueous and organic phases were taken into separate polypropylene tubes which were then subjected to γ-ray spectrometry using Ge detectors. Distribution ratios (*D*) were obtained from radioactivities of each phase.

Figure 1 shows the variations of the *D* values of Mo and W as a function of HCl concentration [HCl]. Tungsten was not extracted in 0.1–2 M HCl, suggesting that W exists as cationic and/or neutral species in the HCl concentration range. Both the distribution ratios of Mo and W rise with an increase of [HCl], indicating that the anionic chloride complexes of Mo and W are formed. It has been reported that the extracted species of Mo and W are $MoO_2Cl_3^-$ or $MoO_2Cl_4^{2-}$ and $WO_2Cl_3^-$, $WO_2Cl_4^{2-}$ or $WOCl_5^-$, respectively [3-5]. To examine the extracted species of Mo and W, the distribution ratios of Mo and W were also investigated in 11 M HCl as a function of Aliquat 336 concentration. The plots of log *D* vs. log Aliquat 336 concentration provided the linear slopes of 1.08 ± 0.04 for Mo and 1.21 ± 0.02 for W, showing that the net charges of the Mo and W chloride complexes are both -1. The extracted species of Mo and W are, therefore, $MoO_2Cl_3^-$ and $WO_2Cl_3^-$ or $WOCl_5^-$, respectively; the solvent extraction behavior of the mononuclear compounds of Mo and W is successfully investigated. For the chemical studies of Sg, further speciation analysis of the extracted compounds will be performed.

We are also developing a solvent extraction apparatus with microchips [6]. In the future, chemical experiments of Sg will be performed using a rapid chemistry assembly with the microchip.

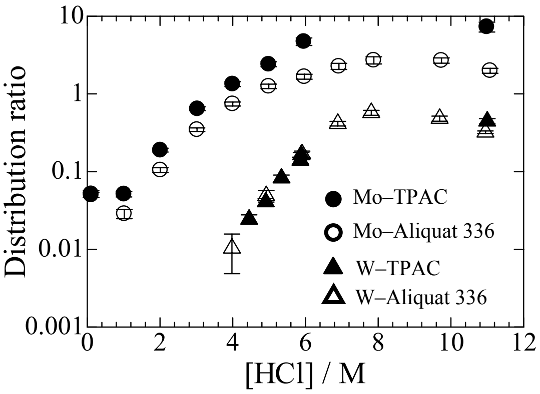

Figure 1: Extractions of Mo and W from hydrochloric acid with 0.05 M TPAC-chloroform and 0.05 M Aliquat 336-chloroform solutions.

References

[1] M. Schädel *et al., Radiochim. Acta* **1997**, *77*, 149-159.
[2] M. Schädel *et al., Radiochim. Acta* **1998**, *83*, 163-165.
[3] K. A. Kraus *et al., J. Am. Chem. Soc.* **1955**, *77*, 3972-3977.
[4] R. Caletka, V. Krivan, *J. Radioanal. Nucl. Chem.* **1990**, *142*, 373-382.
[5] I. Nelidow, R. M. Diamond, *J. Phys. Chem.* **1955**, *59*, 710-718.
[6] K. Ooe *et al. J. Nucl. Radiochem. Sci.* **2007**, *8*, 59-62.

Underpotential deposition – a method to study the chemistry of hassium

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The transactinide elements with Z = 108 - 116 are predicted to be noble metals. Therefore, electrodeposition represents a method to study the chemical behaviour of these transactinides and to prepare samples for α-spectroscopy in one step.

Electrodeposition of noble metals is influenced by the standard potential, the applied potential, the temperature, the oxidation state, and the concentration of ions in solution. This is described by the Nernst equation. In case of single atoms, the Nernst equation is not valid anymore, because the deposition behaviour is strongly influenced by the metal-metal interaction with the electrode material [1]. This phenomenon is called underpotential deposition.

Hummrich et al. [2] studied the deposition of tracer amounts of metal ions in detail and concluded that an electrolysis duration of around 10 sec is necessary for thermodynamic studies. ^{270}Hs seems to be a promising candidate for a first electrodeposition experiment with transactinides. Its half-life was recently estimated from the measured Q_α value via a Viola-Seaborg-type relationship to be around 23 s [3].

Recently, electrodepositions of short-lived Ru and Os isotopes were carried out. The Ru was produced in the ^{249}Cf(n_{th},f) reaction at the TRIGA Mainz reactor. The Os was produced in the ^{nat}Ce(^{40}Ar,xn) reaction, this was the first ever chemistry coupled on-line to TASCA at GSI [4]. In these experiments with Os as well as in the experiments with Ru, the nuclear reaction products were transported by a He KCl jet to a degasser and flushed with the electrolyte solution into the electrolysis cell. The cell is described in detail in [2].

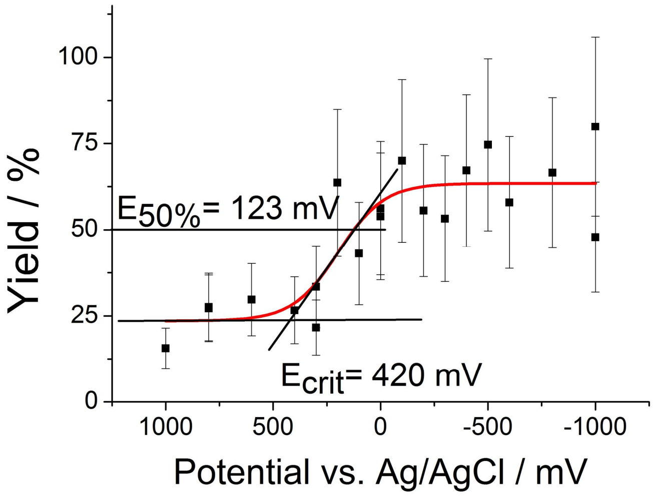

Figure 1: Deposition yield of Os on Pd electrodes as a function of the potential vs. Ag/AgCl.

The electrolyses were conducted at different potentials. After a preset electrolysis time, the electrodes were placed manually in front of γ detector and were measured. Figure 1 shows the deposition yield of Os on Pd electrodes.

Based on the results of these studies, a fully automated electrochemistry apparatus ELCH is under construction.

ELCH will be operated behind TASCA using physically preseparated isotopes. The evaporation residues will be thermalized in a recoil transfer chamber and transported by a KCl jet to ELCH. The KCl clusters will be collected in an ARCA type degasser and flushed into the electrolytic cell by the electrolyte solution. In ELCH, the two working electrodes are made from long metalized tapes which will be pulled through the electrolysis cell and stepped in front of an array of PIN diodes. The optimized surface area to electrolyte volume ratio will allow short electrolysis times and the tape system guarantees a fast transport of the samples from the cell to the detector. This is necessary for kinetic studies of the deposition behaviour of osmium and thermodynamic studies of the deposition of hassium.

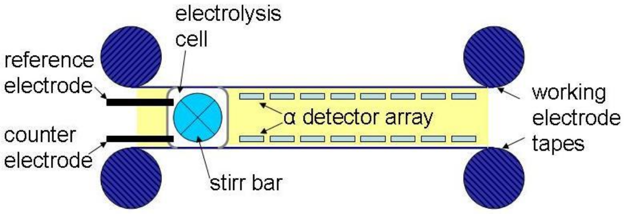

Figure 2: Schematic drawing of ELCH with the electrolysis cell (left) and the working electrode tapes stepped in front of an α-detector array.

[1] B. Eichler, J.V. Kratz, *Radiochim. Acta*, **2000**, *88*, 475-482.
[2] H. Hummrich et al., *Radiochim. Acta* **2008**, *96*, 73-83.
[3] J. Dvorak et al., *Phys. Rev. Lett.* **2008**, *100*, 132503.
[4] J. Even et al., *GSI Scientific Report 2007*, GSI-Report **2008**-*01*, 154.

A SISAK Extraction System for Chemical Studies of Hassium

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The fast solvent extraction system SISAK [1] was used to investigate the reaction between OsO_4 and NaOH as a model study for similar hassium experiments. Such a SISAK experiment would be one of the first attempts to study Hs in the liquid phase. The successful Rf SISAK experiments [2, 3], performed at LBNL in Berkeley, indicated that the SISAK system and its liquid scintillation detectors is sensitive enough to detect Hs, even though the cross section for producing Hs will be about 3 orders of magnitude lower.

The first investigation of the reaction between HsO_4 and NaOH was performed in a gas phase experiment [4]. The interaction of HsO_4 was slightly weaker with NaOH (though not quite clear) than that of OsO_4. This is in fair agreement with theoretical predictions [5]. Performing a SISAK extraction experiment in liquid phase as outlined in this work would give more detailed information about the reaction and would be a comparatively safe first-time Hs experiment with SISAK, since we already *know* that HsO_4 reacts with OH^- from the gas-phase experiments.

The reaction was studied with 2.7-min ^{181}Os produced at the Oslo Cyclotron Laboratory (OCL). Two high efficiency ("80%" and "50%") HPGe detectors were used to measure γ-rays from ^{181}Os. The distribution of OsO_4 between NaOH solutions and pure toluene was studied for different NaOH concentrations. See the resulting extraction curve in Fig. 1.

It is assumed that the reactions occurring are:

$$OsO_4(aq) + NaOH(aq) \leftrightarrow Na[OsO_4(OH)](aq) \qquad (1)$$

and

$$OsO_4(aq) \rightarrow OsO_4(org) \qquad (2)$$

The distribution ratio is:

$$D = \frac{[OsO_4]_{org}}{[OsO_4]_{aq} + [OsO_4(OH)]^-} \qquad (3)$$

which can be rewritten as:

$$D = \frac{K_D}{1 + K[OH^-]} \qquad (4)$$

where K and K_D are equilibrium constants for reactions (1) and (2), respectively. The solid line in Fig. 1 is a fit of equation (4) to the data and is in reasonable agreement with it, although not perfectly. A large number of experiments were performed in Oslo to carefully study the behavior of Os in this chemical system [6].

Based on the results from Oslo a "proof-of-principle" experiment with α-decaying Os isotopes was performed at the GSI Helmholtzzentrum für Schwerionenforschung in Darmstadt. Using α-decaying Os isotopes, the full SISAK setup [7] as it would be used for an Hs experiment with double α-detector arrays was tested. A beam of $^{40}Ar^{11+}$ from the UNILAC irradiated a ^{nat}Ce target in the gas-filled separator TASCA (Tran Actinide Separator and Chemistry Apparatus) [8] producing $^{172\text{-}175}$Os. In the focal plane of the separator a Recoil Transfer Chamber (RTC) was mounted. It was flushed with a He/O_2 mixture. To ensure fast and complete oxidation of Os to its volatile tetroxide, the gas transported Os directly to a quartz wool-plug heated to 600°C and then to the SISAK apparatus. This successful experiment proved the system to be suitable for studying Hs in principle. This was the first SISAK- run behind TASCA. Details about the results from the GSI experiment will be reported at the workshop.

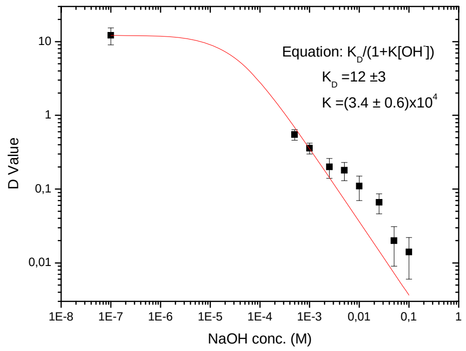

Figure 1: Distribution ratio of OsO_4 between NaOH solution and toluene vs. concentration of NaOH, obtained using the SISAK system in Oslo. The solid line shows the fitting of the result with equation (4).

References

[1] J.P. Omtvedt, et al., J. Alloy Compd. **1998,** 271-273, 303-306.
[2] J.P. Omtvedt et al., Journ. Nucl. Radiochem. Sciences, **2002**, 1, 121-124.
[3] L. Stavsetra, et al., Nucl. Instrum. Method Phys. Res. Sect. A, **2005**, 2-3, 509-516.
[4] A. von Zweidorf et al., Radiochim. Acta, **2004**, 12, 855-861.
[5] V. Pershina, Radiochim. Acta, **2005**, 7, 375-376.
[6] F. Samadani, Master thesis, University of Oslo, **2006**.
[7] J.P Omtvedt, et al., Eur. Phys. J. D, **2007**,45, 91-97
[8] M. Schädel, Eur. Phys. J. D, **2007**,45, 67-74

MicroSISAK – Improvements of a device for continuous liquid-liquid-extraction on a microliter scale

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For the investigation of the chemical properties of the heaviest elements (transactinides Z>103), fast and efficient chemical separation systems are required. Here, special challenges arise from low production rates in the order of a few atoms per minute in the case of Rf (Z=104) down to a few atoms per week for element 112 and short half-lives in the order of seconds.

One approach in this field is the fast centrifuge system SISAK based on liquid-liquid-extraction coupled to a liquid scintillation counter (LSC) for the detection of α-particles and spontaneous fission (SF) events. SISAK has successfully been applied in chemical investigations of element 104 [1-3]. However, this system requires flow rates in order of ml/s resulting in a high consumption of organic solvents and extracting agents. Furthermore, LSC is the only applicable detection method.

We report here on a “MicroSISAK”-system, a device based on microreaction technology. With this system, continuous liquid-liquid-extractions at flow rates in the order of ml/min are possible. The system is based on a commercially available micro-mixer[4] for efficient mixing of the two liquid phases and a subsequent teflon-membrane for fast phase separation. A scheme of the setup for our experiments is shown in figure 1.

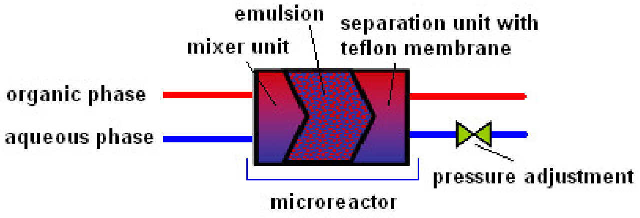

Figure 1. Schematic setup for experiments with MicroSISAK

Different prototypes of MicroSISAK have been designed at the IMM and tested with respect to mixing efficiency, separation power, and extraction yield. The separation of the two phases is caused by the lipophilic character of the teflon membrane and so it depends on the pressure, that is given to the emulsion. If we put back pressure from the outlet of the aqueous phase over the membrane, the separation yield can increase. An apparatus for the pressure adjustment was created and is shown in figure 2. The aqueous phase from the output of the MicroSISAK is connected to a vial of polyethylene-terephtalate where we can apply a constant pressure via a gas flow and a pressure regulator. The range of the regulator is from 0 to 1000 mbar with a sensitivity of 5 mbar.

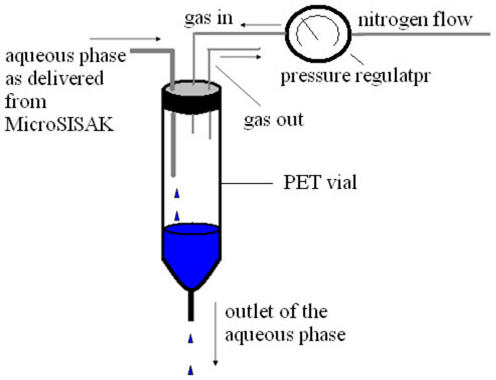

Figure 2: Scheme of the pressure regulation apparatus

For different flow rates, we adjust the back pressure at the aqueous phase until we get the best separation: This is a pure organic phase at the one outlet of MicroSISAK and no organic (just aqueous phase) in the PET vial. The results of the experiments show, that we need more back pressure for higher flow rates (see Fig. 3).

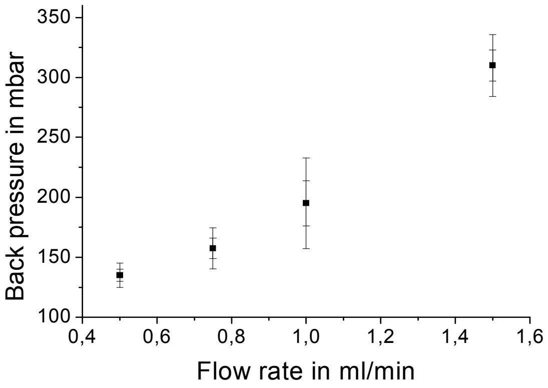

Figure 3: Required backpressure at different flow rates

Separation systems for Hf, the lighter homologue of Rf, and Tc, the lighter homologue of Bh, have been investigated.

Further experiments to investigate the chemical properties of Rf are currently being prepared.

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[1] J.P. Omtveedt, et al, *Journal of alloys and compounds* **1998**, *271-273*, 303-306
[2] J.P. Omtvedt, et al, *Journal of Nuclear and Radiochemical Sciences Vol. 3, No. 1* **2002**, 121-124
[3] J.P. Omtvedt, et al, Europeen Physical Journal D **2007**, *45*, 91–97
[4] http://www.imm-mainz.de (April **2009**)

Automated Chemistry Efforts at LLNL Using Eichrom's DGA Resin

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A new separation scheme for Db is being investigated based primarily upon Eichrom's DGA (N,N,N',N'-tetra-n-octyldiglycolamide (DGA Resin, Normal) or N,N,N',N'-tetrakis-2-éthylhexyldiglycolamide (DGA Resin, Branched)) products. Using Hf, Zr, Am, Nb, Ta, and Pa tracers the chemical differences between these elements in this system have been studied. Decontamination factors from actinide species and Rf homologs have been measured. How small of a separation system can be effectively utilized in accelerator-based experiments has been studied. Minimizing the scale of the chemical process allows the system to be performed as rapidly as possible, permitting greater flexibility in the range of half-lives that can be accessed by this liquid chemical system. Automation of the process is proceeding, which will allow the experiment to perform with minimal operator interaction. The system being created will permit other single-column chemistries to be employed with minimal system re-work. Dual or multiple column chemistries can be employed providing that the eluant from the first column is the load solution for the second, or can easily be changed with a mixing tee.

Extraction Chromatographic Studies of Rf and Db Homologs

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Due to the short half-lives of the transactinide elements, fast and efficient separations are necessary so that the chemical properties of these can be studied and compared to those of their lighter homologs. Separations based on extraction chromatography resins show promise for achieving the required short separation times, high yields and high separation factors that are necessary for transactinide studies. Extraction systems that can be applied to the study of element 104, Rf and element 105, Db have been investigated. The adsorption of Zr and Hf, the lighter homologs of element 104, on different resins has been investigated from various acid matrices to evaluate the suitability of these systems for the study of Rf. Similarly, Nb and Ta have been used to assess the system for Db.

TASCA – A New Tool in the Quest for Superheavy Element Studies

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The **T**rans**A**ctinide **S**eparator and **C**hemistry **A**pparatus (**TASCA**) project [1, 2], which is focusing on the separation and investigation of neutron-rich transactinide nuclides produced in actinide-target based reactions, has successfully finished its commissioning and has entered the research program which includes chemical investigations of superheavy elements (SHE) after preseparation with the gas-filled separator and nuclear structure and reaction studies.

The central device of TASCA is a gas-filled separator in a DQQ configuration operated either in the "High Transmission Mode" (HTM, DQ_hQ_v) or in the "Small Image Mode" (SIM, DQ_vQ_h) [1-5]. In the HTM, the unsurpassed transmission of TASCA is exploited. In contrast, the SIM provides unique possibilities due to its small spot size in the focal plane at a still relatively high transmission.

Extensive studies have been performed to obtain optimized parameter sets for (i) the target thickness and stability, (ii) the gas pressure and the gas filling (He, H_2, and mixtures), (iii) the dipole setting ($B\rho$) and quadrupole focusing, (iv) Recoil Transfer Chambers (RTCs) [6] (window material and thickness, support structures, and size and shape of the chamber), (iv) gas-jet transport of pre-separated products, and (vi) the coupling and performance of devices like ROMA and the Automated Rapid Chemistry Apparatus (ARCA). Results of many of these parameter studies were compared with TASCA model calculations [7] and very good agreement was achieved. These results confidently demonstrate that we are able to perform trustworthy SHE experiments with TASCA.

Efficiency measurements with catcher foils behind the target and in the focal plane showed very good agreement with model calculations for the fairly asymmetric reaction ^{22}Ne(^{181}Ta,xn)^{198m,199}Bi [2]. To confirm this agreement in a more symmetric reaction and to obtain a standard reaction to test and check the TASCA performance, detailed studies were performed with well known reactions of ^{48}Ca with targets of ^{206,207,208}Pb leading to ^{252,253,254}No. Assuming cross sections of 0.5 µb, 1.3 µb, and 2 µb [8] for the production of ^{252}No, ^{253}No, and ^{254}No, efficiencies of 54%, 56%, and 50%, respectively, were obtained for the HTM and a (80x36) mm^2 16-strip FPD; model calculations [7] predict 54%. Equally good is the agreement in the SIM where a 30% efficiency was measured for the reaction ^{48}Ca(^{208}Pb,2n)^{254}No.

As one of the crucial tests in the TASCA commissioning program, we studied the isotopes ^{260}Rf, ^{261a,261b}Rf, and ^{262}Rf synthesized in the very asymmetric reaction ^{22}Ne + ^{244}Pu [9]. The performance of TASCA was as anticipated. Efficiencies and magnetic settings were as expected. Rf isotopes were not only measured in the FPD but were also collected in an RTC and were transported to ROMA for decay measurements [9] and to ARCA for chemical investigations. One highlight of the experiment was the first transactinide chemistry behind TASCA. It was performed in ARCA with pre-separated 78-s ^{261a}Rf and it demonstrated that aqueous-phase transactinide chemistry behind TASCA can now be performed [10].

The new set-up termed *TA*sca *S*mall *I*mage mode *Spec*troscopy (TASISpec) [11] exploits the small spot in SIM. It provides a unique possibility to build a compact Si-detector box for α-, e$^-$, and SF-fragment measurements, and to pack Ge-detectors in close geometry, resulting in a highly efficient set-up for coincidence measurements with γ- and X-rays.

In conclusion, the performance of TASCA as a separator is well understood and, as the most versatile and highest efficient instrument in SHE research, it is ready to explore the physics and chemistry of SHE.

References

[1] M. Schädel *et al.*, GSI Sci. Rep. 2005, GSI Report 2006-1, **2006**, p. 262, and http://www.gsi.de/TASCA

[2] M. Schädel *et al.*, GSI Sci. Rep. 2008, **2009**, in print.

[3] M. Schädel, Eur. Phys. J. D **2007**, 45, 67.

[4] A. Semchenkov *et al.*, Nucl. Instr. and Meth. in Phys. Res. B **2008**, 266, 4153.

[5] Ch.E. Düllmann *et al.*, Nucl. Instr. and Meth. in Phys. Res. B **2008**, 266, 4123.

[6] Ch.E. Düllmann *et al.*, GSI Sci. Rep. 2006, GSI Report 2007-1, **2007**, p. 146.

[7] K.E. Gregorich *et al.*, GSI Sci. Rep. 2006, GSI Report 2007-1, **2007**, p. 144.

[8] Yu. Ts. Oganessian *et al.*, Phys. Rev. **2001**, C 64, 054606.

[9] A. Gorshkov *et al.*, GSI Sci. Rep. 2008, **2009**, in print.

[10] J. Even *et al.*, GSI Sci. Rep. 2008, **2009**, in print.

[11] L.-L. Andersson *et al.*, GSI Sci. Rep. 2008, **2009**, in print.

A new TASCA focal plane detector and data acquisition system

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The new, highly efficient, gas-filled TransActinide Separator and Chemistry Apparatus (TASCA) has been recently put into operation at GSI with the aim to study chemical and physical properties of superheavy elements with $Z \geq 104$. Based on the results of magnetic field model calculations of the dipole and the quadrupoles, two modes of operation of TASCA, the "High Transmission Mode" and the "Small Image Mode" have been realized [1]. The test experiments showed that the image size in the HTM is larger than the detector size of both detector types, as it was expected according to ion transport calculations and Monte-Carlo simulations [1,2]. The TASCA FPD working group has decided to build a detector setup based on a DSSSD as a stop detector and a backward array consisting of single-sided silicon strip detectors (SSSD). A *veto* detector for light fast ions that penetrate the DSSSD will be mounted behind the stop detector. The new setup will feature a (144 x 48) mm^2 large detector, which will accept > 90% of all evaporation residues reaching the focal plane. The geometrical detection efficiency for α-particles emitted from implanted nuclei will be > 70%. A DSSSD structure with an active area of (72 x 48) mm^2 has 72 strips on the front side and 48 strips on the back side. The strip width and the interstrip distance on the both sides of the DSSSD are 900 µm and 100 µm, respectively. Two adjacent DSSSDs form the stop detector with an active area of (144 x 48) mm^2. A SSSD structure with an area of (72 x 48) mm^2 has 8 strips, which are 72 mm long. The strip width and the interstrip distance of the DSSSD are 5.65 mm and 100 µm, respectively.

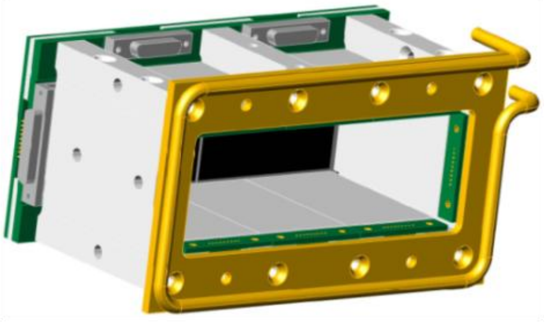

Figure 1: A drawing of the new TASCA FPD array.

In total, the new TASCA detector array has to process the signals of 320 strips. The signals are amplified using 10 32-channel preamplifiers and 40 8-channel amplifiers with integrated multiplexers. The amplifiers have two amplification ranges: x1 and x10. Totally 80 amplified analog signals (40 in the low energy range and 40 in the high energy range) are guided to 3 32-channel ADC CAEN V785. 3-Bit codes of strip numbers from multiplexers are stored in 4 32-bit registers SIS3620. One of them has 1 MHz time clock counter for time stamping. The data acquisition system is self-triggering; each signal above the threshold initiates readout of ADC modules and registers. To reduce the readout time a zero suppression mode is activated in all ADCs. The GSI MBS code and GO4 code are used for data readout and data visualization and analysis, respectively.

References

[1] A. Semchenkov et al. NIM B **266**, 4153 (2008).

[2] K. Gregorich et al. GSI Sci. Rep. 2006, 144 (2007).

From New Actinide Target Technology to Heavy Element Chemistry

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We are studying the chemical properties of the heaviest elements to explore if relativistic effects cause a noticeable change in chemical behavior compared to their lighter homologues. The heaviest elements are synthesized at the Lawrence Berkeley National Laboratory's (LBNL) 88-inch Cyclotron using the compound nucleus Berkeley Gas-filled Separator (BGS).

We have expanded the capabilities of the BGS for use of radioactive targets (^{244}Pu, ^{242}Pu), and thus have gained access to several significantly longer-lived isotopes of elements 104-108 with half lives long enough (^{261m}Rf, 78s; ^{263}Db, 34s; ^{265}Sg, 7s; ^{269}Hs, 10s) for chemical separations in aqueous and gas phases with more complex second generation chemical reaction systems, such as metal-organic-complexes.

Independent verification of element 114

We have independently verified the production of element 114 in the reaction of 244-MeV ^{48}Ca with ^{242}Pu [1]. Two chains of time and position correlated decays have been assigned to ^{286}114 and ^{287}114. The observed decay modes, half-lives, and decay energies agree with published results [2, 3]. The measured cross sections at a center-of-target energy of 244 MeV for the ^{242}Pu(^{48}Ca,3-4n)^{287,286}114 reactions were 1.4 +3.2/-1.2 pb each, which are lower than the reported values.

A new method to produce nuclear targets

We have developed a new method, Polymer-Assisted Deposition (PAD), to create crack-free homogenous metal oxide films for use as targets in nuclear science applications. Metal oxide films of europium, thulium, and hafnium were prepared as models for actinide oxides. Films produced by a single application of PAD were homogenous and uniform and ranged in thickness from 30 to 320 nm. The reapplication of the PAD method (six times) with a 10% by weight hafnium(IV) solution resulted in an equally homogeneous and uniform film with a total thickness of 600 nm. Thulium(III) oxide (Tm_2O_3) targets were irradiated with heavy-ion beams to test their performance for nuclear science applications. A 210-MeV ^{40}Ar beam with intensity up to 50 particle nA was used. Subsequent inspection of the targets with atomic force microscopy and scanning electron microscopy revealed no significant changes in surface homogeneity, and target flaking was not observed [4-5].

Heavy element mass analysis and detector facility

The study of production, decay, and nuclear structure of superheavy elements will be at the forefront of low energy Nuclear Physics research for years to come. Unfortunately, the atomic (Z) and mass number (A) assignments in all of these experiments are somewhat uncertain. We will build a facility for determination of the atomic (Z) and mass number (A) of single atoms of heavy elements. The headline experiment for this new facility will be the Z and A identification of superheavy elements. The facility will consist of an RF gas-stopper cell at the focal plane of the BGS followed by a mass analysis magnet for the determination of A .The mass-separated products will be transported to a low-background detector station where K X-rays of the daughter nuclides will be detected in coincidence with the α-decay, thus providing Z identification. Measurements of the nuclear structure of isotopes throughout the upper half of the periodic table will be possible. This single-atom Z and A identification will be a capability unique in the world, providing the framework for a far-reaching and enduring nuclear research program. .

References

[1] L. Stavsetra, K. E. Gregorich, J. Dvorak, P.A. Ellison, I. Dragojević, M.A. Garcia, H. Nitsche: Independent verification of element 114 production in the ^{48}Ca + ^{242}Pu reaction, PRL, submitted April 15, 2009.

[2] Yu. Ts. Oganessian, et al., Phys. Rev. C 70, 064609 (2004).

[3] Yu. Ts. Oganessian, et al., J. Phys. G 34, R165 (2007).

.

[4] M. A. Garcia, M. N. Ali, T. Parsons-Moss, P. D. Ashby, H. Nitsche: Metal Oxide Films Produced by Polymer-Assisted Deposition (PAD) for Nuclear Science Applications,. Thin Solid Films, doi:10.1016/j.tsf.2007.11.127 (2008).

[5] M. A. Garcia, M. N. Ali, N. N. Chang, T. Parsons-Moss, P. D. Ashby, J. M. Gates, L. Stavsetra, K. E. Gregorich, H. Nitsche: Heavy-ion irradiation of thulium(III) oxide targets prepared by polymer-assisted deposition. Nucl. Instrum. Methods A, doi:10.1016/j.nima.2008.04.077 (2008).

RIKEN GARIS as a Promising Interface for Superheavy Element Chemistry –Production of ^{261}Rf Using the GARIS/Gas-jet System–

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We have been developing a gas-jet transport system coupled to the RIKEN gas-filled recoil ion separator GARIS as a novel technique for superheavy element (SHE, $Z \geq 104$) chemistry [1,2]. The performance of the system has been investigated using ^{206}Fr, ^{245}Fm, and ^{255}No produced in the ^{169}Tm(^{40}Ar,3*n*), ^{208}Pb(^{40}Ar,3*n*), and ^{238}U(^{22}Ne,5*n*) reactions, respectively [1,2]. The results revealed that the GARIS/gas-jet system is a promising tool for next-generation SHE chemistry, i.e., identifying SHE nuclides under low background conditions with high efficiency of the gas-jet transport. In order to produce SHE nuclides with long half-lives for chemical experiments, hot fusion reactions based on a ^{248}Cm target should be considered. Since the last experiment with ^{255}No, we have developed a new gas-jet chamber having a large focal plane window of 100 mm diameter to efficiently collect evaporation residues. A rotating target system for the ^{248}Cm material was also installed. In this work, ^{261}Rf produced in the ^{248}Cm(^{18}O,5*n*) reaction was successfully extracted to a chemistry laboratory after the physical separation by GARIS.

A ^{248}Cm$_2$O$_3$ target of 280 μg cm^{-2} thickness was prepared by electrodeposition onto a 0.90 mg cm^{-2} Ti backing foil. The ^{18}O^{5+} ion beam was extracted from the RIKEN Linear Accelerator, RILAC. The beam energy was 95.5 MeV at the middle of the target, and the average beam intensity was 5 particle μA. GARIS was filled with helium at a pressure of 33 Pa. The magnetic rigidity of GARIS was set at 1.73 Tm. The evaporation residues of interest were separated in-flight from the beam and the majority of the nuclear transfer products by GARIS and then guided into the gas-jet chamber of 100 mm i.d. × 20 mm depth through a Mylar window of 0.5 μm thickness which was supported by a circular-hole (2.0 mm diameter) grid with 78% transparency. The ^{261}Rf atoms were stopped in helium gas, attached to KCl aerosol particles, and were continuously transported through a Teflon capillary (2.0 mm i.d. × 10 m length) to the rotating wheel apparatus MANON for α spectrometry. The flow rate of the helium gas was 2.0 L min^{-1}, and the inner pressure of the gas-jet chamber was 49 kPa. In MANON, the aerosol particles were deposited on 200-position Mylar foils of 0.5 μm thickness placed at the periphery of a 420 mm diameter stainless steel wheel. The wheel was stepped at 30-s intervals to position the foils between seven pairs of Si PIN photodiodes (Hamamatsu S3204-09). Each detector had a 38% counting efficiency for α particles, and the energy resolution was 60 keV FWHM for the detectors which look at the sample from the collection side.

Figure 1 shows the sum of α-particle spectra measured in the seven top detectors of MANON. The beam dose of 6.3×10^{17} was accumulated. As shown in Fig. 1, α peaks of ^{261}Rf (68 s, 8.28 MeV [3]) and its daughter ^{257}No (24.5 s, 8.222 and 8.323 MeV [3]) are clearly seen under the extremely low background conditions. A total of 168 α events were registered in the energy range of interest, including 58 time-correlated α pairs. The 7.687-MeV peak is due to ^{214}Po, a descendant of the natural radioisotope ^{222}Rn in the room. By comparing the spectrum measured with a focal plane Si detector in a separate experiment, the gas-jet transport efficiency of ^{261}Rf was evaluated to be 52±12%. The transport efficiency of GARIS was 7.8±1.7% for the focal plane of 100 mm diameter, referring to the cross section of 13 nb [4]. The production yield of ^{261}Rf at the chemistry laboratory is 0.5 atoms min^{-1} under the present experimental condition. These results suggest that the GARIS/gas-jet system is promising to explore new frontiers in SHE chemistry: (i) the background radioactivities originating from unwanted reaction products are strongly suppressed, (ii) the intense primary heavy-ion beam is absent in the gas-jet chamber and hence high gas-jet transport efficiency is achieved, and (iii) the beam-free conditions also make it possible to investigate new chemical systems that were not accessible before.

In the workshop, we will also present the recent result on ^{265}Sg produced in the ^{248}Cm(^{22}Ne,5*n*) reaction using the GARIS/gas-jet system.

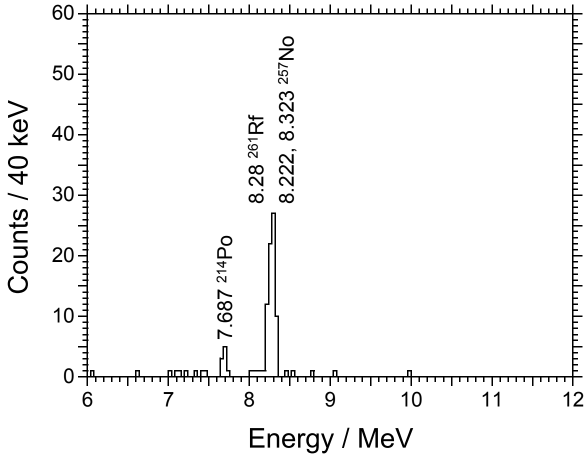

Figure 1: Sum of α-particle spectra measured in the seven top detectors of MANON.

References

[1] H. Haba *et al.*, *J. Nucl. Radiochem. Sci.* **2007**, *8*, 55.
[2] H. Haba *et al.*, *J. Nucl. Radiochem. Sci.* **2008**, *9*, 27.
[3] Ch. E. Düllmann, A. Türler, *Phys. Rev. C* **2008**, *77*, 064320.
[4] Y. Nagame *et al.*, *J. Nucl. Radiochem. Sci.* **2002**, *3*, 85.

PURECOLD for β–α–pile-up suppression – a status report

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The unambiguous identification of isotopes of the heaviest elements is a very important prerequisite for the investigation of their chemical properties. The time-resolved spectroscopic recording of consecutive decays of the investigated isotope as well as its descendents gives the highest possible confidence of assigning an observed event to the isotope of interest. However, the formation of unwanted by-products in the used nuclear reactions may lead to large amounts of unwanted events in energy regions where the decay of the investigated isotopes or their daughter nuclei is expected. Especially the specific decay of ^{212}Bi and ^{212}Po as decedents of ^{220}Rn might disturb the measurement due to the consecutive emission of high energetic β- and α-particles with a sum energy of more than 10.5 MeV within a very short time period of typically less then 1000 ns. Conventional spectroscopy electronics is not able to resolve the consecutive hits of the β- and α-particle in time leading to so called pile-up pulse height amplification.

First measurements using PURECOLD

A new spectroscopic detection system was designed to distinguish pile-up signals from real α-events. We present here some results obtained with the final hardware implementation of this measurement system (PURECOLD) using a pair of 250 mm^2 PIN diode detectors mounted face to face at a distance of 3 mm. On the surface of one of the PIN diodes ^{232}U was deposited to efficiently detect pure α-events as well as pile-up signals from the ^{212}Bi(β)/^{212}Po(α) decay.

The discrimination of pile-up signals is based on the sampling of the leading edge of the fast timing amplifier signal using sixteen consecutive fast voltage comparators (set-up time 150 ps). The voltage level of each comparator is individually programmable using digital analog converters. The time stamps of passing through these thresholds are recorded with a timing resolution of 0.5 ns. This results in a separation of the pile-up events from pure α-events in a time versus energy plot as depicted in Fig. 1. The time difference between two consecutive threshold passings will exceed 10 ns in the case if a preceding β-particle hits the detector before an α-event will be registered.

The spectroscopic part of PURECOLD consists of two independent acting amplifiers and analog digital converters to process events from 3 to 30 MeV and from 5 to 300 MeV using separated signal branches. In Fig. 2 a reconstructed α-spectrum from ^{232}U and its descendents is shown together with the spectrum of the identified pile-up events of the ^{212}Bi(β)/^{212}Po(α) pair.

To demonstrate the performance of the PURECOLD system we will present results of ongoing optimization measurements determining pile-up suppression factors and also half-live determinations from correlation measurements for the short-lived ^{216}Po, ^{215}Po, and ^{212}Po using a simple table top experiment as described above.

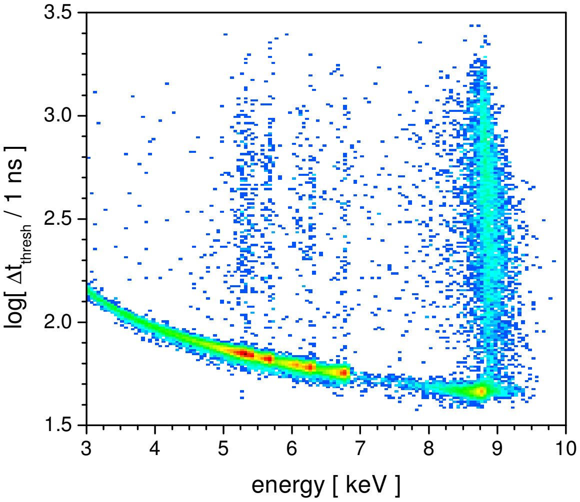

Fig. 1: Time difference between passing the first and 10th threshold versus α-particle energy. A gap from 55 ns to 75 ns separates the true α-events of ^{212}Po from the β-α pile-up events of the decay of ^{212}Bi and ^{212}Po.

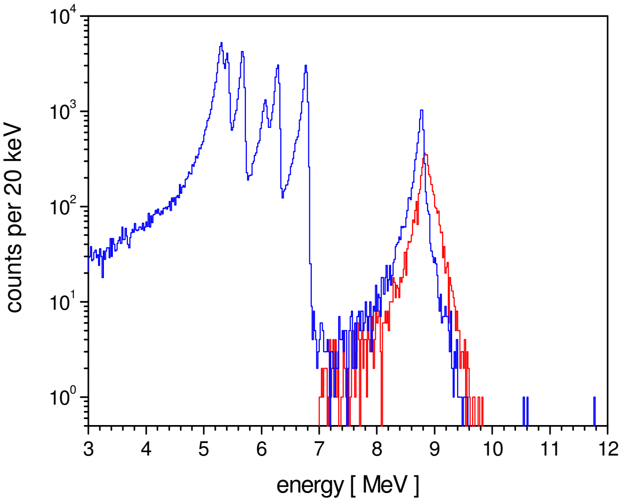

Fig. 2: α-spectra of ^{232}U and its descendants using the non-optimized PURECOLD spectroscopic system. Detected pile-up events are shown in red and the remaining events considered as purified alpha spectrum in blue.

Stopping force estimations for element 114 in Mylar and argon gas

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The use of the Dubna Gas-Filled Recoil Separator (DGFRS) [1] as a preseparation device for gas phase chemistry experiments with elements 112 and 114 led us to the task of reevaluating the stopping force (STF) of super heavy elements (SHE). In the experiment the DGFRS and the IVO-system [2] were separated by a thin Mylar window of vaiable thickness so that the evaporation residues (EVR) can pass it. The reaction product collection chamber (RPC-chamber), filled with gaseous argon and mounted behind the Mylar foil, had to be as small as possible to increase the transport efficiency to the thermochromatography detector COLD. Due to scarce availability, we measured STF data for Hg, Rn and No in Mylar and gaseous argon at energies of around 30 to 50 MeV. These data were used for an extrapolation of the STF of the SHE 114, eka-lead to be used in ongoing experiments.

Stopping force measurements

A 265 MeV $^{48}Ca^{18+}$ beam was delivered from the U-400 cyclotron at the Joint Institute of Nuclear Research (JINR) in Dubna, Russia. The beam was passed through a 1.6 µm Ti window, a 1.6 µm Ti target backing into the target materials. Three different target materials were irradiated: neodymium oxide ($^{nat}Nd_2O_3$, 300 µg/cm^2), dysprosium oxide ($^{nat.}Dy_2O_3$, 330 µg/cm^2) and lead (^{208}Pb, 420 µg/cm^2, 1 µm Cu degrader in front). The mainly produced evaporation residues in the fusion of ^{48}Ca with target nuclei are according to HIVAPSI [3] ^{185}Hg, ^{206}Rn or ^{254}No, respectively. The EVR's were separated in the DGFRS according to their momentum and guided on an exchangeable Mylar foil of 2, 3 or 5 µm thickness separating the RPC-chamber from the DGFRS. The RPC-chamber had a depth of 18 mm and was filled with thermalizing argon gas of a pressure between 0.0 and 0.4 bar. In the center of the focal plane of the separator at the back wall of the RPC-chamber a 20 x 20 mm silicon detector (PIN-diode) was mounted to detect the signals and the alpha decays of implanted products. To gain different data points for the evaluation the Mylar foil thickness and the argon pressure were varied.

Extrapolation to element 114

For the extrapolation of the stopping range to element 114 we used the assumption of proportionality of the STF of a heavy ion to the velocity dependent effective charge (q_{eff}) and the STF of a proton (see eqns. 1-3). This procedure requires that the heavy ion as well as the proton have the same incident velocity. In equation 2 v_{TF} is the Thomas Fermi velocity that includes the Bohr velocity v_0. Thus a proton STF can be derived which is uncorrected for any other STF terms.

$$-\left.\frac{dE}{dx}\right|_{Z_1} = q_{eff}^2\left(E_I, Z_1, A_1\right)\cdot\left[-\frac{dE}{dx}\right]_{Z=1} \qquad (1)$$

$$q_{eff}^2\left(E_I, Z_1, A_1\right) = Z_1^2\left(1 - e^{v_1/v_{TF}}\right) \qquad (2)$$

$$v_{TF} = Z_1^{2/3} v_0 \quad with \quad v_0 = c/137.036\ [m/s] \qquad (3)$$

These proton STF (S/q^2_{eff}) were plotted against $1/\ln(Z_1)$ an empirically established dependence of the STF on Z of the heavy ion. The obtained values were compared with the frequently used stopping code SRIM 2008 [4] and finally used to setup the gas chromatography experiment with elements 112 and 114.

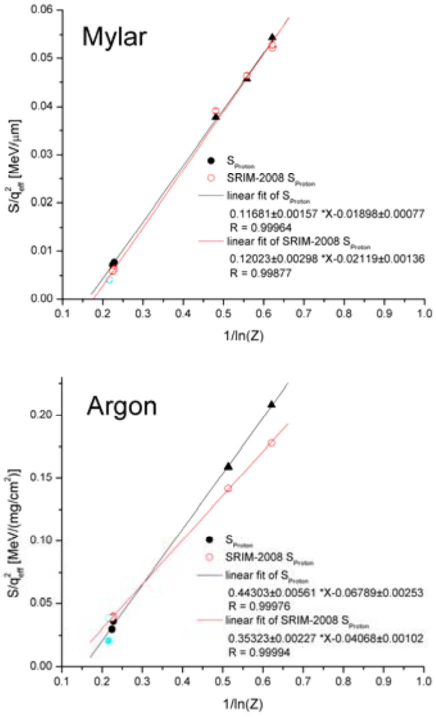

Fig. 1: *Stopping force dependence on Z.* Stopping force values from H. Paul [5] (full triangles) and from our measurements (full circles) in Mylar (upper graph) and gaseous argon (lower graph). The empty circles are simulated SRIM-2008 values. The lines (black for experimental values, red for SRIM-2008 values) represent linear regressions showing the dependence of the STF from the atomic number Z..

References

[1] K. Subotic et al., Nucl. Instr. Methods A, **2002**, 481, 65.
[2] Ch.E. Düllmann et al., Nucl. Instr. and Meth. A, **2002**, 479**,** 631.
[3] R. Dressler, PSI Scientific Report 1998, Volume I, **1999**, 131.
[4] J.F. Ziegler, et al., SRIM-2008, www.srim.org, **2008**.
[5] H. Paul, www.exphys.uni-linz.ac.at/stopping, **2003**.

Preparation of ^{244}Pu-targets by electrodeposition

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In the **T**rans**A**ctinide **S**eparator and **C**hemistry **A**pparatus TASCA at GSI, transactinide elements (Z>103) can be produced in fusion reactions with actinide targets. In a former beamtime at TASCA it was shown, that the production of Rf in the reaction ^{244}Pu(^{22}Ne, 4-6n)$^{260-262}$Rf is possible [1]. In the next experiment at TASCA, the element 114 should be produced in the reaction ^{244}Pu(^{48}Ca, 4n)^{288}114 and its chemical behavior should also be investigated.

Since ^{244}Pu is available only in very limited amounts, the target preparation technique should give high deposition yields. The electrochemical deposition is the most suitable technique for target preparation. To optimize the electrochemical deposition parameters, several depositions were done with ^{239}Pu [2]. This Pu-isotope is available in larger amounts.

The electrodeposition cell consists of polytetrafluorethylene PTFE. Deposition of ^{244}Pu is performed from an isobutanolic solution. A 2-µm thick banana shaped Ti foil of 1.74 cm^2 is the backing for the ^{244}Pu layer and serves as cathode. A Pd foil in the same geometry and size serves as anode. To prevent loss of the organic solution, the whole setup is cooled by two water-cooled Ti blocks at 15 °C. The plating-time is about 6 hours. The current density is limited to 1.15 mA/cm^2 (corresponding to a current of 2 mA). Investigations have shown that at a higher current, the Pu-layer peels off the Ti backing. The voltage is about 160 V in the beginning and about 200 V at the end.

The deposition yield was either determined by α-particle measurement of the target layer or by neutron activation analysis of the supernatant solution. For this, aliquots of the plating solution before and after the plating process have been irradiated at the TRIGA Mark II research reactor of the Institut für Kernchemie. The so produced ^{245}Pu could easily be measured by γ-spectroscopy [$T_{1/2}$ = 10.5 h, lines at 308 keV (5%), 327 keV (25%) and 560 keV (5%)]. This method is element- and isotope specific. The homogeneity of the ^{244}Pu layer is investigated by radiographic analysis [3]. Four targets were produced with areal densities as given in Table 1.

Target	^{244}Pu layer [µg/cm^2]
#562	673
#594	724
#623	790
#624	785

***Table 1**:* ^{244}Pu targets produced for the synthesis of element 114

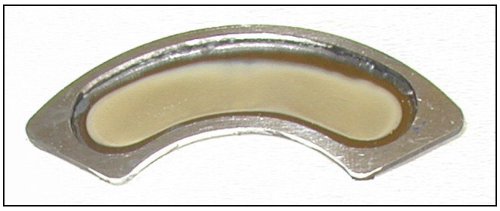

Figure 1: ^{244}Pu target #562, 673 µg/cm^2

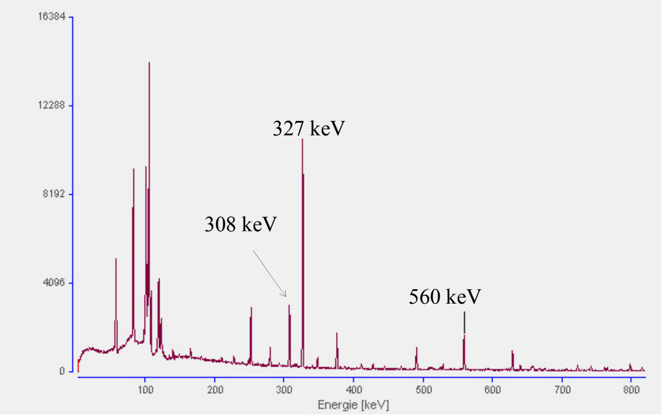

Figure 2: Gamma-spectra of ^{245}Pu in the supernatant solution

References

[1] A. Gorshkov et al., GSI Scientific Report 2008, p. 140, **2009**.

[2] J. Runke et al., *Die Herstellung von Plutoniumtargets für TASCA durch Elektrodeposition*, in: Jahresbericht UMZ, **2008**, in press.

[3] D. Liebe et al., *Improvements at the radiographic analysis of radioactive targets*, in: Jahresbericht UMZ, **2006**.

Coupling TASCA with SHIPTRAP

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The Trans-Actinide Separator and Chemistry Apparatus TASCA [1] at GSI is specially designed for maximum transmission of SHE produced in hot fusion reactions using actinide targets. The central part of TASCA is a gas-filled separator equipped with one dipole- (D) and two quadrupole magnets (Q). This configuration allows two operation modes. The DQ_hQ_v mode gives the highest possible transmission, whereas the smallest image size results from the DQ_vQ_h mode, where the indices indicate horizontally (h) and vertically (v) focusing quadrupoles. Using the reaction ^{206}Pb(^{48}Ca,2n)^{252}No the transport efficiency of TASCA was determined to be as high 54% in the high tramsmission mode and 30% in the small image mode.

With TASCA EVaporation Residues (EVRs) are separated from the heavy-ion (HI) beam and from a significant fraction of unwanted by-products of the nuclear reaction used to synthesize the element of interest. EVRs are extracted from the separator through a thin window. This separates TASCA's low-pressure region ($\leq$ 1 mbar He) from the Recoil Transfer Chamber (RTC) in which EVR are thermalized at about ambient pressure and made available for transport to a chemical or physical separation device using a gas-jet transport system. Recently this technique has successfully been applied for chemical studies of Rf produced in the reaction ^{244}Pu(^{22}Ne,5n)^{261}Rf.

The SHIPTRAP [2] facility at GSI is a Penning-trap mass spectrometer currently coupled to the in-flight velocity filter SHIP [3]. SHIPTRAP consists of three basic elements: (i) a stopping gas cell with a radiofrequency quadrupole (RFQ) for subsequent extraction of the stopped ions, (ii) a beam cooler and buncher RFQ trap and (iii) the Penning trap unit for mass measurements. Recently, first direct mass measurements of No-isotopes have been performed with SHIPTRAP using the reaction ^{206}Pb(^{48}Ca,2-4n)$^{252-254}$No and SHIP as a pre-separator. The EVR, as delivered from SHIP, are stopped in the gas cell (He, 50 mbar) and extracted as a quasi-continuous low energy beam into the buncher trap. Here they are cooled due to collisions with a buffer gas and subsequently transported in bunched mode to the Penning trap unit for mass measurement.

We report here on a plan to connect TASCA with SHIPTRAP in order to benefit from the high transport efficiency of TASCA for asymmetric fusion-evaporation reactions which are not applicable at SHIP so far. Coupling will be performed via a gas-jet transport system using carbon as aerosol material. The performance of a gas-jet loaded with C-clusters has recently been investigated at the TRIGA-reactor in Mainz. Here transport times of about 500 ms and transport efficiencies in the order of 70% have been determined. We assume that similar values can be achieved with the RTC connected to TASCA.

For the connection of a C-cluster gas-jet to SHIPTRAP a new ion source has to be developed. Two ion source concepts will be investigated:

- coupling of the gas-jet with a skimmer unit attached to a high-pressure ECR-source
- combination of laser ablation with a laser ion source or with an ECR-source, respectively

Both concepts will be tested at the TRIGA-TRAP facility [4] in Mainz before relocation to TASCA and SHIPTRAP. Figure 1 shows a schematic view of the set-up as planned so far.

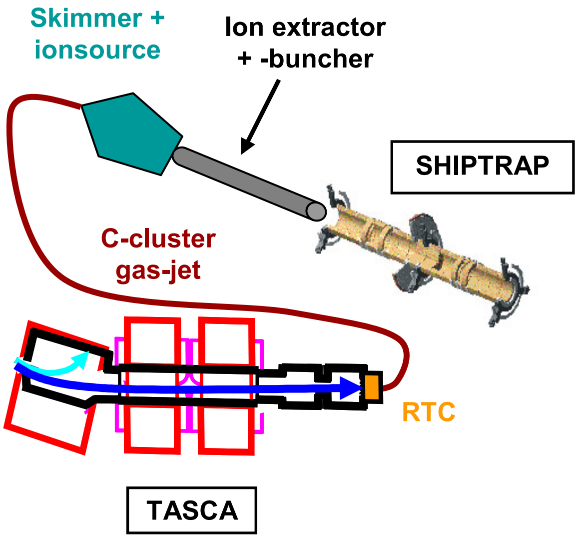

Figure 1: Coupling of TASCA and SHIPTRAP with a C-cluster gas-jet, a skimmer-unit and a subsequent ion source.

References

[1] M. Schädel et al., Eur. Phys. J. D 45 (2007) 67
[2] M. Block et al., Eur. Phys. J. D 45 (2007) 39
[3] G. Münzenberg et al., Nucl. Instrum. Methods 161 (1979) 65
[4] J. Ketelaer et al., Nucl. Instr. Meth. Phys. Res. A594 (2008) 162

The Helmholtz Institute Mainz: Structure, Symmetry, and Stability

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Shortly before Christmas 2008, scientists from Mainz University and from GSI proposed to establish a new research institute in the framework of the Helmholtz Gemeinschaft HGF on the campus of Mainz University. The proposal was examined by the president of HGF and writing of a full proposal was requested by him at the end of January 2009. The final proposal was delivered to the HGF on April 02, 2009 [1] and was reviewed by an international review committee on April 26-28 at Mainz. The unrestricted recommendation was to realize this institute as proposed. The establishment of the Helmholtz Institute Mainz (HIM) was approved by the senate of the HGF on June 5, 2009.

Structure

HIM consists of the following sections: Hadron Structure with Electromagnetic Probes EMP, Hadron Spectroscopy and Flavour SPECF, Symmetry of Matter and Antimatter MAM, Stability and Properties of Superheavy Elements SHE, Accelerator Physics and Intergrated Detectors ACID, and a Theory Floor THFL. Each sections has its own section leader heading scientific and technical personnel. In addition, the institute houses 3 Junior Research Groups.

Funding

In the steady state phase of HIM, funding by the HGF amounts to 5 M€/year. The state of Rhineland-Palatinate contributes another 0.5 M€/year and constructs a new institute building, 20 M€, and finances its operation.

SHE Section

The SHE section comprises two pillars. These are devoted to SHE physics and chemistry, respectively. Each is headed by a full professor (W3) linked to the University. Research fields are synthesis and decay of SHE, nuclear structure studies, atomic physics, and chemistry of SHE. Embedded into the program is the commissioning [2] of TRIGA-Trap with the development of single-ion mass measurements, and the development of collinear laser spectroscopy at TRIGA-Laser. The following milestones in SHE research are envisaged: Confirmation of element 114 production in the ^{48}Ca + ^{244}Pu reaction is foreseen, followed by a stepwise approach to new elements, e.g., element 120 in the ^{54}Cr + ^{248}Cm reaction. Nuclear structure studies will pinpoint Nilsson levels in deformed nuclei in the region Fm – Ds [3]. Embarking from the No region where direct mass measurements were performed [4], mass mapping of the SHE landscape with single ions will proceed to heavier elements. Coupling of SHIPTRAP to TASCA by a gas-jet system and an ECR source [5] will broaden the range of accessible nuclei in SHIPTRAP. Pioneering experiments on the chemical characterization of elements with Z>112 are planned. Here, an important question is whether the sub-shell closure of the $7p_{1/2}$ shell in element 114 makes this element a very volatile noble-gas like element with very weak van der Waals interaction with a Au surface. For lighter transactinides, electrodeposition [6] will be sensitive to the metal-matal interaction with the electrode material. Liquid-liquid extraction [7] can be used to study complex formation with inorganic ligands. A new field will focus on the formation of new compound classes, e.g., volatile metal complexes [8] or organometallic compounds.

ACID section

Because of the limitations of the UNILAC accelerator, i.e. 25% duty cycle, time sharing operation for FAIR, it is proposed that the ACID section, in close collaboration with GSI and U. Ratzinger, IAP/Frankfurt University, designs and constructs the prototype of the first cavity of a superconducting cw linac with 100% duty cycle. The invest budget for this “demonstrator” is on the order of 1 M€. The review committee has recommended that its construction be carried through with the highest priority. This includes the final beam dynamic layout of the linac, comprising start to end simulations considering the injection parameters of the present HLI, loss management and error studies. In addition, the technical layout of the machine, taking into account cryostat design, focusing elements, beam diagnostics, rf-design of cavities and power transmitters are to be completed. The short-term goal is to have a power tested fully functional low-energy demonstrator ready by the end of 2011. At the beginning of 2012, the latter can be installed at the HLI at GSI and beam commissioning can start with the acceleration of light ions, followed by cw beam tests with heavy ions at maximum beam intensity. If successful, funding of the full cw linac for SHE research will be negotiated with the HGF.

References

[1] H. Stöcker, G. Krausch, F. Maas, Proposal for the Establishment of a Helmholtz Institute, 02.04.**2009**.

[2] J. Ketelaer et al., *Nucl. Instr. Meth.* **2008**, *A594,* 162-177.

[3] R.-D. Herzberg, P.T. Greenlees, *Progr. Part. Nucl. Phys.* **2008**, *61*, 674-720.

[4] M. Block et al., GSI Annual Report **2008**.

[5] K. Eberhardt, J.V. Kratz, Contribution to the NuSTAR Forschungsschwerpunkt of the BMBF, **2008**.

[6] H. Hummrich et al., *Radiochim. Acta* **2008,** *96,* 73-83.

[7] J.P. Omtvedt et al., *J. Nucl. Radiochem. Sci.* **2002**, *3,*121-124

[8] Ch.E. Düllmann et al., *Radiochim. Acta* **2009**, in press.