

## **GSI Annual Report 2013 (extracts)**

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# Study of the $^{48}\text{Ca} + ^{249}\text{Bk}$ fusion reaction leading to element $Z = 117$ : long-lived $\alpha$ -decaying $^{270}\text{Db}$ and discovery of $^{266}\text{Lr}$ \*

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The fusion-evaporation reaction  $^{48}\text{Ca} + ^{249}\text{Bk}$ , was studied at the gas-filled TransActinide Separator and Chemistry Apparatus (TASCA) [1], which was significantly upgraded [2] now being able to register nuclei with half-lives from sub- $\mu\text{s}$  to a few days. We observed four decay chains among them two long ones comprising seven  $\alpha$  decays and a spontaneous fission, both chains have similar properties (Fig. 1). Our data is largely consistent with previously reported data [3] on the decay chains assigned to  $^{294}117$ . In addition to data from [3], a hitherto unknown  $\alpha$  branch in  $^{270}\text{Db}$ , which populated the new isotope  $^{266}\text{Lr}$ , was identified.  $^{270}\text{Db}$  with a half-life of  $1.0_{-0.4}^{+1.9}$  h is the most long-lived  $\alpha$ -decaying nucleus above No ( $Z = 102$ ). The decay chain members from  $^{290}115$  to  $^{266}\text{Lr}$  all decay with  $T_{1/2} \gtrsim 1$  s, which opens prospects for their chemical investigation and off-line studies.

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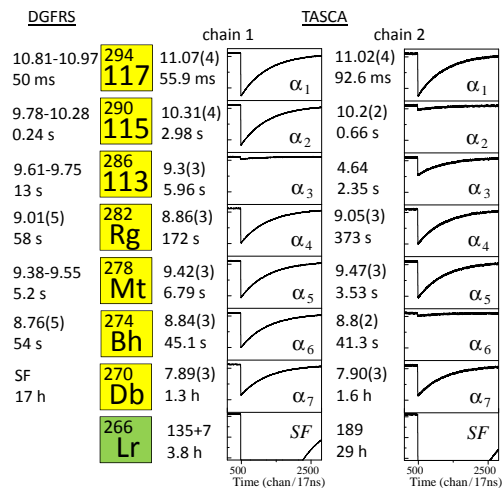


Figure 1: Decay chains assigned to  $^{294}117$  from this work (together with traces of members) and data from [3].

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## Element 115 studied with TASISpec

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An experiment was conducted at GSI to fingerprint the proton number of one or several isotopes along anticipated decay chains of element 115 by means of high-resolution coincidence spectroscopy of  $\alpha$  decays and photons. The fusion-evaporation reaction  $^{48}\text{Ca}+^{243}\text{Am}$  [1] was used. The residues were separated from primary beam and background by TASCA [2-4] and guided into the TASISpec set-up [cf. Fig. 1(a)] [5, 6].

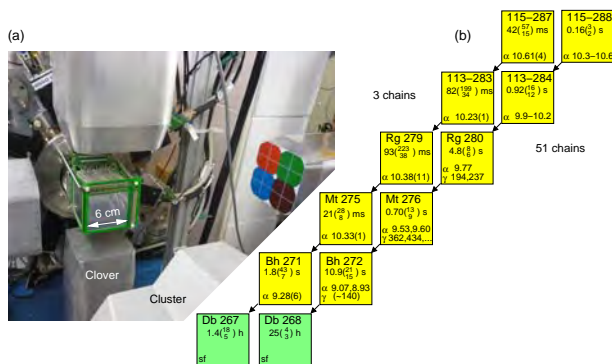


Figure 1: (a) Photograph of the TASISpec  $\alpha$ -photon coincidence set-up [5] in the focal plane of the TASCA gas-filled separator at GSI [2]. See text for details. (b) Proposed decay chains of  $^{287,288}115$  based on the combined data and assignments of Refs. [7-9].

22 and 1 correlated decay chains were found to be consistent with 31 and 2 previously reported chains associated with  $^{288}115$  and  $^{287}115$ , respectively [cf. Fig. 1(b)] [7, 8]. 16 prompt  $\alpha$ -photon coincidences were recorded along the  $^{288}115$  chain [9-11]. Seven short chains of types recoil- $\alpha$  ( $\alpha$ )-fission deserve specific attention [12].

The conclusion of Ref. [9] notes that “thirty correlated  $\alpha$ -decay chains were observed following the reaction  $^{48}\text{Ca}+^{243}\text{Am}$ . Decay schemes arising from high-resolution spectroscopic coincidence data, in conjunction with comprehensive Monte-Carlo simulations, open the

door for direct nuclear structure insights of these heaviest man-made atomic nuclei. Previous assignments linking the majority of the decay chains to the decay of  $^{287,288}115$  [7, 8] are confirmed. This includes first candidates for  $Z$ -fingerprinting the decay of Mt by means of characteristic  $K$ - $X$  ray detection. There is clearly potential for direct determination of the atomic number of the descendants of superheavy elements.”

Following press releases the topic received significant media attention [13], not least due to precisely element 115 starring in various computer games – and Area 51 [14].

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<http://www.youtube.com/watch?v=YW4zZo3F2ac>

# Electronic Structure, Properties and Volatility of Chlorides and Oxychlorides of Group-4 Elements Zr, Hf, and Element 104, Rf

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Volatility of halides, oxyhalides and oxides of elements at the beginning of the 6d-series, i.e., of Rf through Hs has been studied using gas-phase separation techniques [1]. While the understanding of nature of molecule-surface interactions has been gained in group 6 through 8, such an understanding in group 4 and 5 has never been achieved. Neither have the experimentally observed trends in volatility of the halides,  $Zr < Hf > Rf$  and  $Nb < Ta > Db$ , been explained.

Recently, we have studied volatility of the group-5 bromides,  $MBr_5$  ( $M = Nb, Ta, \text{ and } Db$ ), on the basis of calculated molecular properties and a model of dispersion interaction [2]. We have shown that the properties of these compounds change smoothly in the group, so that the volatility should also change smoothly. In this work, we undertake a similar study for the group-4 chlorides. With this aim in view, we have calculated the electronic structures and properties of  $MCl_4$  ( $M = Zr, Hf, \text{ and } Rf$ ).

The calculations were performed with the use of the 2-component relativistic X2C DFT method implemented in the DIRAC12 program package [3]. The uncontracted triple-zeta relativistic basis sets of Dyall were used [4]. The obtained results, for the B88/P86 exchange-correlation potential (Table 1), show an increase in the bond lengths and a decrease in the ionizations potentials in the group, though the polarizabilities show a “zig-zag” behaviour. The latter are much reduced by relativity, with the effect increasing with  $Z$ . Using these properties and a model of dispersion interaction [5], adsorption enthalpies,  $-\Delta H_{\text{ads}}$ , of the chlorides on a quartz surface were determined (Table 1).

Table 1: Equilibrium bond lengths,  $R_e$  (in Å), dissociation energies,  $D_e$  (in eV, from Ref. [6]), ionization potentials, IP (in eV), polarizabilities,  $\alpha$  (in a.u.), and adsorption enthalpies,  $\Delta H_{\text{ads}}$  (in kJ/mol), of  $MCl_4$  ( $M = Zr, Hf, \text{ and } Rf$ ) on quartz

	$R_e$	$D_e$	IP	$\alpha$	$\Delta H_{\text{ads}}$
ZrCl <sub>4</sub>	2.336	20.34	11.00	103.6	106.5
HfCl <sub>4</sub>	2.316	20.80	11.00	99.3	103.0
RfCl <sub>4</sub>	2.370	19.40	10.96	101.2	102.7

The obtained  $-\Delta H_{\text{ads}}$  are indicative of the following trend in the adsorption strength:  $Zr > Hf > Rf$ . Using a correlation of  $-\Delta H_{\text{ads}}$  with the sublimation enthalpies,  $\Delta H_{\text{sub}}$  of 104.2 kJ/mol was predicted for RfCl<sub>4</sub>. Thus, according to the calculations, RfCl<sub>4</sub> should be the most volatile compound in its group, in agreement with previous

Table 2: Equilibrium bond lengths,  $R_e$  (in Å), dissociation energies,  $D_e$  (in eV), ionization potentials, IP (in eV), and dipole moments,  $\mu$  (in D), of  $MOCl_2$  ( $M = Zr, Hf, \text{ and } Rf$ )

	$R_e(M-Cl)$	$R_e(M=O)$	$D_e$	IP	$\mu$
ZrOCl <sub>2</sub>	2.386	1.741	21.76	10.76	3.67
HfOCl <sub>2</sub>	2.352	1.753	21.01	10.63	4.33
RfOCl <sub>2</sub>	2.385	1.822	19.40	10.35	5.20

predictions [7]. The obtained trend is, however, in contradiction with the experimental observations [1].

To check the possible (at the presence of oxygen in the chromatography column) formation of the oxychlorides, as well as their volatility, we have also performed the electronic structure calculations for  $MOCl_2$  ( $M = Zr, Hf, \text{ and } Rf$ ). The obtained total energy differences between  $MOCl_2$  and  $MCl_4$  are indicative of the following trend in the oxychloride formation:  $Zr > Hf > Rf$ . This means that the assumed earlier preference of Hf to form the oxychloride should not take place. The other properties of interest are summarized in Table 2. The  $MOCl_2$  are shown to be stable as a bent structure, though the difference in the total energy with respect to the flat one is very small. The Rf compound should be less stable than those of Zr and Hf, similarly to  $MCl_4$ . The central atom - ligand separation increases from Hf to Rf, which also leads to an increase in the dipole moments. Such an increase in  $\mu$  should result in a decrease in the volatility of  $MOCl_2$  with  $Z$  as adsorption on a quartz surface due an increase in the dipole moment - surface charge interactions. Thus, volatility of the oxychlorides should change as  $Zr < Hf < Rf$ . This trend is, however, also in disagreement with the experimental observations [1]. Thus, the experimentally determined reversed trend in group 4 cannot find its theoretical explanation; neither can the one in group 5 be interpreted.

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# Relativistic coupled cluster study of the MAu and M<sub>2</sub> dimers of Hg, Cn, and Fl

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The electronic structure of Cn (Z=112) and Fl (Z=114) experiences strong relativistic effects due to the relativistic stabilization and contraction of their valence orbitals, the 7s orbital in case of Cn, and the 7p<sub>1/2</sub> orbital in case of Fl. Thus, their molecular and chemical properties are expected to be determined by relativity.

These two elements are the heaviest atoms studied in chemical experiments. Their relative inertness makes them good candidates for studies of volatility through their adsorption on metal surfaces (such as gold) using the gas-phase thermochromatography technique [1, 2]. By performing calculations of the binding energy of the MAu dimers, the trends in the adsorption enthalpies of the atoms on a gold surface,  $-\Delta H_{\text{ads}}^{\text{Au}}(M)$ , can be estimated. Bonding of an element in the solid state in the first approximation can be described by the bonding in its M<sub>2</sub> dimer. Thus, in order to obtain a theoretical insight into the trends in the binding of Hg, Cn, and Fl to a gold surface, and their binding in a solid, *ab initio* study of the molecular properties of their MAu and M<sub>2</sub> dimers was performed.

The calculations were carried out within the infinite-order two-component relativistic Hamiltonian obtained after the Barysz-Sadlej-Snijders (BSS) transformation of the Dirac Hamiltonian in a finite basis set [3]. This method treats scalar and spin-orbit relativistic effects simultaneously, and for valence molecular properties achieves similar accuracy to that of the 4-component Dirac Hamiltonian, while significantly reducing the computational effort. Electron correlation was treated in the framework of the single reference coupled cluster approach with single, double, and perturbative triple excitations (CCSD(T)). Faegri's dual family basis sets [4] were used; the basis sets were extended to convergence of the calculated molecular properties. All the calculations were performed using the DIRAC08 computational program package [5].

Table I contains the calculated bond lengths,  $R_e$ , and dissociation energies,  $D_e$ , of the MAu and M<sub>2</sub> molecules. Hg<sub>2</sub> is the only system for which experimental parameters are available [6]. The excellent agreement of the calculated binding energy with the experiment in case of Hg<sub>2</sub> lends credence to our predictions for the rest of the systems.

The calculated trend in the bond length of the MAu molecules is  $R_e(\text{FlAu}) > R_e(\text{CnAu}) > R_e(\text{HgAu})$ . The increase in  $R_e$  from HgAu to CnAu corresponds to the weakening of the bond in CnAu, and the  $R_e$  of FlAu is larger than that of CnAu due to the participation of the more diffuse 7p<sub>1/2</sub> and 7p<sub>3/2</sub> orbitals of Fl in the bond [7]. The present calculations predict the HgAu molecule to be the most stable of the three, followed by FlAu, and CnAu as

Table 1: Equilibrium bond lengths,  $R_e$  (in Å), and dissociation energies,  $D_e$  (in eV), of the MAu and M<sub>2</sub> molecules. The values in parenthesis are experimental for Hg<sub>2</sub> [6].

	HgAu	CnAu	FlAu
$R_e$	2.640	2.720	2.791
$D_e$	0.563	0.368	0.446
	Hg <sub>2</sub>	Cn <sub>2</sub>	Fl <sub>2</sub>
$R_e$	3.744 (3.69)	3.461	3.547
$D_e$	0.050 (0.046)	0.084	0.117

the least stable. Bonding in CnAu is weaker than in HgAu due to the strong relativistic stabilization of the 7s orbital in Cn compared to the 6s in Hg. FlAu should be stronger bound than CnAu due to the 7p<sub>1/2</sub> orbital in Fl being more accessible than the 7s of Cn for the bonding with the 6s orbital of gold [7]. Assuming that the trends in the binding energies of the gold containing diatomic molecules are a good indication of the trends in the adsorption enthalpies of the atoms on a gold surface, according to the current calculations Fl should be stronger adsorbed on gold than Cn.

Comparing the results for the three M<sub>2</sub> dimers, we observe that Hg<sub>2</sub> has the longest and Cn<sub>2</sub> the shortest bond. The trend in the dissociation energy is  $D_e(\text{Fl}_2) > D_e(\text{Cn}_2) > D_e(\text{Hg}_2)$ . Cn<sub>2</sub> and Hg<sub>2</sub> dimers behave as van der Waals systems; thus Cn<sub>2</sub> has a higher dissociation energy of the two, due to the strong relativistic contraction of the 7s orbital, which causes a decrease of  $R_{\text{vdW}}(\text{Cn})$  and a subsequent increase in the binding energy of the dimer. Group-14 elements, including Fl, behave like chemically bound systems, where  $D_e$  decreases with Z due to the spin-orbit splitting and the relativistic stabilization of the np<sub>1/2</sub> orbital. Thus, Fl<sub>2</sub> is the least bound dimer in group 14 [7]; however, due to the different type of interaction, it is stronger bound than both Hg<sub>2</sub> and Cn<sub>2</sub>.

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## Upgrade preparation for the 1.4 MeV/u gas stripper system for FAIR

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A key projectile for the FAIR facility will be  $^{238}\text{U}$ . In routine operation of the GSI UNILAC,  $^{238}\text{U}$  is generated by a MEVVA ion source that delivers ions with comparably low charge states ( $4^+$ ), which are accelerated to 1.4 MeV/u in the high current injector (HSI). The 1.4 MeV/u beam passes a region of high gas density, in which the charge is increased to  $28^+$  by stripping of electrons. [1] Generally, higher intensities at charge states, preferably above  $28^+$ , are desirable. This would allow to operate the accelerator more reliable and efficiently.

To optimize the stripping efficiency and potentially increase the achieved ion charge states a program to upgrade the gas stripper has started. The modified stripper setup is depicted in Fig. 1. As a first modification, switching from the continuously fed supersonic  $\text{N}_2$ -jet to a pulsed gas injection, synchronized with the beam timing structure, has been implemented. This allows to increase the gas pressure inside the stripper chamber during a beam pulse, while still reducing the total gas flow. The gas injection was positioned inside a T-fitting, which was installed in the main stripper chamber to match the beamline. In a first test the pressure in the main stripping section as function of the opening time of the valve and the pressure along the beamline adjacent to the gas stripper section were measured.

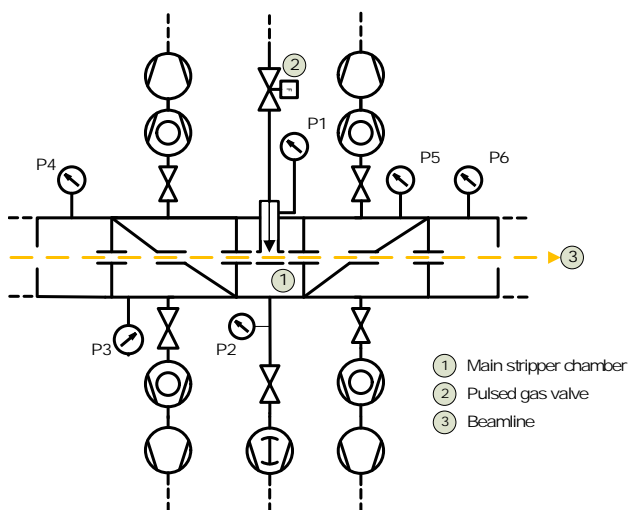


Figure 1: Schematic view of the modified 1.4 MeV/u gas stripper as to be used for first measurements with a pulsed gas valve in beam experiments in 2014.

The gas pressure on the valve was 3 MPa. The dependency of the pressure inside the stripper chamber and the pumping performance on the valve opening time was evaluated

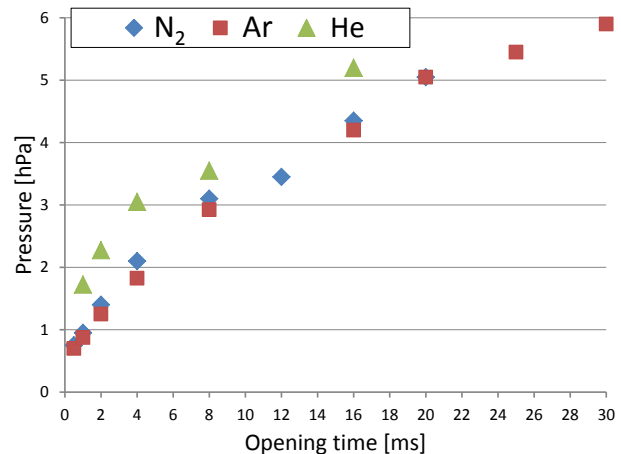


Figure 2: Pressure measurements at the gas stripper for different stripper gases as function of the opening time of the pulsed gas valve.

for three different gases ( $\text{N}_2$ , Ar and He). These gases are planned to be investigated as potential stripper gases in the future, together with Xe.

The results of the pressure measurements near the gas outlet in the main stripper chamber (diaphragm vacuum gauge at P1, Fig. 1) are shown in Fig. 2. As the pressure varied according to the pulsed gas flow regime, only the maximum pressure values during one pulse are shown. Note that the vacuum gauge was placed at an entry point on top of the main stripper chamber, so the shown pressures do not represent the real pressure in the beamline. The pressure increases with increasing opening time and starts to level off at longer opening times.  $\text{N}_2$  and Ar are pumped at about the same rate whereas He is pumped less efficiently. Therefore, the pressure at the vacuum gauge is higher for He at the same opening times. The measured pressure increases to values above 5 hPa, independent of which gas was used. The pressure in the adjacent beamline was also measured (vacuum gauges P2-P6, Fig. 1).

The optimum conditions with respect to the pressure in the main stripping region and the adjacent beamline as well as the experimental charge state distribution will be evaluated in beam experiments in 2014.

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