GSI Annual Report 2015 (extracts)

Contributions SHE Chemistry Group

Contributions with Participation of SHE Chemistry Group Members

Predictions of Adsorption of Cn, Fl and their homologs Hg and Pb, respectively, on a Hydroxylated Quartz Surface

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Both experimental and theoretical research in the area of chemistry of the superheavy elements (SHE) have recently been focusing on investigations of properties of elements 112 (Cn) and 114 (Fl): their unusually high volatility was expected due to the strongest relativistic effects on their electron shells and a closed-shell, $6d^{10}7s^2$, and a quasi-closed shell, $7s^27p_{1/2}{}^2$, electronic ground states, respectively [1]. First experimental studies of their volatility as adsorption on a gold surface of detectors of a chromatography column have indeed shown that these elements are very volatile [2-4]. They, however, disagree on the trend in volatility between Cn and Fl: Cn < Fl [3] while Cn > Fl [4]. Further experiments are under way to solve this contradiction and to differentiate between these elements and their lighter homologs Hg and Pb [5]. This aim should be achieved with the use of two types of detector surfaces – quartz and gold – presumably giving very different adsorption enthalpies, ΔH_{ads}(M).

Earlier, we have predicted ΔH_{ads} of these elements on gold via 4c-DFT cluster calculations [6] and on quartz using a van der Waals (vdW) adatom-slab model and atomic DCB CC calculations [7]. In this work, we predict ΔH_{ads} of Hg, Cn, Pb, and Fl on a quartz surface at ~ 21 °C using the periodic ADF BAND code [8].

Two types of the hydroxylated α-quartz (001) surface were considered; vicinal and germinal silanols. Various adsorbate coverage - from the full till a very low one - was modeled by the (1x1), (2x2) and (4x4) supercells. Also, various adsorption positions were tested. Fig. 1 shows, e.g., adsorption of adatom M on the (2x2) supercell simulating the vicinal silanol (80% at 21 °C).

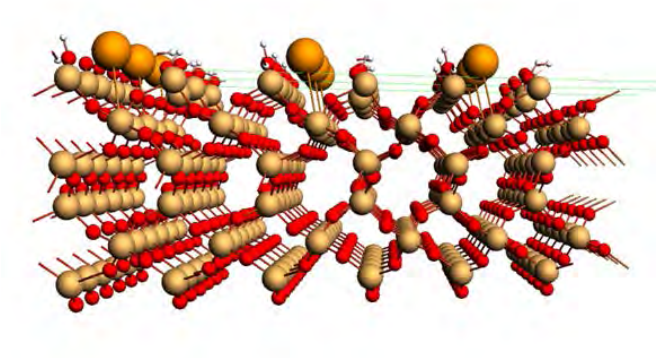

Fig. 1. Atom M (M = Hg, Cn, Pb, and Fl) adsorbed on a hydroxylated quartz surface, a (2x2) supercell (a side view).

Results of the calculations have shown that adsorption on vicinal silanols is stronger than on germinal, and that the preferred adsorption position of all the adatoms is a hollow one, on top of the Si atoms in the second row from the top (Fig. 1).

Table 1 summarizes the calculated ΔH_{ads} of Hg, Cn, Pb, and Fl on the hydroxylated (001) α-quartz surface for the (4x4) supercell, as well as our earlier calculations of ΔH_{ads} of these elements on quartz using a vdW model [6], also in comparison with experimental data. One can see that the ADF BAND data agree pretty well with experiment for Hg and Pb at zero coverage, which gives credit to the results for Cn and Fl: these elements should not adsorb on quartz by chemical forces. However, the vdW adsorption should give positive ΔH_{ads} values [7]. A high inertness of Cn and Fl towards quartz is a result of strong relativistic effects.

Thus, according to the present calculations and those of [6,7], the following behavior can be predicted for the coming experiments: Pb should adsorb on quartz at 21 °C; Hg should not adsorb on quartz and should adsorb on gold at 21 °C; Cn should not adsorb on quartz and should adsorb on gold at $T_{ads} \approx$ -20 °C, while Fl should not adsorb on quartz at 21 °C and should adsorb on gold at 21 °C showing a smooth distribution along the detectors' area.

Table 1. Predicted $-\Delta H_{ads}$ (kJ/mol) of Hg, Cn, Pb, and Fl on a hydroxylated α-quartz (001) surface and experimental data at zero coverage

Method	Hg	Cn	Pb	Fl	Ref.
ADF	54	-38	152	-22	This
vdW[a]	(26.3)	26.0	(27.3)	21.0	7
Exp.	42 ± 2	-	165 ± 4	-	9

[a] a vdW model plus atomic DCB CC calculations.

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Improvements for superheavy element chemistry behind TASCA

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Experiments on the chemical properties of flerovium (Z=114, Fl), produced in the reaction ^{244}Pu(^{48}Ca, 3-4n)^{288,289}Fl, were performed at GSI, Darmstadt using the COMPACT gas-chromatography setup coupled to the gas-filled recoil separator TASCA [1]. The first experimental investigations of Fl at GSI were performed in 2009 and two radioactive decays of Fl were observed with negligible background [1]. The two most long-lived known Fl isotopes are ^{289}Fl ($T_{1/2}$ =$2.1^{+0.8}_{-0.4}$ s) and ^{288}Fl ($T_{1/2}$ = $0.69^{+0.17}_{-0.11}$ s) [2]. Due to their relatively short half-lives a fast transport time from TASCA to COMPACT is crucial. The transport time is directly linked to the volume of the Recoil Transfer Chamber (RTC) [3], which in turn is attached at the TASCA the focal plane. During the experiment reported in [1], TASCA was operated in its Small Image Mode (SIM) [4]. Relative to the High Transmission Mode (HTM), the SIM has a smaller image size at the focal plane at the cost of a lower transmission.

For experimental investigations on Fl, performed in 2014/15 several improvements to the experimental set-up have been performed, to optimize the overall efficiency. Initially this was studied with short-lived Hg and Pb isotopes:

1) Optimizations of i) the quadrupole magnets focusing in HTM and ii) window size of the RTC resulted in a higher overall efficiency.

2) Several RTCs were developed and tested to identify the minimum transport time. For Fl studies, a fully Teflon®-covered RTC and direct connection to COMPACT appears to be the best suited. It suppresses encounters of Fl with any reactive surfaces prior to those in COMPACT. SRIM simulations showed a gas mixture of 50% He and 50% Ar and an RTC depth of 20 mm to ensure complete stopping of Fl isotopes inside the RTC. In addition, the transport time as function of the gas flow rate was optimized, leading to faster flush-out and hence an improved overall efficiency. Online experiments with ^{183}Hg ($T_{1/2}$ = 9.4 s) produced in the ^{48}Ca+^{142}Nd reaction showed that 62% of all Hg entering the RTC were deposited in COMPACT. The amount of Hg entering the RTC was measured in a separate experiment by placing a Si focal plane detector, behind the RTC window in the RTC position. Using a chopped beam (0.1 s beam on and 5 s beam off), a flush-out profile for Hg was measured. Based on the Hg decay-in-flight observation in a SiO_2-covered COMPACT array, 50% of Hg entering the RTC reached COMPACT within 0.4 s, at a gas flow rate of 2 L/min.

3) Different combinations of COMPACT detectors with either a Au or SiO_2 surface, can be used in an isothermal or thermochromatography mode. This broadens the range of experimentally accessible chemical reactivity. Preparatory experiments have shown, that Pb deposits on SiO_2, whereas Hg deposits only on Au [5]. To distinguish between Pb- and Hg-like behavior, a first section of the COMPACT detector is covered with a SiO_2 surface (cf. Fig. 1).

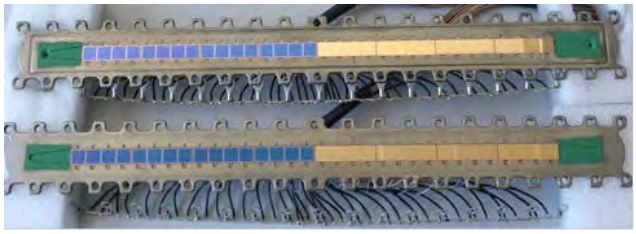

Fig. 1: COMPACT detector array with SiO_2 and Au surfaces.

Experiments on Fl using this improved system have been performed and are currently under analysis. The setup is also suitable for chemical studies of element 113, where the isotope ^{284}113 ($T_{1/2}$ = $0.97^{+0.12}_{-0.10}$ s) [6, 7, 8] can be produced as daughter after α-decay of ^{288}115 in the reaction ^{243}Am(^{48}Ca,3n)^{288}115.

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Radiochemical investigation of the kinematics of multi-nucleon transfer reactions

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Multi-nucleon transfer reactions have attracted renewed attention as a promising tool for the production of neutron-rich transactinide isotopes [1-3]. Based on this renewed interest we performed a ^{48}Ca + ^{248}Cm bombardment at an incident energy 10% above the Coulomb barrier at TASCA to investigate angular-resolved distributions and recoil energies of isotopes between Cf and Fm. Identification of isotopes of these elements was performed within the offline radiochemical separation technique. Thus, only long-lived isotopes with atomic mass numbers between 246 and 256 were accessible. For the analysis of the reaction kinematics, the estimation of the most probable number of evaporated neutrons and the recoil energies as function of the angular distribution was evaluated for each nuclide. We used the stacked catcher foil technique to recieve information about the penetration by using several few μm thin Ni-foils, which were arranged behind one another. Each foil was divided into four arc-segments thus collecting reaction products emitted at different angles. This allowed to simultaneously measuring both, recoil ranges and angular distributions.

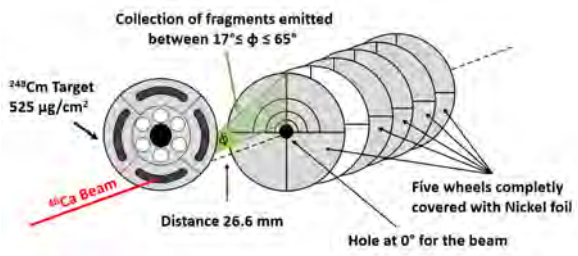

Figure 1: Schematic of the experimental setup: Five frames were completely covered with Ni-catcher foils of different thicknesses (1x3 μm, 3x1 μm and 1x5 μm). The first wheel was designed to stop sputtered ^{248}Cm target material. On the second wheel, only one quadrant was covered with a 5μm nickel foil to catch all transfer products emitted into this quadrant to provide a 100 % reference.

Irradiation of ^{248}Cm with ^{48}Ca ions was performed at the TASCA target position at a beam energy of 5.78 MeV/u, which corresponds to a center of mass energy of 220 MeV in the center of target. The target was irradiated for 10 hours with an average beam intensity of 4.6 x 10^{12} particles/s. The transfer products were stopped in the Ni catcher foils covering laboratory angles from ϕ = 17° - 65°. After end of bombardment, the foils were dissolved in dilute nitric acid, and a scavenger precipitation with $Fe(OH)_3$ using ammonia was performed. Different elements were separated on a cation-exchange column with 0.12 M α-HiB and 0.14 M α-HiB at pH=4.80 into a combined Fm/Es fraction and a Cf/Bk fraction, respectively. This allowed seperating nuclides with adjacent atomic numbers by subsequent column chromatographic treatment. The identification of Bk-, Cf-, Es-, and Fm-isotopes was achieved by detecting α-particles, considering known cross-section ratios for different isotopes of a given element [4], and their characteristic decay properties as well as precursor effects.

Kinetic energies corresponding to recoil range in the stopping material were evaluated using the stopping power systematics of ions in matter [5]. The data will be investigated within Volkov's generalized Q_{gg} systematics including corrections for the breaking of nucleon pairs in the multi-nucleon transfer process [3,6]. Information of the angular distributions, and the total kinetic energy loss (TKEL) can be used to generate deflection functions for the visualisation of the rotational movement through the nucleon-transfer process.

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Progress on the 1.4 MeV/u pulsed gas stripper for the GSI UNILAC

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The GSI Universal Linear Accelerator (UNILAC) will serve as part of the injector system for the Facility for Antiproton and Ion Research (FAIR) [1]. An UNILAC upgrade program has already started dedicated to meet the high demands of FAIR [2]. As part of this program a pulsed gas stripper has been developed, to replace the 1.4 MeV/u N_2 gas-jet stripper.

The gas stripper device is equiped with a pulsed gas injection, synchronized with the beam pulse timing, to increase the effective gas density during a beam pulse transit [3]. The aim is to enhance the beam intensity behind the stripper by increasing the fraction of ions with the desired charge state. In past measurement series, H_2 was identified as most promising stripper gas to achieve this goal [4]. Initial measurements using U-beams on the pulsed H_2-target showed an increased stripping efficiency into the desired 28^+ charge state of about 60 %. At back-pressures up to 12 MPa, increasing charge states were observed [5]. The behavior of the charge state distribution suggested even higher charge states at further increased gas densities. This is depicted in Fig. 1 by the charge state distributions of U-ions after passing the H_2-target at increasing target thicknesses. The target thickness is estimated from energy loss measurements using SRIM2013 [6].

In 2015, the whole setup was improved enabling for higher gas densities. A new generation of pulsed gas valves was used, which can tolerate an increased back-pressure up to 30 MPa. This improvement allows for target thicknesses of the H_2-target of about $\leq 70\,\mu g/cm^2$. Additionally, a new specialized power supply was commissioned, improving the reliability and control of the gas valves. The required opening time to reach a maximum gas density is about $\leq 250\,\mu s$, depending on back-pressure and gas type. The gas supply for the valves was modified to deliver H_2 with the desired back-pressure.

The flange, equiped with the new injection system, is mounted on top of the main stripper chamber. A four-stage differential pumping system provides for the required vacuum pressures ($\leq 10^{-3}$ Pa) in the adjacent beam line towards the High Current Injector (HSI) and the Alvarez section. A roots vacuum pump, installed on the bottom of the main stripper chamber, as well as several turbo pumps are used (see [4]).

Systematic measurements of charge fractions, beam emittance and energy-loss were conducted using ^{238}U-, ^{209}Bi-, ^{50}Ti- and ^{40}Ar-beams on H_2- and He-targets, as well as N_2 as a reference stripper gas. The experimental data are still under evaluation.

Additionally, measurements of the gas flow through the stripper cell and the gas pressure near the interaction zone were conducted. The gas flow was measured to 1.0 ± 0.5 L/min using the H_2-target at 8 MPa back-pressure for operation with U^{28+}-beams (100 μs, 1 Hz repetition rate). This is a significantly reduced gas consumption compared to the N_2 gas-jet stripper operation (22 L/min).

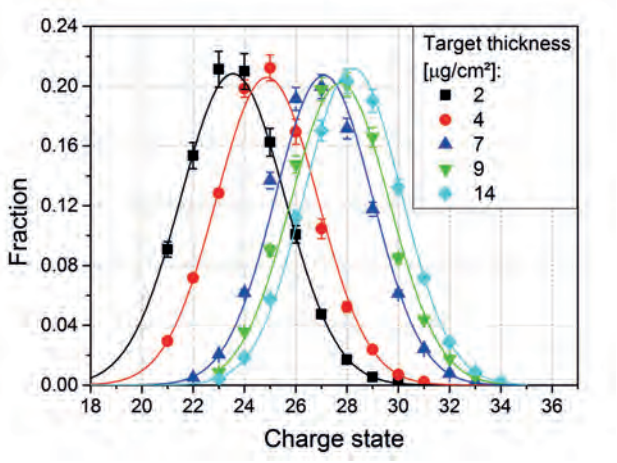

Figure 1: Charge state distributions of U-ions passing through the H_2-target at increasing target thicknesses.

First measurements in mixed beam operation as well as in long-pulsed beam operation were performed. Mixed beam operation was succesfully tested by stripping U- and Ti-beams in H_2- and N_2-targets, respectively. In long-pulsed beam operation with high repetition rates (up to 5 ms, 50 Hz), the use of H_2 and He at increased back-pressures is limited by the pumping system. However for this operation mode, the performance of the pulsed gas cell with N_2 remains the same as with the N_2-jet stripper.

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Decay spectroscopy at SHIP using a new focal plane detection system

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Nuclear structure experimental studies in the heavy and superheavy element regime at the extremes of sensitivity require continuous advancements in detector development. A focal plane detection system was designed and developed in-house at GSI, Darmstadt, to explore this region in decay spectroscopy studies [1]. Initially, a study was performed on the superheavy nucleus ^{257}Db with the LISE filter at GANIL, France, with the data currently under analysis. To assess the performance and capabilities of the apparatus in combination with the SHIP separator, an experimental campaign over a variety of heavy nuclei was completed in October 2015 at GSI.

The measurement apparatus comprises a double sided silicon strip detector (DSSD) of 60x60 strips (1 mm strip pitch, 300 μm thickness) for the detection of recoil implantations and subsequent α/e^-/spontaneous fission decays surrounded in a box formation by four single sided silicon strip detectors (SSSD) upstream, to measure α-decay escapes and conversion electron emission. The detection arrangement had one Ge clover detector (behind the DSSD and separated by a thin 1.5 mm Al cap) for γ-ray/X-ray decay measurements (up-to 5 Ge detectors are possible). The electronics readout system used was charge sensitive preamplifiers coupled to the FEBEX3A/B digitisers [2] developed at GSI and previously employed by the TASCA group [3].

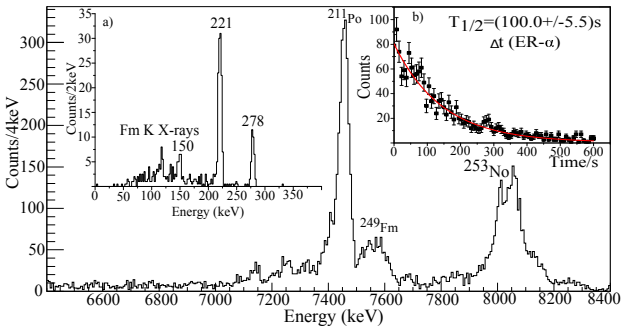

Figure 1: Alpha-decay of ^{253}No measured in the DSSD. Inset a) Gamma rays in coincidence with the α-decay of ^{253}No, inset b) time difference between evaporation residues (ER) implantation and subsequent α decay.

The primary beam of $^{48}Ca^{10+}$ in parasitic mode (5 Hz macropulse, 5 ms wide) was impinged on a variety of targets. Initially, the previously measured [4] heavy odd-A actinide ^{253}No was produced in the reaction ^{207}Pb(^{48}Ca,2n)^{253}No. In coincidence with the α-decays from ^{253}No, γ-rays from excited states in ^{249}Fm were measured. In this deformed mid-shell region of the nuclear chart, ^{254}Lr and ^{255}No were also produced. The data analysis is ongoing.

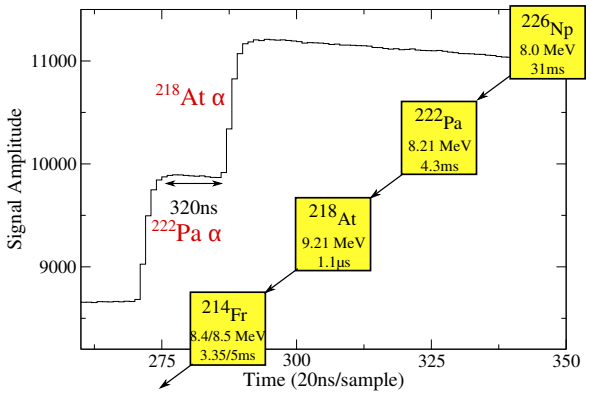

Figure 2: ^{226}Np decay chain measured in the reaction ^{181}Ta(^{48}Ca,3n)^{226}Np. The relatively fast decay of ^{218}At can be measured through examination of individual traces correlated to the ms decay of ^{226}Np and ^{214}Fr.

In order to study the evolution of the N=126 shell closure above protactinium, which is not yet well understood [5], neutron deficient isotopes in the uranium-plutonium region were studied in addition. To that end, the reaction ^{48}Ca + ^{181}Ta$\rightarrow^{229}$Np* was selected with a two-fold purpose of populating neutron deficient neptunium isotopes to improve on data in this region, while at the same time testing the performance of the electronics in a region with known relatively fast decays. An example of a decay chain correlated to ^{226}Np is shown in Fig 2. The fast decay of ^{218}At measured demonstrates the capability of detecting events in the sub-microsecond range with an almost dead time free system. In conclusion, the new SHIP focal plane detection system has been successfully tested at GSI, with a broad collection of data taken.

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Laser spectroscopy of nobelium

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Relativistic effects influence the valence electron configuration of the heaviest elements affecting their physical and chemical properties. These effects can be described using state-of-the-art numerical approaches for highly correlated, relativistic many-electron systems, such as Multi Configuration Dirac-Fock and Relativistic Coupled-Cluster calculations [1]. To benchmark these calculations, a comparison between measured and predicted atomic properties is needed. At present, no laser spectroscopy data is available for the transfermium elements (Z>100). Thus, laser spectroscopic studies of these elements are of great interest.

In our experiments we aim to search for atomic levels in the element nobelium (Z=102) applying the Radiation Detected Resonance Ionization Spectroscopy (RADRIS) [2] technique. For such studies the isotope ^{254}No, $t_{1/2}$ = 51s was chosen. It was produced via the fusion evaporation reaction ^{208}Pb(^{48}Ca,2n)^{254}No using the ^{48}Ca ion beam provided by the UNILAC. The fusion-products were separated by the velocity filter SHIP and about four ^{254}No ions/s entered a buffer gas cell where they were stopped in 95 mbar argon gas of 99.9999% purity and were subsequently collected on a tantalum filament. After an appropriate collection time, the primary beam was blocked and the accumulated ions were re-evaporated as neutral atoms by a short heating pulse. We then employ a two-step photoionization process to ionize the atoms. In case of resonant excitation and final photoionization, the ions were transported by suitable electric fields to a silicon detector where they were identified by their characteristic α decay.

Laser light for the first excitation-step was provided by four tunable dye lasers and an optical parametric oscillator (OPO) system and transported to the experimental setup using UV-fibers. The dye lasers were pumped by two excimer lasers operated at 248 nm and 308 nm. The OPO, pumped by a frequency-tripled Nd:YAG laser, was operated in a frequency mixing mode, delivering laser light in the wavelength range below 410 nm. The 351 nm light for the second, non-resonant step was provided by a high power excimer laser and was transported to the experimental setup by highly reflective UV-mirrors. The laser wavelengths were measured with a high precision.

Different theoretical models predict the *7s7p* ^{1}P$_1$ atomic level in the range of 322-385 nm [3]. The large spectral range to be covered and the low production rate makes the experiment challenging. After the level search in 2007 and 2014, the setup was further optimized [4]. In summer 2015, a resonance was observed at a wavelength of about 333.8 nm and the resulting α-decay spectrum is shown in Fig. 1. Baking the filament increased the effieciency of the setup increased by about a factor of two, cf. 1 (c). These results will be published soon. In addition, some Rydberg states were identified in the 2015 beamtime. This enables the determination of the first ionization potential of No with high accuracy in future beamtimes.

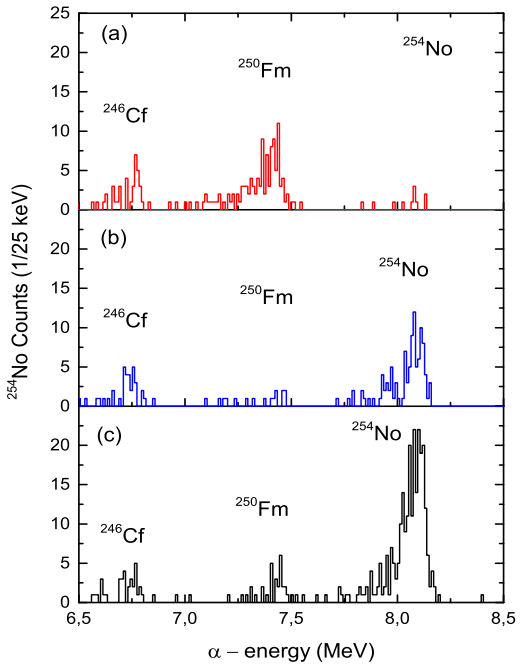

Figure 1: α-decay spectrum of ^{254}No. The measurements with laser off (a), lasers on (b) and after conditioning the filament(c). The laser excitation energy λ_1= 333.8 nm, λ_2= 351 nm with laser pulse energy E_{λ_1} = 200 μJ and E_{λ_2} = 36 mJ. The measurement time was 300 s for (a) and (b) and 600s for (c)

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High current proton and carbon beam operation via stripping of a molecular beam at GSI UNILAC

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The experimental program of the future facility for Antiproton and Ion Research (FAIR) project requires a high number of cooled anti-protons per hour [1]. The FAIR proton injector linac has to deliver a 70 MeV, 35 mA pulsed proton beam at a repetition rate of 4 Hz [2].

During recent machine investigations at the GSI a high current proton beam was achieved in the Universal Lineral Accelerator (UNILAC) [3, 4]. In preparation for this the ion source was equipped with a newly developed 7-hole extraction system and optimized for single charged hydrocarbon beam (isobutane gas) operation. This beam was accelerated to 1.4 MeV/u and cracked in a new pulsed gas stripper into protons and charged carbon. The new stripper setup provides for high density gas pulses synchronous with the transit of the beam pulse close to the beam trajectory. With this setup a proton (up to 4.3 mA) as well a carbon-beam (up to 9.5 mA) intensity record at a beam energy of 1.4 MeV was achieved. The proton beam was accelerated up to 3.6 MeV/u inside the first Alvarez-section with full transmission.

MACHINE DEVELOPMENT

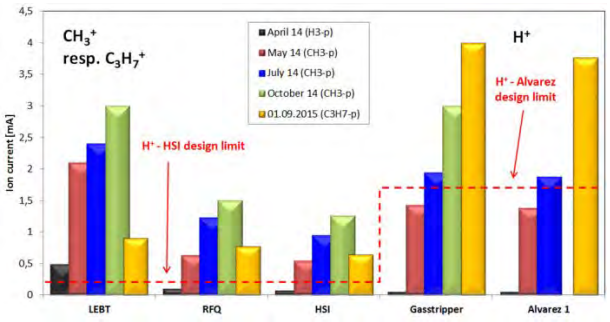

Figure 1: Measured beam current along HSI and Alvarez 1; the proton design limit was exceeded in the post stripper section.

The MUCIS ion source [5] was operated with isobutane (C_4H_{10}) gas, while a high-current $C_4H_7^+$-beam (0.7 emA) was delivered to the HSI. Due to the increased HSI design limitation for the $C_4H_7^+$-beam, an improved beam transmission compared to a pure proton beam is evident. The proton intensity should a theoretical increase by a factor of 7, while a factor of 6.4 was achieved (Fig. 1). The charge state distribution could be changed easily by varying the target density, while the proton beam intensity remains the same (Fig. 2). The proton beam transmission along the GSI-poststripper decreased to 70% (Fig. 3).

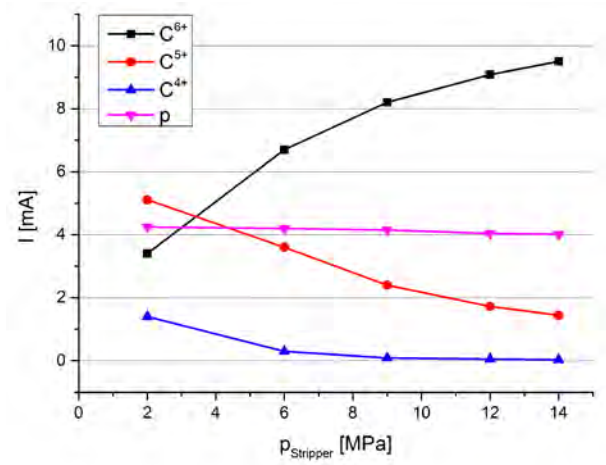

Figure 2: Measured ion current for proton- and carbon-beams (with different charge state) depending on stripper target density; stripping of an isobutane beam.

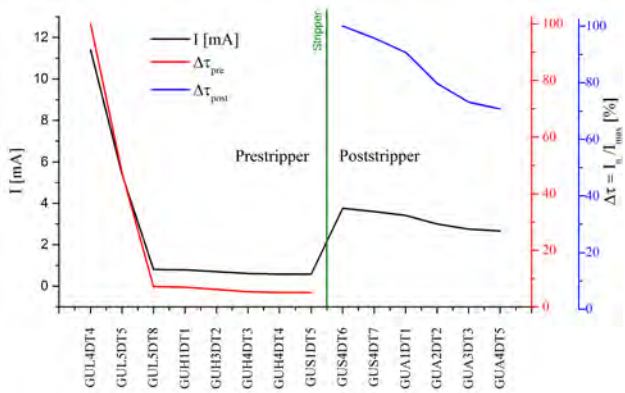

Figure 3: Measured (electrical) beam current of pre- ($C_4H_7^+$) and poststripper transmission (p) along GSI-UNILAC.

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