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# Production and decay of element 114: high cross sections and new nucleus <sup>277</sup>Hs\*

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#### Introduction

Discoveries of new superheavy elements (SHE) were reported from FLNR, Dubna, Russia [1], including observations of element 114 isotopes produced in the <sup>48</sup>Ca+<sup>242,244</sup>Pu reactions. Successful independent studies of some of the reactions studied in Dubna were reported [2,3], most recently also the observation of one atom each of <sup>286,287</sup>114 produced in the <sup>48</sup>Ca+<sup>242</sup>Pu reaction at LBNL [4]. Predictions on the existence of an "island of stability" in the region of SHE have substantiated, despite the small number of observed events in every confirmation experiment. All successful confirmation experiments reported cross sections lower than those from FLNR by factors of two or more. Nevertheless, these cross sections are unexpectedly high compared to extrapolations from lighter systems [5], and intriguingly constant over a large range of 112 < Z < 118. A thorough understanding of the underlying production mechanism is still missing; location and extension of the "island of stability" in the region of spherical SHE is still far from being established. To help shedding more light on these problems, a <sup>48</sup>Ca+<sup>244</sup>Pu experiment was performed at the gas-filled TransActinide Separator and Chemistry Apparatus (TASCA) [6,7], which was optimized for the study of <sup>48</sup>Ca-induced fusion reactions with actinide targets. TASCA's efficiency for this nuclear reaction type is currently unsurpassed.

#### **Experimental**

The UNILAC accelerated a pulsed <sup>48</sup>Ca beam (~ $2 \cdot 10^{12}$  s<sup>-1</sup>), which passed through <sup>244</sup>PuO<sub>2</sub> targets (average thickness: 438 µg/cm<sup>2</sup> <sup>244</sup>Pu). Beam energies inside the targets were 241.3-246.2 MeV (E\*=39.8-43.9 MeV; hereafter referred to as 42-MeV run) and 236.4-241.0 MeV (E\*=36.1-39.5 MeV; 38-MeV run). 2.44 \cdot 10^{18} (42-MeV run) and 1.15 \cdot 10<sup>18</sup> (38-MeV run) projectiles passed through the targets. Nuclear reaction products entered

TASCA, operated in "high transmission mode" [7], and were separated in 0.8 mbar He gas. The detection system consisted of a Multi Wire Proportional Counter (MWPC) and a focal plane detector box (FPDB). The FPDB consisted of a Double Sided Silicon Strip Detector (DSSSD; pitch size: 1 mm; 144 vertical / 48 horizontal strips) and Single Sided Silicon Strip Detectors (SSSSD) mounted perpendicular in the backward hemisphere of the DSSSD [8]. The MWPC provided a signal for ions recoiling from the target and allowed distinguishing these from radioactive decays of species implanted in the DSSSD. The energy resolution of the FPDB was 25 keV FWHM for 8.1 MeV  $\alpha$ -particles fully stopped in the DSSSD and 170 keV for  $\alpha$ -particles that deposited a fraction of their energy inside the DSSSD and the remainder in the SSSSD. The detection efficiency was 72% for  $\alpha$ -particles and 100% for SF. The efficiency for focusing element 114 EVRs into the DSSSD was (60±6)% [9]. Data acquisition was triggered by events registering more than 300 keV in the DSSSD or more than 500 keV in a SSSSD. More details are given in [10,11].

#### Results

We searched for decay chains from <sup>288,289</sup>114 and their daughters [1]. Upon identification of a chain, additional  $\alpha$ -particles occurring in the same pixel as the chain, in between registration of the EVR and the terminating SF, were searched. Due to a damaged target, the background rate was increased during portions of the runs. SFs terminating decay chains recorded during these periods were required to occur outside beam pulses. Based on the event rate only 0.02 (<sup>289</sup>114) and 0.05 (<sup>288</sup>114) random chains from unrelated background events were expected. The search yielded nine EVR- $\alpha$ -SF chains (<sup>288</sup>114) and four EVR- $\alpha$ - $\alpha$ (- $\alpha$ )-SF chains (<sup>289</sup>114). Ten chains were measured in the 42-MeV run and three chains in the 38-MeV run (Figs. 1 and 2). The agreement of our data (Table 1) with that of [1] is good in all cases except for chain #9. The data measured for the EVR, the first, and the second  $\alpha$ -particle suggest assigning chain #9 to  $^{289}114 \rightarrow ^{285}112 \rightarrow$  $^{281}$ Ds.  $^{281}$ Ds then decayed by emission of a (8.727±0.025)-MeV  $\alpha$ -particle 5.688 s after the decay of <sup>285</sup>112, during the beam-off period, where background is low. <sup>281</sup>Ds has

<sup>\*</sup> Work supported by the BMBF (06MT247I, 06MT248, 06MZ223I); the GSI-F&E (MT/TÜR, MZJVKR); the Swedish and Norwegian (177538) Science Councils; the US DOE (DE-AC03-76SF00098; DE-AC02-05CH11231; NNSA Fellowship); the Govt. of India (TADDS). \*c.e.duellmann@gsi.de

undergone SF in all ten previously observed decays [1] with  $T_{1/2}=11.1^{+5.0}_{-2.7}$  s. Based on background rates, the probability to register an  $\alpha$ -like event with properties as exhibited by the observed one is only 0.1%. We thus assign it to a so far unobserved  $\alpha$ -branch in <sup>281</sup>Ds. Considering this  $\alpha$ -decay and the thirteen measured SF decays from [1] and our work, an  $\alpha$ -decay branch  $b_{\alpha}$  of  $9^{+16}_{-7}$ % results after correcting for detection efficiency differences for  $\alpha$ -decay and SF. The chain was terminated 4.5 ms later by SF of new <sup>277</sup>Hs.

The B• $\rho$  of element 114 EVRs in 0.8 mbar He was measured to (2.29±0.11) T•m.



Figure 1: Decay chains assigned to <sup>288</sup>114 (chains 1-8) and <sup>289</sup>114 (chains 9, 10) observed during the 42-MeV run. A black triangle in the lower right corner of a box indicates that the beam was off at the time of the event.



Figure 2: Same as Figure 1, but showing decay chains observed during the 38-MeV run.

Table 1.	Decay	properties	([1]	] and thi	s work)
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Isotope	Decay	$T_{1/2}$ (this work)	$T_{1/2}$ (combined)
<sup>289</sup> 114	α	$0.97^{+0.97}_{-0.32}$ s	$2.1^{+0.8}_{-0.4}$ s
<sup>288</sup> 114	α	$0.47^{+0.24}_{-0.12}$ s	$0.69^{+0.17}_{-0.11}$ s
<sup>285</sup> 112	α	$30^{+30}_{-10}$ s	$29^{+11}_{-6}$ s
<sup>284</sup> 112	SF	$101^{+50}_{-25}$ ms	$99_{-16}^{+24}$ ms
<sup>281</sup> Ds	SF/a:91/9	$20^{+20}_{-7}$ s	$13^{+5}_{-3}$ s
<sup>277</sup> Hs	SF	$3^{+15}_{-1}$ ms	$3^{+15}_{-1}$ ms

#### Discussion

Z=108 is a deformed proton shell closure in N~162 isotopes. The observed <sup>277</sup>Hs lifetime is short compared to half-lives of the Hs isotopes near the deformed N=162 shell closure, indicating reduced shell stabilization in the N=169 nucleus <sup>277</sup>Hs. Macro-microscopic model predictions of  $T_{1/2}(SF)$  for the neighboring isotopes are 46 ms  $(^{276}\text{Hs})$  and 0.98 ms  $(^{278}\text{Hs})$ , the geometric mean being 6.7 ms [12]. This is similar to our observed lifetime. The odd neutron is expected to hinder SF decay significantly. Thus, the drop in  $T_{1/2}(SF)$  when increasing N above 162 may be more severe than suggested by [12]. <sup>275</sup>Hs (N=167) decays by  $\alpha$ -particle emission with T<sub>1/2</sub>=0.19 s [1]. The experimental trend with prevalent  $\alpha$ -decay in Hs isotopes with N=157-167, but predominant SF in lighter as well as in heavier isotopes is close to that in [10], which suggests dominant α-decay from N=154 to N=166 but SF for N>168. This indicates that stability vanishes rapidly with increasing distance from N=162.

Measured cross sections in the 38-MeV run were  $8.0^{+7.4}_{-4.5}$  pb (3n channel) and  $2.8^{+4.2}_{-2.1}$  pb (4n channel), and in the 42-MeV run,  $3.5^{+3.3}_{-2.0}$  pb (3n channel) and  $9.8^{+3.9}_{-3.1}$ pb (4n channel). Error bars include statistical uncertainties only (68.3% confidence level); the systematic uncertainty is estimated to 14%. In contrast to any other confirmation experiment, we confirm the large cross sections as reported from FLNR [1]. In fact, our measured cross sections are higher than those reported from the DGFRS. These high cross sections call for investigations of the details of the production mechanism. Production rates that follow from these values encourage using this nuclear reaction to produce relatively long-lived isotopes of element 114, in particular for envisaged chemical investigations [13] or for  $\gamma$ -spectroscopic studies that allow shedding light on the nuclear structure in this SHE region and may facilitate unique Z identification.

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## **COMPACT Coupled to TASCA for Element 114 Chemistry\***

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The unambiguous identification of new superheavy elements (SHE) is a very difficult task for both, physicists and chemists due to very low production rates, absence of a link to known isotopes, and unknown chemical, and decay properties. Chemical studies of SHE are of great importance because they can identify the proton number of the studied isotopes. Coupling of chemistry setups to physical recoil separators allows chemical experiments with extremely high sensitivity due to a strong suppression of unwanted byproducts in the preseparator [1]. At GSI, the new gas-filled separator TASCA has been put into operation in 2008. It is designed specifically for chemical studies of transactinides produced in nuclear fusion reactions of <sup>48</sup>Ca beams with actinide targets [2].

The highest cross section for SHE formation was observed in the reaction  ${}^{48}\text{Ca}+{}^{244}\text{Pu}$  [3,4] leading to element 114 (E114). First attempts to chemically identify E114 were performed by a PSI-FLNR-LLNL collaboration in Dubna, and an unexpectedly low adsorption enthalpy of element 114 on gold was reported [5], in contradiction with theoretical predictions of the trend in the binding energy, *Eb*, Pb>>E114>Hg>E112 [6]. These studies were performed without preseparation, and the relatively high counting rate from unwanted byproducts led to controversially discussed results. An attempt to observe E114 in the chemistry experiment after preseparation failed [7].

A chemistry experiment with E114 was carried out at TASCA in 2009. Because the lifetimes of even the longest-lived E114 isotopes, <sup>288,289</sup>114, are short [4], TASCA was operated in the Small Image Mode (SIM) [2], which focuses fusion products into a small area of about ~30x40 mm<sup>2</sup>. The lower transmission efficiency of SIM (~35%) compared to HTM (~60%) is compensated by the smaller volume of the Recoil Transfer Chamber (RTC), which allows a faster transport of products to a detection setup. During the experiment on the synthesis of <sup>288,289</sup>114 [4], the operation of TASCA in SIM was successfully tested with a focal plane detector. A beam dose of 0.98·10<sup>18 48</sup>Ca ions at E\*(<sup>292</sup>114)=42 MeV was acquired and two <sup>288</sup>114 decay chains were observed. In the preparation of the experiment two RTCs made of Teflon<sup>™</sup> were tested: a "small" one with a volume of 14 cm<sup>3</sup> and a "large" one with 29 cm<sup>3</sup>. Transport times and yields to COMPACT [8] were optimized for both chambers with short-lived Hg and Pb isotopes produced with <sup>40</sup>Ar and <sup>48</sup>Ca beams. Transport times of 0.6 s and 0.8 s were measured at a gas flow rate 1.3 l/min for the "small" and "large" RTC, respectively. Three different He/Ar gas mixtures with ratios of 30:70, 50:50, and 70:30 were explored. The last one was selected for the E114 chemistry experiment. Two similar COMPACT detectors connected in series were used; each detector consisted of 32 pairs of 1x1 cm<sup>2</sup> PIN diodes covered with a 35-nm thick gold layer. The first detector, kept at the room temperature, was connected directly to the RTC exit via a 2-cm long Teflon<sup>™</sup> tube. The second detector, connected via a 30-cm long Teflon<sup>TM</sup> capillary, was placed downstream of the first one; a temperature gradient from +20 to -162 °C was applied along it. The use of two detectors in series allows detecting species in a wide range of volatilities - from the nonvolatile Pb to the noble gas Rn (Fig. 1). With this setup the adsorption enthalpy of short-lived <sup>288,289</sup>114 on gold was measured with relatively high efficiency under background-free conditions. The data are under evaluation.



Figure1:Pb, Hg and Rn distributions in COMPACT.

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## <sup>244</sup>Pu-targets for production of element 114 at TASCA

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Introduction: In a series of recent experiments at TASCA, production and decay as well as chemical properties of element 114 have been investigated using the <sup>244</sup>Pu(<sup>48</sup>Ca,3/4n)-reaction leading to <sup>288,289</sup>114 [1,2]. Because the <sup>244</sup>Pu target material is available only in very limited amounts, the target preparation technique should give high yields. Easy and complete recovery of the target material is another pre-requisite [3]. Thus, we have chosen Molecular Plating (MP) onto 2 µm thin pinhole-free titanium foils as the target preparation technique. At TASCA, a rotating target wheel is used composed of three banana-shaped target segments with an active target area of 1.44  $\text{cm}^2$  each. The rotating target is confined in a nearly closed container in order to protect the beam line as well as the separator against contamination in the case that a target gets destroyed.

<sup>244</sup>Pu target production and characterization: For the production of one target segment by MP about 1 mg of <sup>244</sup>Pu in the form of its nitrate is dissolved in a small volume (100-200 µl) of nitric acid in a Teflon<sup>™</sup> beaker and mixed with a surplus of isopropanol (800 µl). The mixture is then transferred into the electrochemical deposition cell (EDC) made of Teflon<sup>™</sup> which is subsequently filled up with isobutanol to a total volume of 16 ml [4]. MP is carried out by applying a voltage of 150-200 V at a maximum current density of about 1.2 mA/cm<sup>2</sup>. After 5-6 hours plating time, deposition yields up to 90 % are achieved. The backing foils are produced by cold rolling at GSI [5]. They should be pinhole-free and are precleaned with isopropanol, 6 M hydrochloric acid and water. Prior to use, the foil integrity is checked by optical microscopy to ensure that the backing is pinhole-free. The average foil thickness is determined by weighing, whereas the homogeneity of the foil thickness is checked by  $\alpha$ -particle energy-loss measurements. For a target backing foil with a nominal thickness of 2.2 µm deviations are in the order of  $\pm 0.2$  µm.

The <u>target thickness</u> is determined by two independent methods: (i) *a-particle spectroscopy*. After the deposition is completed, the target is dismounted from the EDC, dried under an infrared lamp and measured with a surface barrier  $\alpha$ -detector at a distance of about 30 cm. (ii) The Pu-content of the solution in the EDC is determined by *Neutron Activation Analysis*. Subsequent to MP an aliquot of the supernatant solution in the EDC (1 ml) is irradiated for 2 h in the TRIGA Mainz research reactor with a thermal neutron flux of 7 x 10<sup>11</sup> cm<sup>-2</sup>s<sup>-1</sup>. Here, 10.5 h-<sup>245</sup>Pu is formed via the reaction <sup>244</sup>Pu(n, $\gamma$ )<sup>245</sup>Pu.

The Pu content of the irradiated solution is determined by means of  $\gamma$ -spectrometry using the prominent  $\gamma$ -lines resulting from the <sup>245</sup>Pu decay at 327 keV,

308 keV, and 560 keV, respectively [3]. Table 1 comprises all  $^{\rm 244}{\rm Pu}{-}{\rm targets}$  produced for TASCA so far.

Table 1: <sup>244</sup> Pu-targets for TASCA		
Target#	Thickness [µg/cm <sup>2</sup> ]	
08-395	401	
08-482	502	
08-485	490	
08-486	390	
08-487	472	
09-562	673	
09-594	724	
09-623	790	
09-624	785	

The homogeneity of the Pu-layer is checked with radiography [6] using a commercial radiographic imager (FLA 7000 from FUJIFILM Corp.). Figure 1 shows a picture of a target segment. Here, the brown layer indicates the Puoxide deposit. Also shown is a 3-dimensional plot of the activity distribution. With this technique it could be shown that the active target area is completely covered and, in addition, that Pu is homogeneously distributed over the entire target area. From this one can conclude that variations in target thickness are in the order of 15%.



Figure 1: <sup>244</sup>Pu target wheel for TASCA as used for the production of element 114. The brown layer indicates the Pu-oxide deposit.

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# Recovery of <sup>244</sup>Pu from irradiated targets for production of element 114\*

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The <sup>244</sup>Pu targets [1] (PuO<sub>2</sub> electrodeposited on Ti backing) were irradiated during recent bombardments with <sup>48</sup>Ca<sup>10+</sup> ions to produce <sup>288, 289</sup>114 [2,3]. During these bombardments with up to 3.6 x  $10^{18}$  ions, targets and backings underwent changes that made reprocessing and production of new targets for forthcoming experiments desirable.



Figure 1: Teflon vessel.

Recovery of <sup>244</sup>Pu from one arc-shaped segment was accomplished as follows: The Al-target frame was inserted into a Teflon vessel containing a cavity into which the frame could be inserted, see Figure 1. With a sharp knife, the target was cut out of the frame, the frame was removed, and the target together with the Ti backing was dissolved in hot conc. HCl. The dissolution of the Ti backing was incomplete. The central part of the backing that had received the highest beam intensity did not dissolve. The resulting solution with the remainder of the undissolved Ti was evaporated to near dryness, transferred into a 10 ml measuring flask and filled with 8 M HCl. An aliquot of that solution was removed, evaporated to dryness, and the  $\alpha$ -particle activity was determined. The total activity was used for yield determination. The results indicate that more than 80 % of the Pu had been recovered.

\*Sponsored in the frame of a GSI R&D project (MZJVKR).



Figure 2: Alphaspectrum of the Pu eluate.

The Pu/Ti solution in 8 M HCl was transferred to a AG 1x8 anion-exchange column (3 x 50 mm), and was washed subsequently with 10 x 1 ml of 8 M HCl to remove the Ti and the <sup>241</sup>Am from the column. Then, the Pu was eluted from the column in 8 x 1 ml of 0.5 M HCl. Figure 2 shows the spectrum of  $\alpha$  particles of an aliquot of the eluate. Due to the isotopic composition of the plutonium (97.9 %  $^{244}$ Pu, 1.3 %  $^{242}$ Pu, 0.7 %  $^{240}$ Pu,  $^{<}_{220}$  0.1% other), the main  $\alpha$  activities are associated with  $^{238}\text{Pu}$  and  $^{239}$  Pu. 100 µl of that solution was removed, filled up to 2 ml and was irradiated with thermal neutrons in the TRIGA reactor at the Institute of Nuclear Chemistry at the University of Mainz at 100 kW together with a second reference sample containing 9.62  $\mu$ g <sup>244</sup>Pu for 6 h. After a decay time of 18 h, both samples were assayed for the 327.6 keV γ-activity of <sup>245</sup>Pu at a Ge detector, see Figure 3. The activation analysis showed a <sup>244</sup>Pu recovery of 89 %.



Figure 3: Gammaspectrum of <sup>245</sup>Pu solution.

- [1] K. Eberhardt *et al.*, contribution to this report.
- [2] Ch. E. Düllmann et al., contribution to this report.
- [3] A. Yakushev et al., contribution to this report.

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# The Performance of TASCA in the <sup>48</sup>Ca+<sup>206,207,208</sup>Pb Reactions

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The gas-filled recoil separator TASCA (TransActinide Separator and Chemistry Apparatus) was installed in a dipole-quadrupole configuration (DQQ) at the UNILAC at GSI [1-3]. An extensive commissioning program [4] was carried out at TASCA studying a large number of experimental parameters and nuclear reactions. Depending on the polarity of the quadrupole magnets TASCA can be operated in two modes: the so called High Transmission Mode (HTM) and the Small Image Mode (SIM). Dispersion values of 9 and 1 mm per one percent change of  $B\rho$  were calculated for the HTM and SIM, respectively [3]. Ion optical calculations of the HTM and SIM were performed using Monte-Carlo simulations [5]. Important characteristics of TASCA in both modes were investigated using <sup>48</sup>Ca+Pb reactions and a 16-strip 80x35 mm<sup>2</sup> large position-sensitive silicon-strip detector based focal plane detector (FPD). Optimal magnetic settings and gas pressures were established by centering spatial distributions of  $\alpha$ -decaying evaporation residues in the FPD. Transmission measurements were performed with targets of well determined thicknesses.

#### HTM:

Measured spatial distributions of <sup>254</sup>No ions are shown in Fig. 1 for different helium gas pressures and a constant dipole magnet setting of  $B\rho$ =2.08 Tm. Solid curves show the calculated [5] distribution of <sup>254</sup>No ions in the FPD of TASCA at different gas pressures. Well centered distributions were observed in the range of 0.8 to 1.0 mbar pressure range. These distributions are in very good agreement with the calculated ones. However, at lower and higher gas pressures the <sup>254</sup>No distributions are horizontally shifted off-center. This means that the deflection angle of <sup>254</sup>No in the dipole magnet is changing. This is related to a change of average charge of <sup>254</sup>No ions. Such an effect was observed also at the Dubna gas-filled separator and it was explained with so-called "density effect" [6].

An average value of  $(57\pm5)$  % for the transmission of No isotopes synthesized in <sup>48</sup>Ca on <sup>206-208</sup>Pb reactions was deduced using the cross-section data for fusionevaporation reactions from [7]. This value is in good agreement with the calculated value of about 52 % for the HTM of TASCA.

#### SIM:

To find optimal settings for the quadrupole magnets in the SIM is more difficult than for the HTM. Various settings for the quadrupole focusing were tested to obtain best values. A 40-mm diameter image size was taken as a "reference" best value. Again, deviations between this optimized result, which is in agreement with theoretical calculations, and distributions obtained at pressures lower than the optimal He pressures were observed.

A transmission of (35±5) % was deduced at optimized SIM settings for the reaction  ${}^{48}Ca+{}^{208}Pb$ . This value is in good agreement with calculated values.



Fig 1: Spatial distributions of <sup>254</sup>No ions in the FPD at different pressures of the He filling-gas. TASCA was operated in HTM. Dashed lines show the Gaussian fit.

Pure hydrogen and He-H<sub>2</sub>-mixtures were used as filling gases in both modes as well. The optimal magnetic settings and gas pressures were investigated and the corresponding average charges of nobelium ions were determined. Within a 10% uncertainty, the measured transmissions for He, H<sub>2</sub> and mixtures of both gases were identical. When pure H<sub>2</sub> and a mixture of He and H<sub>2</sub> were used, we observed a better background suppression of targetlike ions as compared with pure helium.

More detailed information on the TASCA performance in <sup>48</sup>Ca+Pb reactions and average charges of the nobelium ions in various gases will be given in [8].

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## **TASISpec – Heading towards its first experiment.**

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TASISpec (TAsca Small Image mode Spectroscopy) [1, 2] is a Si and Ge detector setup optimised for particle- $\gamma$ -X-ray coincidence spectroscopy of superheavy elements in conjunction with the TASCA separator [3]. The detection system consists of 192 Si strips distributed over one double sided silicon strip detector (DSSSD) and four single sided silicon strip detectors (SSSSD). The DSSSD is the focal plane detector into which the residual nuclei are implanted and their subsequent decay products such as fission fragments or  $\alpha$  particles are detected. The SSSSDs form a "box" upstream from the DSSSD and they are used to detect  $\alpha$  particles which have escaped detection in the DSSSD, conversion electrons (CE) and possibly the second fission fragment. A seven-crystal Ge cluster detector is mounted directly behind the DSSSD and four clover detectors are mounted behind the four SSSSDs. The complete setup is thus composed of a total of 23 Ge crystals.

The setup is constructed to enable multi-coincidence spectroscopy such as  $\alpha$ - $\gamma$ -CE and  $\alpha$ - $\gamma$ - $\gamma$  with unprecedented  $\gamma$ -ray efficiency and thus reveal essential information necessary to build reliable level schemes for superheavy elements.

During 2009 a thorough evaluation of the commissioning experiments was performed [2]. This involved amongst others detection efficiencies and implant-decay correlation times. As an example, the decay of <sup>253</sup>No has been explored. The half life of the ground state was previously determined to  $T_{1/2} = 1.56(2) \min [4]$ . In the present analysis the half life is determined to  $T_{1/2} = 1.61(21)$  min where the uncertainty originates mainly from the small number of  $\alpha$  particles, which could be included in the analysis. In Fig. 1 the DSSSD hitpattern is shown. It shows the pixels in which the 8.0 MeV  $\alpha$  particles relating to the decay of the ground state of <sup>253</sup>No were detected. As can be seen in this figure the implants are nicely focused into a very narrow spot. Since the half life of <sup>253</sup>No is rather long the pixels in the very centre of the focal spot where the implantation rates are at its highest, were excluded when determining the half life. This minimises the risk of random correlations between the incoming evaporation residues and the  $\alpha$ particles.

In a commissioning run in July 2009 was the reaction  $^{208}$ Pb( $^{48}$ Ca,1n) $^{255}$ No applied to explore the benefits

gained from the usage of pulse-shape electronics. Pulse shape analysis could yield particle identification due to distinct ionisation schemes in the semiconductor material for different incidenting particles, like e.g.  $\alpha$  particles and CE.

In the beginning of 2009 TASISpec was granted beamtime for its first main beam experiment. The experiment is scheduled for spring 2010 and will aim to explore Kisomers in <sup>253</sup>No in detail.



Figure 1: The position of the detected  $^{253}$ No correlated alpha particles in the DSSSD. On the *x* and *y*-axis the size of the DSSSD is indicated.

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## Pilot-Test Experiment with Os of a SISAK Setup for Hs-Chemistry Studies

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A liquid-liquid extraction system for investigating chemical properties of element 108, hassium, was developed [1] using  $\gamma$ -emitting <sup>181</sup>Os produced at the Oslo Cyclotron Laboratory (OCL). The system is targeted for the fast solvent extraction system SISAK [2] and based on OsO<sub>4</sub> reacting with NaOH. Such an experiment would be the first attempt to study Hs in the liquid phase. Successful Rf and Db experiments [3-5] performed at LBNL in Berkeley indicate that SISAK with its liquid scintillation detectors is sensitive enough to detect Hs, even though the Hs cross section is ~3 orders of magnitude lower.

The first investigation of the reaction between  $HsO_4$ and NaOH was performed in a gas phase experiment [6]. The interaction of  $HsO_4$  appeared somewhat weaker with NaOH than that of  $OsO_4$ , in fair agreement with theoretical predictions [7]. The liquid-liquid extraction Hsexperiment proposed in the work presented here is based on results from this gas-phase experiment. In aqueous solution, it is assumed [1] that the reactions occurring are:

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

$$Na[OsO_4(OH)] + NaOH \leftrightarrow Na_2[OsO_4(OH)_2]$$
(2)

$$OsO_4(aq) \rightarrow OsO_4(org)$$
 (3)

The distribution ratio between NaOH solution and toluene, which was selected as organic phase because it is also suitable as solvent for the liquid scintillation detection used by SISAK, is given by:

$$D = \frac{[OsO_4]_{org}}{[OsO_4]_{aq} + [OsO_4(OH)]^{-} + [OsO_4(OH)_2]^{2^{-}}}$$
(4)

which can be rewritten as:

$$D = \frac{K_D}{1 + K_1 [OH^-] + K_1 K_2 [OH^-]^2}$$
(5)

where  $K_I$ ,  $K_2$  and  $K_D$  are equilibrium constants for reactions (1), (2), and (3), respectively. Experiments were performed in Oslo, utilizing manual extractions and SI-SAK on-line measurements to carefully study the behavior of Os in this chemical system, see Samadani et al. [1] for details. The results are summarized in Fig. 1.

Based on the results from Oslo a "proof-of-principle" experiment with  $\alpha$ -decaying Os isotopes was performed at GSI: the full SISAK setup [8], as it would be used for a Hs experiment with double  $\alpha$ -detector arrays to simultaneously measure both phases (for the aqueous phase done indirectly, after a second extraction step) was set up and tested. <sup>40</sup>Ar<sup>11+</sup> ions from the UNILAC irradiated a <sup>nat</sup>Ce target in the gas-filled separator TASCA (TransActinide Separator and Chemistry Apparatus) producing <sup>172-175</sup>Os.



Figure 1: Comparison of data from OCL and GSI, together with fit of eq. (5) to OCL data.

In the separator focal plane a Recoil Transfer Chamber (RTC) was mounted. It was flushed with a He/O<sub>2</sub> gas mixture, which passed an oven (run at 600°C) mounted at the exit of the RTC to ensure fast and complete oxidation of Os. The volatile osmium tetroxide was transported to SISAK by the He/O<sub>2</sub> gas and dissolved in NaOH solution. After extraction into toluene the  $\alpha$ -activity was measured in on-line flow cells by liquid scintillation detection. This was the first SISAK experiment behind TASCA. Results from this run using  $\alpha$ -decaying <sup>172</sup>Os agree well with those of  $\gamma$ -measurements obtained in Oslo, as shown in Fig. 1. This successful experiment proved that the system is suitable for studying Hs.

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## Liquid – liquid – extraction with the MicroSISAK – system

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For studies of the chemical properties of the heaviest elements (Z > 103), it is indispensable to use fast and efficient systems, because of their short half-lives and low production rates [1]. A promising possibility for liquid chemistry in this field is the liquid – liquid – extraction by using micro reaction technology [2]. Therefore the *Institut für Mikrotechnik Mainz* developed a device, that agitates two liquids via a static digital mixer and separates them again via a hydrophobic Teflon membrane. The past experiments have shown that the principal idea could be realised with this apparatus called *MicroSISAK*.

The most important parameters to look at, are the separation of both liquid phases and the extraction yield. Last year, we reported on an efficient apparatus, with that the separation was increased by setting a backpressure at the outlet of the aqueous phase of our device [3]. The higher the flow rate we use to put the liquids in the microreactor, the higher is the needed backpressure to separate them. The correlation of flow rate and back pressure is shown in Figure 1 for a yield of separation not less than 98%.



Figure 1. Required back pressure for a separation yield over 98% depending on the flow rate

The extraction yield has been studied in different phase systems (see Table 1):

Table 1: reviewed extraction systems

nuclid / amount	aqueous solution	extraction agent
Hf-181 ~10 <sup>-6</sup> mol/l	H <sub>2</sub> SO <sub>4</sub> 0,5 mol/l	TOA 1 mol/l in
		toluene
Hf-181~10 <sup>-6</sup> mol/l	HNO <sub>3</sub> 6 mol/l	DBP 0,25 mol/l in
		toluene
Tc-99m carrier-free	HNO3 0,01 mol/l	TPAC 10 <sup>-4</sup> mol/l in
		CHCl <sub>3</sub>

The calculation of the extraction yield results from the  $\gamma$ decay of the used nuclide. The activity in the organic phase after mixing and separation is compared with the one in the aqueous phase before the latter is pumped in the micro reactor. After some batch experiments, the three extraction systems were tested with different flow rates. The results of two systems are compared with the batch experiments in Figure 2 and 3.



Figure 2. Extraction yield vs. flow rate for Hf with TOA, the red line shows the results of the batch experiments



Figure 3. Extraction yield vs. flow rate for Tc with TPAC, the red line shows the results of the batch experiments

For these three extraction systems and the attempted flow rates, the extraction yield ranges between 15% and 45%. This is unsatisfactory.

To find out, why the extraction is not as good as expected, there are experiments running to explore the effects of the contact time of the emulsion. There are also plans to run the MicroSISAK with higher temperature to check out the influence of the diffusion between the two phases.

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### Accuracy studies and first mass measurements at TRIGA-TRAP\*

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**Introduction:** At present atomic masses are obtained with the highest precision by measuring the cyclotron frequency of a stored ion in a Penning trap [1]. TRIGA-TRAP at the research reactor TRIGA-Mainz is so far the only Penning trap mass spectrometer installed at a nuclear reactor, which provides short-lived neutron-rich radionuclides in the mass region 80 < A < 140 by thermal-neutron induced fission. The goal of TRIGA-TRAP is to perform mass measurements on these nuclides in order to contribute to improvements of nuclear structure studies, nucleo-synthesis calculations and the predictive power of nuclear mass formulas [2].

**Results:** At TRIGA-TRAP the time-of-flight ion cyclotron resonance (TOF-ICR) method is routinely used to determine cyclotron frequencies of ions produced by offline ion sources. A laser ablation ion source [4] provides carbon clusters  ${}^{12}C_n^+$  as reference ions for mass calibration and to study systematic uncertainties of the mass spectrometer. One systematic uncertainty due to nonlinear magnetic field fluctuations depends on the time elapsed between two measurements  $\Delta t$ . The magnitude of this effect was determined in a long-term measurement of the cyclotron frequency of  $C_{20}^+$  to be

$$\frac{\Delta \omega_{ref}}{\omega_{ref}} = 4 \cdot 10^{-11} \,/ \min \cdot \Delta t.$$

A trap misalignment leads to a systematic uncertainty in the mass calibration which increases with the mass difference of the ion of interest to the reference ion  $(m - m_{ref})$ . This so-called mass-dependent systematic effect was investigated by the determination of well-defined frequency ratios r of carbon cluster ions over a broad mass-range. About 70 frequency ratio measurements yield

$$\frac{\Delta r_m}{r} = -1.8(1) \cdot 10^{-9} \ (m - m_{ref}) / u$$

as the result for the mass-dependent systematic effect at TRIGA-TRAP. After correction of these two systematic effects the residual relative systematic uncertainty of the measurements was found to be  $2.5 \cdot 10^{-8}$ . A detailed description of these tests is given in [4]. Ions for a mass measurement of <sup>197</sup>Au have also been

Ions for a mass measurement of <sup>197</sup>Au have also been produced with the laser ablation ion source using a gold foil as target. The literature value of the mass of <sup>197</sup>Au listed in the atomic-mass evaluation (AME) 2003 [5] was confirmed with the TRIGA-TRAP mass spectrometer. The frequency ratio to  $C_{16}^+$  was determined with a precision of 6.9 keV ( $\delta m/m = 3.7 \cdot 10^{-8}$ ). A cross-check with  $C_{15}^{+}$  has been performed at the same time to ensure the accuracy of the measurement. The results of the measurements are shown in Fig. 1.



Figure 1: Results of the mass measurement of  $^{197}Au^+$ . The red lines represent the uncertainty of the literature values in [5], and the data points the results of TRIGA-TRAP.

**Outlook:** Further offline mass measurements on lanthanoids and actinoids will be performed in 2010. Concerning the online coupling to the TRIGA reactor, a gas-jet system is being connected to an ECR ion source to transport and ionize the radionuclides. An RFQ buncher is being installed to provide cooled ion bunches for highprecision mass measurements on neutron-rich fission products.

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# Commissioning of TRIGA-LASER - tests and developments of the LaSpec beamline

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**Introduction:** The major parts of the TRIGA-LASER experiment for collinear laser spectroscopy on exotic fission products from the TRIGA research reactor [1] are now installed and are currently being commissioned. Besides the exploration of unknown ground in the nuclear chart with respect to spins, moments and charge radii, the experiment is the prototype of the the LaSpec experiment [2] at FAIR, GSI and will serve as a development platform until the final installation at FAIR.

Experimental: For commissioning, laser spectroscopy experiments were carried out with stable Rb ions from an offline ion source. The charge exchange cell (CEC) and the optical detection system were installed at the beamline and tested. Charge exchange of the rubidium ions was accomplished with potassium vapor produced in the cell at temperatures of  $100 - 200^{\circ}$  C. A charge exchange efficiency of 99% can be reached. However, laser spectroscopy experiments will be performed with efficiencies of 20 - 40% at about 140° C to avoid artificial broadening of the resonance lines. To change the velocity of the neutralized beam for Doppler tuning, the CEC can be floated to a potential of up to 10 kV. Voltage scans can either be performed with a Heinzinger PNChp10000 high-precision high-voltage power supply programmed via GPIB or with a Kepco HV amplifier controlled by the voltage of a 16 Bit DAC. The fluorescence detector after the CEC is a combination of a light collecting elliptical mirror with a light guide and a photomultiplier. The Hamamatsu R1017 multiplier exhibits 5% quantum efficiency at the Rb transition wavelength of 780 nm.

Results: The first laser spectroscopy experiments were performed with Rb atoms after charge exchange with an acceleration voltage of 10 kV and a maximum Doppler tuning voltage of  $\pm 1$  kV. This range is equivalent to approximately 9.5 GHz and sufficient to record the full Rb hyperfine structure of both naturally abundant isotopes <sup>85</sup>Rb and <sup>87</sup>Rb simultaneously. This is shown in Fig. 1, which shows the complete hyperfine structure as a function of the scanning voltage. The inset shows the strongest peak and demonstrates the resolution. The linewidth of 25 MHz is close to the natural linewidth of 6 MHz. The detection efficiency was optimized and a value of  $\epsilon \approx 1$  photon / 360 atoms was achieved. The background caused by scattered laser light is about 150 kHz at 0.6 mW and therefore still too high. However, a signal-to-noise ratio of  $\approx 2$  in a 6 h integration time, assuming a resonance scan with 20 points, can be achieved with an atom beam intensity of  $1 \times 10^4$  atoms/s. Currently, a



Figure 1: Full hyperfine spectrum of <sup>85</sup>Rb and <sup>87</sup>Rb and enlarged scan of the strongest peak. Spectra are the sum of 10 and 50 scans for the overall spectrum and the single peak, respectively ( $P_{\text{Laser}} = 0.54 \text{ mW}, I = 3 \times 10^7 \text{ atoms/s}$ ).

particle detection system for low count rates is being tested which will allow to perform particle-photon coincidence measurements to increase the sensitivity [3]. Lower straylight background and higher detection efficiency of fluorescence light are the objectives of a new light collection module which is currently being designed and simulated. Coupling to the reactor and first online experiments are expected for the end of 2010.

#### References

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