Extracts of the 2007 Annual Report of the (former) JGU "Institute of Nuclear Chemistry"

The deposition of ^{239}Pu on thin Ti backings by molecular plating

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Many transactinide elements can be produced in heavy ion fusion reactions with ^{244}Pu targets. For this, target thicknesses of 500 µg/cm² are required. Since ^{244}Pu is available only in very limited amounts, the target preparation technique should give high deposition yields. Easy and complete recovery of the target material is another prerequisite. Molecular plating (MP) is well suited for the preparation of lanthanide and actinide targets on metallic and non-metallic backing materials with deposition yields > 80 %. For the molecular plating the actinide material normally is used in its nitrate form, from which 10µl were dissolved in 16 ml isobutanolic solution. The electrodeposition cell consists of Polyetheretherketone (PEEK) and silicon seals.

The isotope ^{239}Pu is used to optimize the deposition parameters for Pu as well as to study the performance of Pu layers on thin Ti backings during irradiation with intense ^{22}Ne beams as applied at the UNILAC accelerator of the Gesellschaft für Schwerionenforschung (GSI). For the **T**rans-**A**ctinide **S**eparator and **C**hemical **A**pparatus TASCA at GSI a rotating target wheel assembly consisting of three banana shaped segments with an active target area of 1.74 cm² is used. Here, thin Ti foils (2 µm) serve as backing material.

In the initial deposition tests with very small amounts of ^{239}Pu (~ 10 kBq or < 5µg) serious adsorption losses occurred due to the special chemical nature of Pu. Pu appears in aqueous solution predominantly in the +IV and the +VI state, where especially Pu(IV) is known to form colloids and polyhydroxides in pH ranges > 1. These species tend to adsorb strongly onto glass and polymers like PEEK and silicon.

To circumvent these problems, the original Pu nitrate solution was fumed to dryness with 8 M perchloric acid to transfer the Pu into the higher oxidation state +VI, in order to suppress the formation of polynuclear Pu species. For further experiments, the Pu is kept in 8 M perchloric acid. Furthermore, different plastic and elastomeric materials were investigated with regard to their adsorption tendency for Pu. Here, equal samples of each material were contacted with 16 ml ^{239}Pu solution (11 kBq total), stirred and treated by ultrasound for 60 minutes. The amount of Pu adsorbed on the surface was measured by α-particle counting. As a result, PTFE shows the least adsorption and therefore was chosen as new cell material. Unfortunately, there was no alternative for the relatively high adsorbing silicon seals. With the new PTFE deposition cell, molecular plating experiments were performed with a total of 200 µg ^{239}Pu corresponding to a target thickness of 125 µg/cm². The first deposition still resulted in a loss of Pu due to adsorption of 27 %, whereas during the second procedure only 4 % of Pu was lost. In further depositions, a loss of material was not longer noticed. This effect leads to the suggestion that the surface of the deposition cell now was saturated with Pu since it was not cleaned between subsequent depositions. Then, the intake of Pu was increased to 1000 µg in order to obtain targets with approximately 500 µg/cm². Here the deposition yield ranged between 70 and 86 %. Table 1 shows all ^{239}Pu targets made in the PTFE cell.

Table 1: ^{239}Pu-targets made in new PTFE Cell

Target	Pu layer [µg/cm²]	Note
425Pu239	26	
414Pu239	108	
416Pu239	116	
417Pu239	476	backing ripped
419Pu239	423	
422Pu239	not determined	target peeled off

Figure 1 shows the picture of the target 416Pu239 and the corresponding alpha particle spectrum used for the determination of the target thickness.

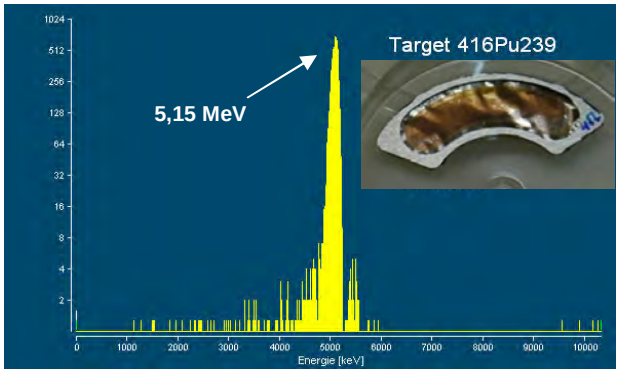

Figure 1: α-spectrum and picture of a target 416Pu239

Current experiments focus on the recovery of the adsorbed Pu by using a mixture of HCl/HF. Following this, a new amount of ^{239}Pu will be conditioned for deposition and three more segments will be prepared for further irradiation tests at GSI scheduled for April 2008.

Acknowledgement

The authors thank the staff of target laboratory at GSI for the preparation of the Ti backings.

Status of the TASCA Commissioning Program*

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The **T**rans**A**ctinide **S**eparator and **C**hemistry **A**pparatus, **TASCA**, project [1] is focusing on the separation and investigation of neutron-rich transactinide nuclides produced in actinide target based reactions. The envisioned research program includes both chemical investigations of transactinide or superheavy elements after preseparation with the gas-filled separator and physics motivated nuclear structure and nuclear reaction studies.

The central device of TASCA is a gas-filled separator in a DQQ configuration. It can be operated in the "High Transmission Mode" (HTM, DQ_hQ_v) and in the "Small Image Mode" (SIM, DQ_vQ_h); see Refs. [1-4] for more details. The separator was installed at the UNILAC beam line X8 and, after having all crucial parts of the control system [5] running, an extensive commissioning program was carried out in 2007. This report briefly summarizes the nuclear reactions applied and the most important parameters studied. A few examples are discussed in a very exemplary way. In addition, recent target developments and the progress in the coupling of chemistry set-ups will be outlined. The first chemical study behind TASCA is described in a separate contribution [6].

All nuclear reactions applied are listed in Table 1 together with the mode of TASCA operation (HTM=H, SIM=S) and the separator gas. Also indicated are experiments aimed to test or apply a recoil transfer chamber (RTC) in addition to measurements performed with a focal plane detector (FPD). As the standard FPD we used a (8x3.6) cm^2 large position-sensitive 16-strip silicon detector. Some experiments were devoted to test prototype double-sided silicon strip detectors (DSSSD) which are planned to be used in future experiments with superheavy elements (SHE).

To understand TASCA as a separator and to build up a solid data base providing good predictive power concerning separator operation for future SHE experiments, we investigated the following most important parameters: (i) the magnetic rigidity of reaction products between Z=76, Os, and Z=102, No, produced at different recoil velocities, and the corresponding best settings of the dipole magnet, (ii) the quadrupole focusing, which is especially relevant for the SIM, (iii) the target thickness dependence of the separator transmission - strongly depending on the asymmetry of the nuclear reaction -, and (iv) the optimum gas pressure with respect to focusing and to transmission - being quite different in the HTM and in the SIM. The analysis of a huge amount of data from these experiments is in progress, and it is important to realize that most of the above mentioned parameters influence each other.

Table 1: Nuclear reactions applied in TASCA commissioning experiments; see text for details.

Beam	Target	Product	Mode	Gas	RTC
^{22}Ne	^{nat}Ta	$^{198m-199}$Bi	H + S	He	
	^{179}Au	^{215}Ac	H + S	He	
	^{238}U	^{255}No	H + S	He	
^{30}Si	no	^{30}Si	H + S	Vac	
	^{181}Ta	$^{205-206}$Fr	H	He	
^{40}Ar	^{nat}Ce	^{173,175}Os	H	He	yes
	^{144}Sm	$^{180-182}$Hg	H + S	He	yes
	^{nat}Gd, ^{152}Gd	$^{194-196}$Pb, ^{188}Pb	H + S	He	yes
	^{nat}Lu	^{210}Ac	H + S	He, N_2	
	^{208}Pb	^{245}Fm	H + S	He	yes
	^{232}Th, ^{238}U	targettest, background	H	He	
^{48}Ca	^{144}Sm	^{188}Pb	H + S	He	
	^{206}Pb	^{252}No	H + S	He	
	^{208}Pb	^{254}No	H	He, H_2	
^{54}Cr	^{nat}Gd	$^{209-210}$Ra	H + S		

Always as a first step, the best dipole setting was found in HTM by centring the product distribution with a typical width of ≈ 6 cm on the FPD. A magnetic rigidity range from 1.5 to 2.2 Tm was covered in those experiments. The quadrupole focusing was found to be insensitive to small quadrupole current changes in the HTM while it reacts very sensitively in the SIM. Optimized SIM settings were determined to obtain maximum rates and narrow distributions of ≈ 1.5 cm FWHM.

The target thickness dependence of the transmission was extensively studied in the reactions ^{22}Ne + ^{197}Au (55, 130, 255, 580 µg/cm^2) and ^{40}Ar + ^{144}Sm (75, 190, 380, 930 µg/cm^2) in both modes. A comparison of these data with model calculations [7] will allow selecting an optimum target thickness with the highest product rate for all the envisioned nuclear reactions.

Many experiments were devoted to find the optimum He pressure and to determine the response to pressure changes. For this we checked the spatial distribution and the total rate of the products in the FPD. While a pressure of about 1 mbar is generally best in the HTM, a signifi-

* Work supported by BMBF (06MT247I, 06MT248, 06MZ223I) and GSI-F&E (MT/TÜR, MZJVKR)

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cantly lower pressure in the 0.2 to 0.5 mbar range gives optimum results in the SIM. A more detailed investigation of the pressure dependence is under way.

One of the most interesting but least understood parameter in the operation of gas-filled separators is the gas filling. In addition to He as our standard gas, we did first test experiments with H_2, N_2, and mixtures of He and N_2. In the ^{40}Ar + ^{nat}Lu reaction we probed the influence of small amounts of N_2 in He on the magnetic rigidity and tested the pressure dependence in pure N_2. From the ^{48}Ca + ^{208}Pb reaction clean α-spectra of ^{254}No and its daughter ^{250}Fm were measured in the HTM with He and H_2 fillings. Figure 1 shows an example obtained with 1.5 mbar H_2.

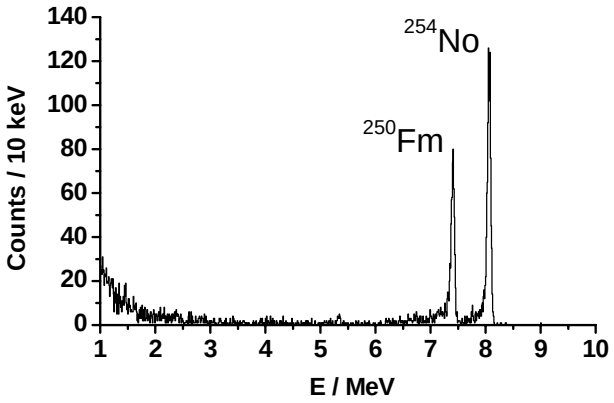

Figure 1: α-spectrum of ^{254}No separated in a 1.5-mbar H_2 gas filling of TASCA. ^{250}Fm is the daughter nucleus.

The ^{22}Ne(^{181}Ta,xn)^{198m,199}Bi reaction was used to check the calculated transmission [7] in the HTM and in the SIM. ^{198m,199}Bi were collected in Al catcher foils directly behind the target (used as the 100% reference value) and in the focal plane. Subsequent γ-ray spectroscopic measurements of these foils allowed determining the transmission and, from a measurement of segments, the spatial product distribution in the focal plane. Very good agreement was found between theoretically calculated transmissions and distributions and the measured ones.

Target development and testing with ^{40}Ar beams of up to 2 μA(particle) continued and concentrated on metallic Th and U targets on 2 μm Ti backings. In addition, large varieties of ^{144}Sm, ^{179}Au and ^{206,208}Pb targets were prepared and used for intense parameter studies at TASCA. Preparations towards new transuranium targets were concentrating on ^{244}Pu. In this ongoing program, considerable progress has been achieved recently.

Commissioning experiments for the RTCs [8], which were built for both two ion-optical modes, focused on finding best conditions for transporting preseparated nuclides to sites where chemistry experiments are envisaged to take place, i.e., a position inside X8 as well as in the nearby radiochemistry laboratory. Suitable nuclides were produced with ^{40}Ar beams, e.g., α-decaying 25-s ^{188}Pb and 4-s ^{245}Fm as well as longer-lived Os, Hg, and Pb isotopes that were identified with γ-ray spectroscopy.

Yields of preseparated Pb isotopes, transported with a He/KCl gas-jet to the chemistry laboratory, were measured as a function of parameters like (i) the thickness of degrader foils installed in front of the RTC window, (ii) the RTC depth, (iii) the pressure inside the RTC, and (iv) the gas-flow rate. Maximum yields of about 65% were obtained for transport to the radiochemistry laboratory through a 10-m long PE capillary at He flow rates of 2.5 L/min at a pressure of 1.2 bar in the RTC.

The product range in the He-filled RTC was measured by inserting catcher foils to positions with different distances from the RTC window. The measured ranges in He turned out to be larger than the values predicted by SRIM calculations, even though the energy loss in the Mylar degrader foil and window agrees well with such calculations. This was confirmed in measurements of EVRs in the FPD after passing through degrader foils.

Preseparated ^{188}Pb was measured after transport into ROMA [9]. Clean α-spectra and high yields allowed determining the half-life of (23.4±0.4) s with better precision than the literature value of (24.2±1.0) [10]. Furthermore experiments were performed where ^{245}Fm was transported into ROMA; see Figure 2. They prove that the TASCA-RTC system is ready for experiments with SHE.

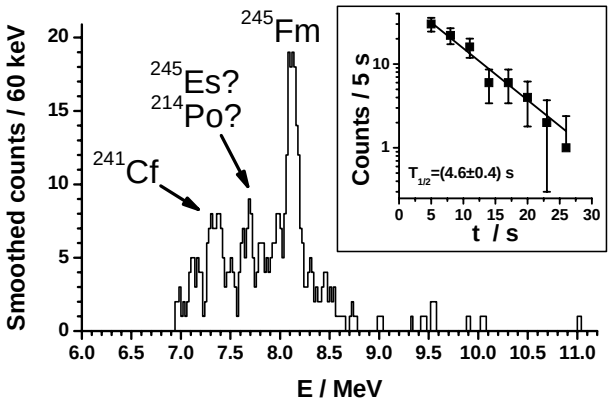

Figure 2: α-spectrum of ^{245}Fm. The insert shows the decay curve for α-particles with 8.05 MeV < E_α < 8.25 MeV.

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First successful chemistry-experiment behind TASCA –Electrodeposition of Os*

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Underpotential deposition has been shown by Hummrich [1] to be a suitable method for studying the chemical behaviour of the transactinides. For such investigations, the nuclide should have a half-life of at least 10 s. Thus, ^{270}Hs ($T_{1/2}$~22 s [2]) is a good candidate for electrochemical experiments. This report describes the first electrodeposition of short-lived isotopes of osmium, the lighter homologous element of hassium.

Os was produced in the reaction ^{nat}Ce(^{40}Ar,xn). The first experiments took place in cave X1 without preseparation. The reaction products were transported via a He/KCl-jet from the recoil chamber, which was directly behind the target, to a direct catch (DC) apparatus and to the Automated Liquid Online Heavy Element Apparatus (ALOHA). DC samples were collected on glass fibre filters which were measured by γ-spectroscopy. No γ-lines of Os isotopes were visible in the spectra due to the high background of transfer products (see figure 1, top), which clearly demonstrates the need for a physical preseparation [3] for such chemistry experiments. Behind TASCA [4], ^{177}Os and ^{176}Os were seen as the main products (see figure 1, bottom).

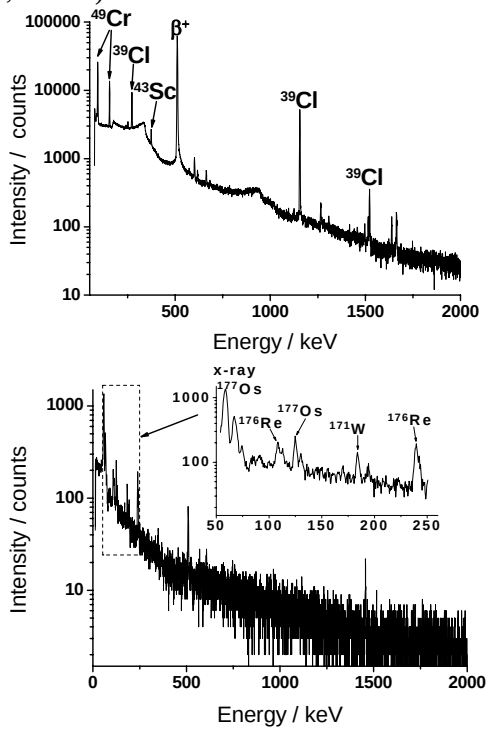

Figure 1: Comparison of the γ-spectra of a sample produced at X1 (top) and at TASCA (bottom).

* Work supported by BMBF (06MZ223I) and GSI-F&E (MZJVKR)

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For the electrochemical studies, the KCl aerosol particles were transported from the HTM-RTC [5] to the radiochemistry laboratory and were deposited on a Ta plate in ALOHA [1]. After 2 min collection time, the sample was dissolved and flushed into the electrolytic cell with 1 ml 0.1 M HCl delivered by a syringe pump. After running the electrolysis for 2 min, the electrodes were measured with a γ detector. The electrolysis was repeated at various potentials vs. an Ag/AgCl reference electrode. Measurements with different electrode materials (Pd, Ni, and palladinated Ni) were performed. The data were analysed according to [6] as shown in figure 2. The $E_{50\%}$-values, i.e., the potential, at which a deposition yield of 50% was observed, were +81 mV for Pd, +67 mV for palladinated Ni, and +10 mV for Ni, with uncertainties of ±50 mV.

To gain information about the deposition kinetics, the electrode potential was kept constant at -800 mV vs. an Ag/AgCl electrode and the electrolysis duration were varied. At room temperature and non-optimal stirring conditions, half of the osmium was deposited on Pd electrodes within (48±10) s and on Ni electrodes within (54±10) s.

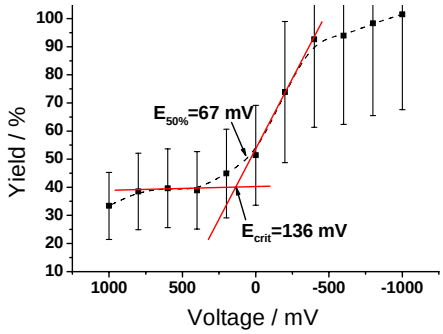

Figure 2: The critical potential on a palladinated Ni-electrode (the dashed line is drawn to guide the eye).

In another experiment with higher beam energy α-decaying ^{172,173}Os were produced and deposited on the Pd electrodes. Due to the small α-branch and the short half-lives (~20 s), only a few counts were detected. It could be shown, however, that it is possible to detect such short-lived nuclides by α spectrometry with an automated electrolytic cell after preseparation with TASCA.

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