

## **Extracts of the 2005 Annual Report of the (former) JGU "Institute of Nuclear Chemistry"**

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## TASCA Target Group Status Report

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The **T**rans-**A**ctinide **S**eparator and **C**hemistry **A**pparatus **TASCA** is currently installed at cave X8 and is dedicated to investigate the chemical and physical properties of the heaviest elements. An overview of the current status is given in a separate report [1].

For the production of Rf and Db isotopes, cold fusion reactions can be applied using Ti-beams with Pb- or Bi targets, respectively. Low melting metals can be substituted by compounds for application in high intensity beams [2]. However, to produce longer-lived isotopes of neutron-rich heavier actinides and transactinides hot fusion reactions with actinide targets are required. Here, possible target materials range from Th up to Cm or heavier elements.

We combined the different methods and capabilities of the four target laboratories involved to solve the challenging task of developing appropriate backings and targets. For first tests, we kept the geometry of the existing target wheel of the nuclear chemistry experiments at GSI unchanged. As we plan to install thinner backings for TASCA – so far 15 to 20  $\mu\text{m}$  thick Be foils were used – it is necessary to fix the backing on an adequate frame, as can be seen in Figure 1.

Al backings and Ti backings, supplied from different companies with different qualities, were produced by cold rolling. C backings were produced by resistance heating and applied without annealing. After two test beam times with  $^{12}\text{C}$  beam (500 pA) and  $^{40}\text{Ar}$  beam (800 pA), we now focus on 3  $\mu\text{m}$  Ti or 35  $\mu\text{g}/\text{cm}^2$  annealed C, alternatively, which will be tested with thermally evaporated  $\text{UF}_4$ , a target material in the near future.

As the target laboratory at GSI is specialised in evaporation, sputtering, and rolling of stable elements up to natural uranium, the foils discussed above were prepared there. The other three laboratories have the expertise of handling highly radioactive materials including the chemical purification of target material and the recovery of rare target material from used targets. After the decision for the appropriate backing has been made, backing foils will be given to the target laboratory at the LMU, where radioactive targets (Ra, Ac, Th, U, Pu, Cm) can be prepared by microevaporation. The group at the TUM offers the analytical capabilities to measure concentra-

tions of actinide elements in solution (also Pu, Cm). For the nuclear chemistry experiments at GSI the radioactive actinide targets are prepared by electrochemical deposition at UMZ.

The search for optimal conditions for the electrochemical deposition of U (as uranyl nitrate) from organic solution by molecular plating on thin Al- and Ti-backing foils is currently going on at LBNL and UMZ [3]. Figure 1 shows a picture of a 265  $\mu\text{g}/\text{cm}^2$   $^{238}\text{U}$ -layer produced by molecular plating on a 25  $\mu\text{m}$  thick Al-backing [3]. Target performance – when exposed to an ion beam – has not been tested yet. At UMZ a new electrochemical cell for improved deposition yield and uniformity of the target layer is under construction based on the design and the experience obtained at RIKEN [4]. Furthermore, optical microscopy, electron microscopy, and energy dispersive x-ray analysis are used for monitoring target thickness, chemical purity and homogeneity of the target surface. UMZ is testing a commercially available autoradiographic imager (Packard Instant Imager).

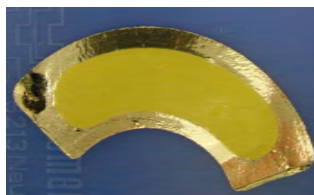


Figure 1:  $^{238}\text{U}$ -target (265  $\mu\text{g}/\text{cm}^2$ ) produced by molecular plating of uranyl-nitrate from isobutanol solution.

A new target station for TASCA is currently developed at GSI. This device is designed so that it can be used with the existing rotating actinide target wheel from GSI and with the new rotating target wheel system that is presently under development at LBNL for the Berkeley Gas-filled Separator (BGS), as well.

All contributions to the TASCA Working Group meetings and Workshops are accessible via [www-w2k.gsi.de/tasca](http://www-w2k.gsi.de/tasca).

### References

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[http://www-w2k.gsi.de/tasca05/images/contributions/TASCA05\\_Haba.pdf](http://www-w2k.gsi.de/tasca05/images/contributions/TASCA05_Haba.pdf)

# Molecular plating of uranium on thin aluminum backings\*

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Molecular plating is widely used for the deposition of lanthanide and actinide compounds on thin metallic backing foils [1]. At GSI and UMZ, this method has been previously applied on Ti or Be foils. Within the requirements of the TASCA-Project (TransActinide Separator and Chemistry Apparatus) very thin foils of Al (2-10µm) as backing material have been proposed [2].

The general usability of very thin Al was investigated by a test series with Gd and U. For these tests, plating conditions were adopted unchanged from previous practices [3]. Although Al foils of less than 10µm in thickness are difficult to manage, they show no significant disadvantages compared to Ti or Be with molecular plating.

2-Propanol (isopropanol) as organic solvent has previously been used for the preparation of actinide targets on Ti and Be backings. But on Al, the stability of the U layer is not acceptable. The surface of most of the targets looked scaly or cracked associated with irreproducible plating yields. In order to solve these problems, an alternative solvent, 2-Methyl-1-Propanol (isobutanol) was tested. Table 1 compares the plating condition of the previously used isopropanol and the newly tested conditions applying isobutanol.

Table 1: Plating conditions for isopropanol and isobutanol (U on Al-Backing)

| Solvent      | Voltage [V]       | Current [mA] | Plating time [h] | Thickn. [µg/cm <sup>2</sup> ] |
|--------------|-------------------|--------------|------------------|-------------------------------|
| Iso-propanol | 100-1200 stepwise | 0.5 -3.0     | 1                | 390 <sup>#</sup>              |
| Iso-butanol  | 150 V             | 0.01-0.5     | 4 - 5            | 650 <sup>##</sup>             |

The hygroscopic character of isopropanol might have been the reason for the poor surface quality since the plating solution as always been exposed to air for a couple of hours prior to the plating. The content of water may cause the relative high current and, thus, it possibly effects the formation of obstructing hydroxides at the surface of the backing foil. Isobutanol, which is also used by the target laboratory of the Lawrence Berkeley National Laboratory, LBNL, is less polar and therefore poorly water-soluble [4]. During numerous experiments, the plating conditions were adjusted to a lower voltage resulting in a longer plating time as shown in Table 1. With isobutanol, the coating looks more homogenous without visible cracks.

That has been approved with 25µm as well as 10µm Al backings. Fig 1 shows a light optical micrograph of an U-target, as usually prepared for the existing rotating actinide target wheel assembly at GSI.

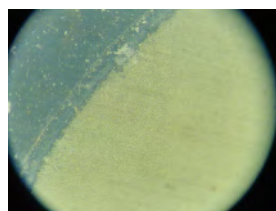


Fig 1: Uranium on Al Backing (25µm), plated from isobutanol within 3 h, Voltage 150 V

Furthermore, for U, a time dependence of the depositions yield has been determined. So far, a maximum yield at about 60 - 65 % is reached at a plating time of 5 h, as shown in Fig 2. In future investigations, the work will also focus on an increase of deposition yield to 75 % or more as achieved with isopropanol.

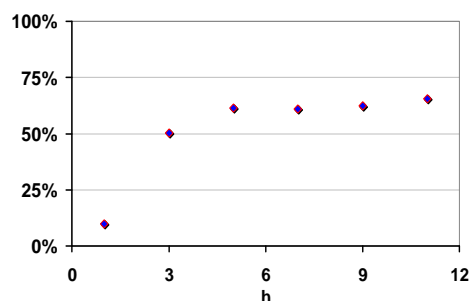


Fig 2: Time dependence of deposition yield at molecular plating of U on 25µm Al backing (from isobutanol with 150 V)

Next we plan to apply these conditions to other elements, especially various lanthanides. Also, the use of autoradiography [5] and REM/EDX will be further investigated to monitor target thickness, homogeneity and surface layer composition, respectively.

## References

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\* F+E Vertrag Mz/JVK Entwicklung und Herstellung von Transuran- u. sonstigen radioaktiven Targets

# corresponds to 74% at an intake of 500µg U /cm<sup>2</sup>

## corresponds to 65% at an intake of 1000µg U/cm<sup>2</sup>

# Autoradiographic Imaging of Uranium Targets

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Molecular plating is commonly used to prepare lanthanide and actinide targets. Subsequently to the plating procedure, the deposition yield is determined indirectly by analyzing the supernatant solution [1]. Alternatively, autoradiographic imaging offers the possibility to investigate the target surface itself, in order to obtain information about target thickness and homogeneity of the deposition layer. Autoradiographic imaging is a mapping of the radioactivity of a material, which is distributed over a certain area. The equipment used here is an InstantImager<sup>®</sup> by Packard Instrument Company with a rectangular detection area of 20 x 24 cm.

## Determination of imager resolution

The spatial resolution given by the manufacturer is 0.5 mm for C-14 ( $E_{\beta} = 0.2$  MeV) [2]. In previous tests, a resolution of about 1.5mm has been determined for Au-198 ( $E_{\beta} = 1.0$  MeV,  $E_{\gamma} = 412$  keV). A gold wire with a diameter of 25  $\mu$ m was neutron activated and subsequently analyzed by the Imager<sup>®</sup>. For uranium, the resolution has been appointed recently to  $\geq 2$  mm. Here, 2 mm-spots of deposited uranyl nitrate were arranged regularly on a Plexiglas carrier with a defined distance of 4mm between each other, see Figure 1. After neutron activation, the images of the uranium spots almost merge into each other despite of their distance.

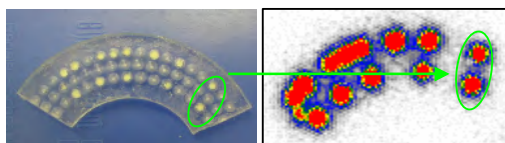


Figure 1:  
Sample carrier with uranyl nitrate and its autoradiographic image after irradiation. The framed uranyl nitrate spots have a distance of 4mm between each other.

## Imaging of UF<sub>4</sub> - Targets

In Figures 2 and 3, the autoradiographic images of two UF<sub>4</sub>-Targets are shown. These targets, which were made by vapor deposition at GSI, have a thickness of 350  $\mu$ g U/cm<sup>2</sup> on a carbon backing with an total area of 31,7cm<sup>2</sup>. They are additionally covered by a 10  $\mu$ g/cm<sup>2</sup> carbon layer. The lower target (B) has already been irradiated by a Ca-48 beam at GSI, the upper one (A) has not been irradiated. To obtain comparable results, both targets were measured simultaneously for 18h. Generally,

from these images, one can conclude that the target material is distributed over the whole area with a maximum in the centerline. The irradiated target shows a loss in total counts of about 10% compared to the non-irradiated one. To judge if this loss is due to the interaction with the ion beam [3], one has to check for possible shifts in the detection efficiency over the entire detection area. Corresponding studies with uranium targets (UF<sub>4</sub> on Al, also made at GSI by vapor deposition) used as standard samples are currently performed.

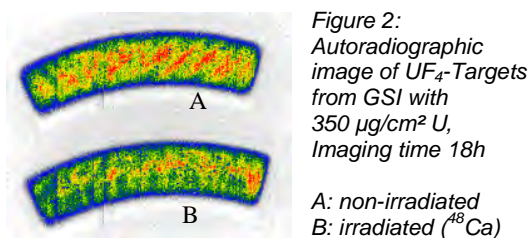


Figure 2:  
Autoradiographic image of UF<sub>4</sub>-Targets from GSI with 350  $\mu$ g/cm<sup>2</sup> U, Imaging time 18h

A: non-irradiated  
B: irradiated (<sup>48</sup>Ca)

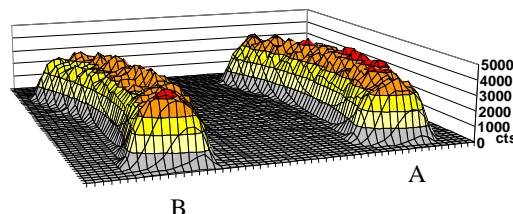


Figure 3:  
3-D diagram of the Targets from Figure 2  
Target area is 31.7cm<sup>2</sup>.  
A (non-irradiated): 1,557,371 tot. gross cts.  
B (irradiated by <sup>48</sup>Ca): 1,472,885 tot. gross cts.

Basically, autoradiographic imaging with the InstantImager<sup>®</sup> is helpful to obtain general information about target surface by using uranium as target material. For quantitative information about target homogeneity and thickness a higher resolution is preferable.

## References

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## Elektrodeposition von Pt als Homologem von Darmstadtium

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Am Beschleuniger erzeugte, kurzlebige Pb-Isotope konnten erfolgreich elektrochemisch auf ein palladiertes Nickelband abgeschieden und anschließend  $\alpha$ -spektrometrisch nachgewiesen werden [1]. Im Rahmen der Chemie der schwersten Elemente ist neben Pb (Homologes von Element 114) auch das Studium von Pt (Homologes von Ds, Element 110) interessant.

Am TRIGA-Reaktor Mainz wurde  $^{197}\text{Pt}$  durch Neutronenaktivierung von metallischem Pt hergestellt. Das Pt wurde in Königswasser gelöst und so verdünnt, dass eine Lösung der Konzentration  $5 \cdot 10^{-6} \text{ M}$  in 0,1 M HCl resultierte. Mit je 1 ml dieser Lösung wurden Abscheidungsversuche an Ni-Elektroden bei einem Potential von -500 mV (vs. Ag/AgCl) durchgeführt.

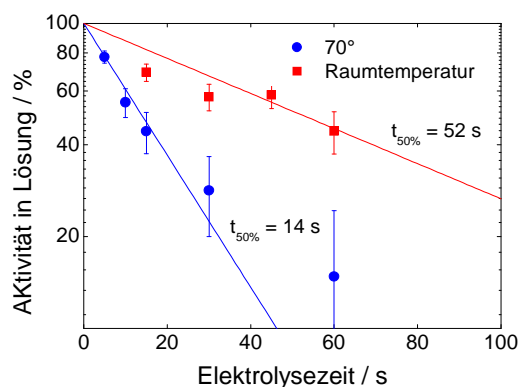


Abb. 1: Geschwindigkeit der Abscheidung von  $^{197}\text{Pt}$  an Ni aus 0,1 M HCl bei einem Potential von -500 mV (vs. Ag/AgCl)

Abb. 1 zeigt die noch in Lösung verbleibende Aktivität als Funktion der Elektrolysedauer. Der  $t_{50\%}$ -Wert gibt die Zeit bis zur Abscheidung der Hälfte der vorhandenen Aktivität an, er beträgt bei Raumtemperatur 52 s und bei 70 °C 14 s.

Bei einer Strahlzeit am UNILAC-Beschleuniger der GSI wurden in der Reaktion von  $^{40}\text{Ar}$  mit natürlichem Nd die kurzlebigen Pt-Isotope  $^{178}\text{Pt}$  ( $t_{1/2} = 21 \text{ s}$ ),  $^{179}\text{Pt}$  ( $t_{1/2} = 21 \text{ s}$ ) und  $^{180}\text{Pt}$  ( $t_{1/2} = 52 \text{ s}$ ) hergestellt. Das  $\alpha$ -Spektrum ist in Abb. 2 dargestellt. Die  $\alpha$ -Energien von  $^{179}\text{Pt}$  (5,17 MeV) und  $^{180}\text{Pt}$  (5,16 MeV) liegen sehr nahe beieinander und können nicht aufgelöst werden. In einem bereits zur Untersuchung von Pb-Isotopen verwendeten Experimentaufbau [2] wurden die Isotope aus der Gasphase über das ALOHA-System in die wässrige Phase (0,1 M HCl) überführt und mit

einer HPLC-Pumpe zur Elektrolysezelle transferiert. Unter kontinuierlicher Aktivitätszufuhr wurde eine 3-minütige Elektrodeposition bei - 500 mV und 70 °C auf Ni-Elektroden durchgeführt. In dieser Zeit stellte sich für  $^{178}\text{Pt}$  ein radioaktives Gleichgewicht ein.

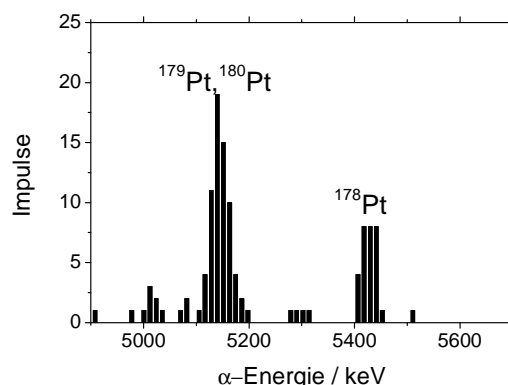


Abb. 2:  $\alpha$ -Spektrum der über die Kernreaktion von  $^{40}\text{Ar}$  mit natürlichem Nd erzeugten Pt-Isotope

Aus der auf den Elektroden abgeschiedenen Aktivität und einem 100 %-Wert der Aktivität, der durch Sammeln des Gasjets auf ein Polycarbonatfilter mit 0,1  $\mu\text{m}$  Porenweite erhalten wurde, ergibt sich eine maximal abscheidbare Aktivität  $A_{\text{max}}$ . Dieser Wert hängt nur von der Halbwertszeit bzw. Zerfallskonstante  $\lambda$  des abgeschiedenen Isotops und der Geschwindigkeit der Abscheidung ab. Mit bekanntem  $\lambda$  und dem gemessenen Wert für  $A_{\text{max}}$  kann  $t_{50\%}$  über

$$t_{50\%} = \frac{\ln 2}{\frac{\lambda}{1-A_{\text{max}}} - \lambda}$$

berechnet werden. Für einen maximal abscheidbaren Anteil von  $42 \pm 8 \%$  für  $^{178}\text{Pt}$  ergibt sich ein  $t_{50\%}$ -Wert von  $29,1^{+12,4}_{-8,3} \text{ s}$ . Da im Vergleich zum zuerst beschriebenen Experiment ein zusätzliches Volumen von ca. 200  $\mu\text{l}$  für die Überführung der Aktivität in die Elektrolysezelle nötig ist, ist  $t_{50\%}$  hier höher.

Die Ergebnisse zeigen, dass elektrochemische Methoden auch für die Untersuchung von Ds geeignet sein könnten.

### Literatur

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# Electrochemical Deposition of Short-lived Pb Isotopes on a Pd Coated Ni Tape

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The coupling of the ALOHA system to an electrolytic cell was successfully used to transfer Pb recoil nuclei from a  $^{219}\text{Rn}$  emanation source into the liquid phase with subsequent electrodeposition on Pd electrodes [1].

So far, the transport of the electrodes from the electrolytic cell to the  $\alpha$ -detector was performed manually. However, in an experiment with superheavy elements, an automated transport is necessary, preferably by using a metal tape as the electrode. For the deposition of Pb as the homolog of element 114, the electrode material of choice is Pd [1], but the production of several meters of Pd tape is too expensive. As a cheap and withstanding material, Ni tape is available, but the deposition of Pb on Ni was found to be slow and reversible. To combine the advantages of both materials, Pd was deposited onto a Ni tape (2 cm x 500 cm x 100  $\mu\text{m}$ ), which was wound onto a PE holder to ensure complete contact with the solution. The deposition was carried out in 1.5 L of 0.1 M HCl containing 50 mg/L Pd at -1000 mV vs. Ag/AgCl for 15 min. As counter electrode, a Pt gauze was used.

An electrolytic cell for electrodeposition experiments with tape shaped electrodes was constructed. The metal tape was sealed with two FEP coated o-rings to prevent the liquid from entering the detector system and to ensure a low frictional resistance for a smooth tape transport. A high volume magnetic stir bar was used for agitating the electrolyte. The  $\alpha$ -detector system consisted of three PIN diodes, with (1 x 2) cm active surface, each. The detectors were mounted at 5 cm distance in an Al-case and were kept under He (see Fig.1).

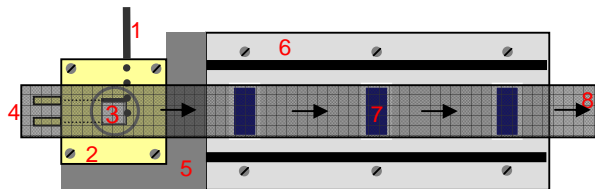


Fig. 1: Schematic of the electrolytic cell with the  $\alpha$ -detection system for electrodeposition and detection of accelerator produced short lived isotopes. 1 Ag/AgCl reference electrode, 2 electrolytic cell with o-rings, 3 high volume magnetic stir bar, 4 electrolyte in- and outlet, 5 spacer, 6 Al-case for detector system, 7 (1 x 2) cm PIN diodes, 8 Pd coated Ni-tape, arrows: stepping direction

The  $\alpha$  emitting  $^{188}\text{Pb}$  ( $t_{1/2} = 25.5$  s) was produced in the reaction  $^{152}\text{Gd} (^{40}\text{Ar}, 4n)$ . A  $^{152}\text{Gd}$  (30% enrichment) target with a thickness of 300  $\mu\text{g}/\text{cm}^2$  on 15  $\mu\text{m}$  Be backing was used. The recoil nuclei were swept out of the recoil chamber by a He/KCl cluster gas-jet at a flow rate of 1.5 L/min and transported through a PE capillary to the ALOHA device. To determine the overall deposition

yield for  $^{188}\text{Pb}$ , the activity was impacted for 3 min on a Kel-F disc. At this collection time, the activity of  $^{188}\text{Pb}$  was nearly in radioactive equilibrium.

After impactation, the disc was switched to the dissolution position and the activity was transferred to the electrolytic cell by continuous cyclic pumping of the electrolyte (0.1 M HCl,  $T = 65^\circ\text{C}$ ).

The rest potential of the Ni(Pd) tape was -250 mV vs. Ag/AgCl. At this potential, Pb deposits spontaneously, so no external potential had to be applied.

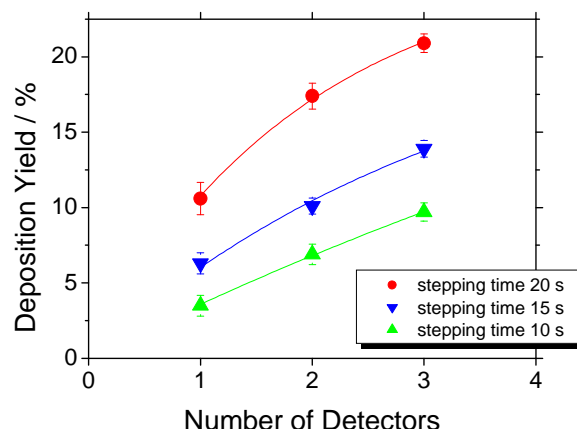


Fig. 2: Deposition yield vs. the number of detectors for various stepping times of the Pd coated Ni tape

The tape was stepped in intervals of 10, 15, and 20 s for several minutes. The deposition yield was calculated by comparing the count rates for each detector with a direct catch on a 0.1  $\mu\text{m}$  nucleopore filter. A maximum deposition yield of 21 % was achieved for a stepping time of 20 s (see Fig. 2).

In this set-up, only three detectors were used. Especially for fast stepping times a big amount of activity was stepped out of the detection system. In a superheavy element experiment, up to 20  $\alpha$  detectors will be available. The yield may be increased significantly by using a pump system with a lower dead volume (now 250  $\mu\text{l}$ ) and a higher electrode surface. The latter can be achieved by using two tapes.

After improving the overall yield, the proposed set-up may be successfully used in aqueous chemistry with element 114. Experiments with homologs suggest, that also the electrochemical deposition of element 108 - 111, 113, 115, and 116 should be possible.

## References

- [1] Hummrich et al., *GSI Scientific Report 2004*, p. 189

# Separation of Tc from fission products with the extraction system MicroSISAK

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MicroSISAK is a miniaturized apparatus developed for performing fast and continuous liquid-liquid separations on a  $\mu\text{l}$ -scale, e.g., for chemical studies of the heaviest elements or for process studies with new types of ligands that are only available in very small amounts. The main components are: (i) an inlet with inter-digital channels for intense mixing of the phases [1]. The mixture is subsequently fed into (ii) a filter unit with a teflon membrane (0.5  $\mu\text{m}$  pore size) for phase separation [2]. The inner volume of these units is in the order a few  $\text{mm}^3$ . All parts are made of Ti and mounted into a Ti-housing. Previously, this set-up has been applied for off-line extractions of Gd, Hf, and Tc [2].

Recently, it could be demonstrated that Tc can be separated from a fission product mixture [3]. For this, a  $^{235}\text{U}$ -target covered with a 15  $\mu\text{m}$  thin Al-foil has been installed in one of the beam ports of the research reactor TRIGA Mainz. Using a gas-jet system, the fission products, attached to KCl-aerosol particles, were transported to the Automated Liquid Online Heavy Element Apparatus ALOHA. Here, the aerosol particles are deposited on a Ta-disc. After a certain collection time the deposit is dissolved with 0.1 M  $\text{H}_2\text{SO}_4$  containing 0.05 M  $\text{KBrO}_3$  and subsequently pumped into MicroSISAK (see figure 1). In the mixer unit the aqueous phase is contacted with  $10^{-4}$  M tetraphenylarsonium-chloride (TPAC) dissolved in  $\text{CHCl}_3$  for selective separation of Tc(VII).

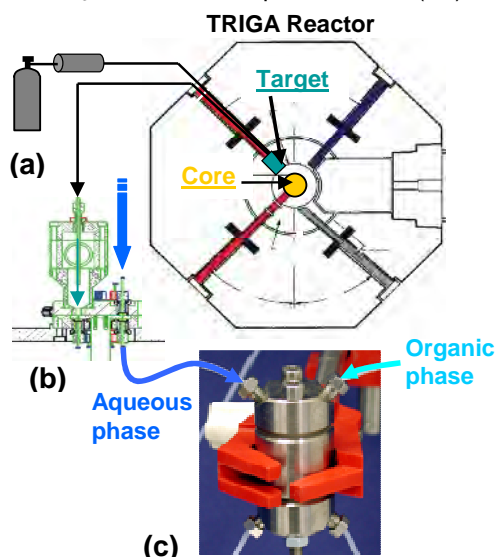


Figure 1: Set-up for the separation of Tc from fission products: (a) Gas-jet system. (b) ALOHA, (c) MicroSISAK housing (see text for details). The arrows indicate the inlet of the two phases.

Aliquots of the outgoing phases were collected and the extraction yield for Tc was concluded from a simultaneous measurement of the 18  $\text{min-}^{104}\text{Tc}$ -activity (358 keV, 531 keV) in both phases with two detectors. Two different types of mixing units have been used (see table 1).

Table 1: Configuration of the mixer units as used for the separation of Tc with MicroSISAK. Shown on the right a microscopic picture of a mixer unit (type A).

| Type | Channels | Width            |
|------|----------|------------------|
| A    | 2x15     | 30 $\mu\text{m}$ |
| B    | 2+1      | 30 $\mu\text{m}$ |

Figure 2 comprises the results for the extraction of Tc for different flow rates of the aqueous and the organic phases, respectively. For efficient extraction a surplus of organic phase improves the extraction yield significantly (blue curve/mixer B). With increasing total flow rates phase separation becomes more and more inefficient and thus the extraction yield is decreasing (red curve/mixer A). The relatively low extraction yield of the batch experiments indicate incomplete formation of Tc(VII) in the aqueous phase.

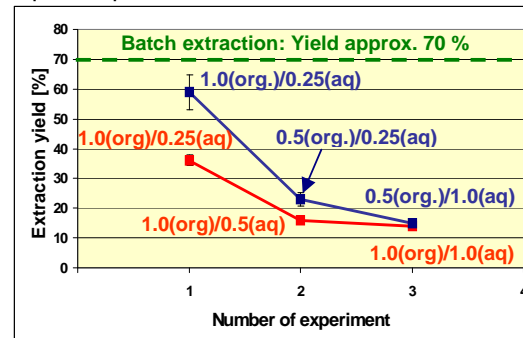


Figure 2: Extraction of Tc(VII) into TPAC/ $\text{CHCl}_3$  with MicroSISAK. The red curve corresponds to configuration A, the blue one to mixer type B. The numbers indicate the flow rate [ml/min] of the two phases (org/aq).

A new version of MicroSISAK with up to 3 subsequent separation units is currently under development at the Institut für Mikrotechnik, Mainz.

## References

- [1] W. Ehrfeld et al., Microreactors, Wiley-VCH Weinheim (2000).
- [2] K. Eberhardt et al., Institut für Kernchemie Annual Report, A5 (2004).
- [3] see: [www.safe.uio.no/ESFCTE05/Talks/ESFCTE05\\_Eberhardt.ppt](http://www.safe.uio.no/ESFCTE05/Talks/ESFCTE05_Eberhardt.ppt)