

Preparation of targets for the gas-filled recoil separator TASCA by electrochemical deposition and design of the TASCA target wheel assembly

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Abstract

The Transactinide Separator and Chemistry Apparatus (TASCA) is a recoil separator with maximized transmission designed for performing advanced chemical studies as well as nuclear reaction and structure investigations of the transactinide elements ($Z > 103$) on a one-atom-at-a-time basis. TASCA will provide a very clean transactinide fraction with negligible contamination of lighter elements from nuclear side reactions in the target.

For TASCA a new target chamber was designed and built at GSI including the rotating target wheel assembly ARTESIA for beam intensities up to 2 μA (particle). For the production of longer-lived isotopes of neutron-rich heavier actinide and transactinide elements, hot fusion reactions with actinide targets are required. Here, possible target materials range from thorium up to curium or even heavier elements.

For the deposition of lanthanide and actinide elements on thin aluminum and titanium backings by means of Molecular Plating (MP), a new deposition cell has been constructed that allows precise temperature control of the organic solvent and stirring of the solution. The electrode geometry ensures homogeneity of the electric field inside the cell. With the new set-up, holmium and gadolinium layers (500 $\mu\text{g}/\text{cm}^2$) on 2–5 μm thin titanium backings have been produced with deposition yields of the order of 90%. Systematic investigations are under way to further optimize the deposition conditions for other lanthanide and actinide elements including uranium and plutonium on different backing materials.

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1. Introduction

Today, we know 111 IUPAC approved chemical elements and have reports about six more elements not yet approved by the IUPAC [1]. The chemical and physical properties of the heaviest known elements (the so-called transactinide elements with $Z > 103$) are a matter of

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current research interest [2]. Experiments on the chemical behavior of rutherfordium (Rf, $Z = 104$) and dubnium (Db, $Z = 105$) showed that they are well placed into groups 4 and 5 of the periodic table. Furthermore, chemical investigations have been performed with seaborgium (Sg, $Z = 106$), bohrium (Bh, $Z = 107$), hassium (Hs, $Z = 108$) and, very recently, with element 112 [3–6]. However, many of the transactinide elements—also called Super Heavy Elements (SHE)—are not yet investigated sufficiently to justify their position in the periodic table, and it became obvious that simple extrapolations of the chemical properties within one group of elements in the periodic table can be misleading.

For the production of neutron-rich isotopes of SHE with half-lives of a few seconds up to about 1 m, as required for chemical studies, hot fusion reactions of actinide target nuclei such as ^{238}U , $^{242-244}\text{Pu}$, ^{248}Cm or ^{249}Cf with light ion beams like ^{18}O , ^{22}Ne , ^{26}Mg or ^{48}Ca are applied. Typical production rates range from a few atoms per minute for Rf down to about 1 atom per day in the case of Hs [2,3]. In cases when α -decay is the most prominent decay mode for the heaviest elements, this provides unique nuclide identification from time-correlated mother–daughter decay chains leading to known nuclei. However, due to their low production rates, chemical separations of SHE are performed on a one-atom-at-a-time scale and thus by-products from nuclear side reactions can seriously interfere with the detection of the SHE even in chemically isolated fractions. In order to overcome this obstacle, chemical techniques are combined with a physical pre-separation of the nuclear reaction products using gas-filled recoil separators like the Berkeley Gas-filled Separator (BGS) [7] or, more recently, the recoil separator TransActinide Separator and Chemistry Apparatus (TASCA) at GSI [8]. The pre-separator provides a very clean transactinide

fraction with negligible contamination of lighter elements from unwanted nuclear reactions in the target.

2. The gas-filled recoil separator TASCA

The TASCA was recently built at beam line X8 of the Universal Linear Accelerator (UNILAC) at GSI [8]. TASCA is specially designed for maximum transmission of SHE produced in hot fusion reactions using actinide targets in order to produce neutron-rich nuclides with half-lives long enough for chemical separations. In addition, a rich nuclear reaction and nuclear structure research program is envisaged with TASCA again focusing on reactions with actinide targets.

A schematic view of the TASCA set-up is shown in Fig. 1. The central part of TASCA is a gas-filled separator, partially making use of existing components from the former NASE [9] separator, namely one dipole-(D) and two quadrupole magnets (Q). From ion-optical calculations based on the model fusion reaction of 5–6 MeV/u ^{48}Ca on 0.5 mg/cm² actinide targets (^{238}U , ^{244}Pu) the DQ_hQ_v configuration was selected as the best choice [10]. This configuration allows two operation modes. The DQ_hQ_v mode gives the highest possible transmission, whereas the smallest image size results from the DQ_vQ_h mode, where the indices indicate horizontally (h) and vertically (v) focusing quadrupoles.

One of TASCA's main foreseen applications is its use as a physical pre-separator for chemical studies [11]. In this technique, Evaporation Residues (EVRs) are separated from the heavy-ion (HI) beam and from a significant fraction of unwanted by-products of the nuclear reaction used to synthesize the element of interest. EVRs are extracted from the separator through a thin window [12,13]. This separates TASCA's low-pressure region

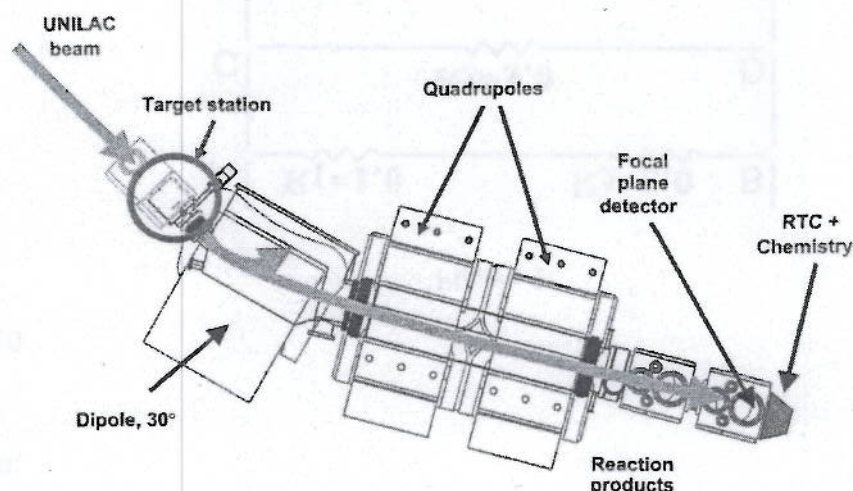


Fig. 1. Schematic view of the TASCA set-up with target station, dipole and quadrupole magnets, and recoil transfer chamber (RTC) for connecting TASCA to a chemistry apparatus. Also shown is the incoming primary beam as delivered from the UNILAC and the flight path of the separated reaction products inside TASCA.

(≤ 1 mbar He) from the Recoil Transfer Chamber (RTC) [13] in which EVR are thermalized at about ambient pressure and made available for transport to a chemistry setup (see Fig. 1). This approach promises significant progress in the chemical investigation of the heaviest elements, as new techniques and new chemical compound classes will presumably become accessible for experimental studies [13].

The TASCA separator has now entered the commissioning phase and a series of successful test experiments have been conducted with TASCA. This includes the first detection of α -emitting francium isotopes in the focal plane detector. Here, ^{211}Fr was produced in the reaction of a ^{30}Si -beam with a tantalum target.

3. The rotating target wheel assembly ARTESIA

For TASCA, a new target station has been built that accommodates: (i) the rotating actinide target wheel in an easily removable cassette, (ii) the newly built drive, (iii) collimators, and (iv) beam diagnostics components. Fig. 2 shows a cross-sectional view of the new target station in the intersection between the high vacuum beam line and the gas filled separator. A beam current transformer upstream of the target—not shown here—allows for continuously monitoring the beam current. To make use of the highest presently available beam intensities at the UNILAC, a windowless differential pumping section was installed (see Fig. 2). The ARTESIA (A Rotating Target Wheel for Experiments with Superheavy-Element Isotopes at GSI

using Actinides as Target Material) target wheel is mounted in a special box together with the target wheel drive and a fiber optics for rotation speed control. Thus, the rotating target is confined in a nearly closed container in order to protect the beam line and the separator against contamination in the case that a radioactive target gets destroyed. Fig. 3 shows the open cassette with the ARTESIA target wheel consisting of three banana-shaped segments. Also shown is the fiber optics for speed control in order to synchronize the target wheel with the pulsed UNILAC beam with 25% duty cycle and 5 ms macro-pulse length. At a rotation speed of 2000 rpm a turn of 120° —corresponding to one segment including frame intersections—takes about 10 ms. Thus, with the current UNILAC duty cycle the irradiation sequence alternates such that after segment 1, first segment 3 is irradiated and subsequently segment 2, and so forth (sequence 1–3–2–1–...).

4. Target and backing materials for TASCA

The transactinoid elements ($Z > 100$) are produced by HI-induced nuclear fusion reactions at accelerators. Depending on the availability of suitable ion beams and target materials, so-called “cold fusion” and “hot fusion” reactions can be distinguished. In cold fusion reactions, medium-heavy particles such as ^{50}Ti , ^{58}Fe or ^{62}Ni fuse with ^{208}Pb - or ^{209}Bi -targets at the lowest possible beam energy. This type of reactions has been used for the first production of the elements 107 up to 112 (see Refs. [2,3] and references

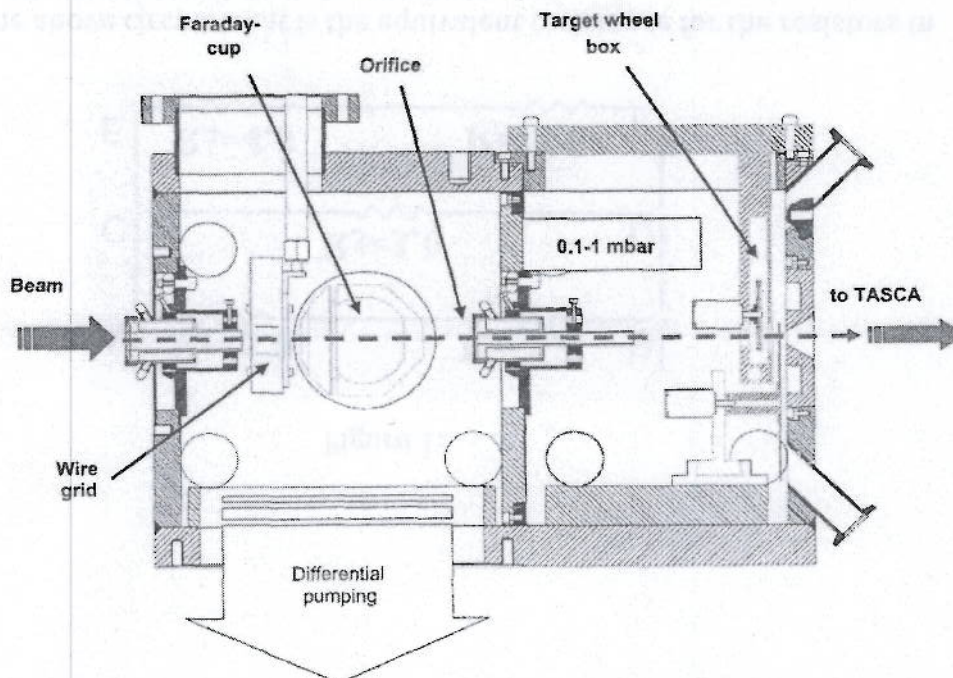


Fig. 2. Cross-sectional view of the target station at TASCA including a grid for beam diagnosis, a movable Faraday-cup and the target-wheel box. By means of differential pumping, a pressure of 0.1–1 mbar in TASCA is achieved.

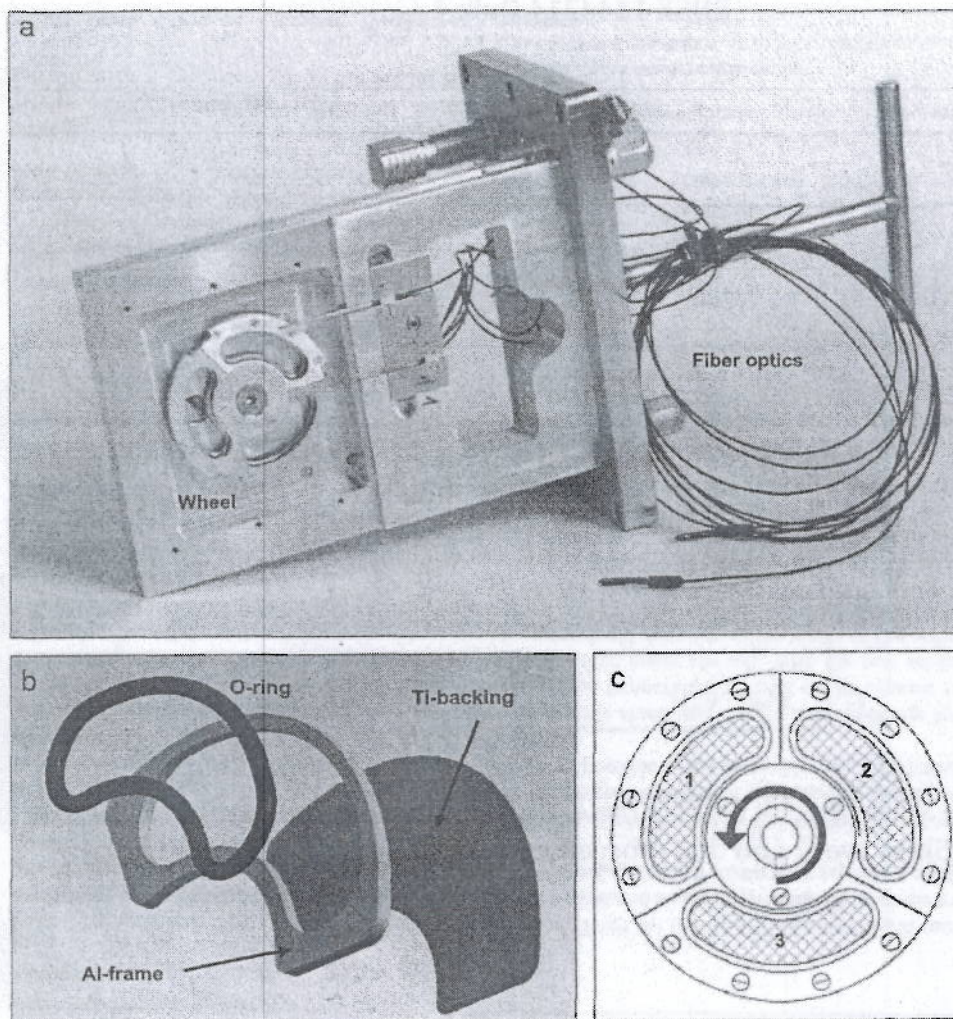


Fig. 3. (a) Open target wheel cassette for TASCA with target wheel assembly ARTESIA. (b) Schematic view of one of the banana-shaped target segments with 2 μm titanium backing, Al-frame, and O-ring sealing. (c) Three segments form one complete target wheel.

herein). However, for chemical investigations, the half-lives of the isotopes as produced in cold fusion reactions are in many cases too short. More neutron-rich and relatively long-lived isotopes of the SHE are produced when actinide targets like ^{232}Th , ^{238}U , ^{242}Pu or ^{248}Cm are used with beams of lighter ions such as ^{18}O , ^{22}Ne or ^{26}Mg . For chemistry studies with TASCA, the following prerequisites must be taken into consideration for choosing a proper target–projectile combination:

- the recoil energy of the EVR must be high enough to pass through the window between the separator and the RTC,
- the availability of the required target material might be limited,
- for chemical studies the half-life of the nuclide under investigation should exceed one second, and
- reactions with high cross-section are clearly preferred.

Table 1 comprises some possible production reactions for SHE applicable with TASCA. In some cases, rare and expensive target materials like ^{244}Pu , ^{248}Cm or ^{249}Bk must be irradiated.

The choice of an appropriate backing material is a crucial point in many HI-reaction studies. Very often, a low- Z material like C ($Z = 6$), Al ($Z = 13$) or Ti ($Z = 22$) is used to prevent the production of nuclides whose decay interferes with the unambiguous identification of single SHE atoms, such as α -particle emitting or spontaneously fissioning ones.

For the same reason, Pd ($Z = 46$) instead of Pt ($Z = 78$) has been employed as the anode material to prevent contamination of the target with a high- Z material. The use of Pt would result in the formation of, e.g., α -decaying Po isotopes, which can hinder the detection of the transactinide element under investigation [14].

Al-backings and Ti-backings in different thickness are produced by cold rolling, whereas deposition by resistance

Table 1

Production reactions for chemical studies of the transactinide elements with TASCA. The half-lives of the Ds-isotopes are predicted to be in the range of seconds

Cold fusion reactions	$T_{1/2}$ (s)	Hot fusion reactions	$T_{1/2}$ (s)
$^{208}\text{Pb}(^{50}\text{Ti},1\text{n})^{257}\text{Rf}$	5.5	$^{244}\text{Pu}(^{22}\text{Ne},5\text{n})^{261}\text{Rf}$	78
$^{209}\text{Bi}(^{50}\text{Ti},1\text{n})^{258}\text{Db}$	4.3	$^{249}\text{Bk}(^{18}\text{O},5\text{n})^{262}\text{Db}$	34
		$^{248}\text{Cm}(^{22}\text{Ne},5\text{n})^{265}\text{Sg}$	7.4
		$^{244}\text{Pu}(^{27}\text{Al},4\text{n})^{267}\text{Bh}$	17
		$^{248}\text{Cm}(^{26}\text{Mg},5/4\text{n})^{269/270}\text{Hs}$	14/22
		$^{232}\text{Th}(^{48}\text{Ca},5/4/3\text{n})^{275-277}\text{Ds}$	Few s ^a
		$^{238}\text{U}(^{48}\text{Ca},3\text{n})^{283}112$	3.8
		$^{244}\text{Pu}(^{48}\text{Ca},3\text{n})^{289}114$	2.7

^a Predicted; see: G. Audi, et al., Nucl. Phys. A729, 3 (2003).

heating is applied for carbon backings [15]. For experiments where the target is placed in a chemically reactive atmosphere containing oxygen, which is a prerequisite for particular gas phase chemistry experiments with SHE [16], beryllium (Be) has to be employed as backing material. Be-foils with thicknesses of 10–15 μm are commercially available from various suppliers.

For the preparation of lanthanide and actinide targets, different methods are applied, e.g., deposition by Physical Vapor Deposition (PVD), by Magnetron Sputtering (MS) and Electrochemical Deposition (ED). Irradiation test with high intensity ^{12}C - and ^{26}Mg -beams on uranium-tetrafluoride (UF_4) and metallic uranium targets on various backings have already been performed and are described in a separate contribution in this issue [15]. So far, 2 μm thin Ti-foils as a backing material proved to be superior to Al and C with respect to mechanical stability and in-beam performance with U as the deposit.

5. Preparation of targets by electrochemical deposition

Because several actinide isotopes like ^{244}Pu and ^{248}Cm are 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 pre-requisite. ED is well suited for the preparation of lanthanide and actinide targets on metallic and non-metallic backing materials with deposition yields approaching 100% [14,17–30]. Here, the deposition of lanthanide elements, U and Cm, respectively, on thin Al- and Ti-backings from organic solutions is described. This technique is referred to as Molecular Plating (MP) [14,25–30].

For MP, the lanthanide or actinide compound, typically the nitrate is dissolved in a small volume (5–10 μl) of nitric acid and the aqueous phase is mixed with a surplus of an organic solvent ($\approx 14\text{ ml}$), usually isopropanol or isobutanol. Under these conditions, no electrolytic dissociation of the organic solvent occurs by applying an electric current and probably the originally dissolved nitrate compound transforms into the oxide during the deposition process [30]. Fig. 4a shows a schematic view of the cell used for molecular plating, as proposed by Haba et al. [31]. Fig. 4b is a photo of the complete set-up with two water-cooled

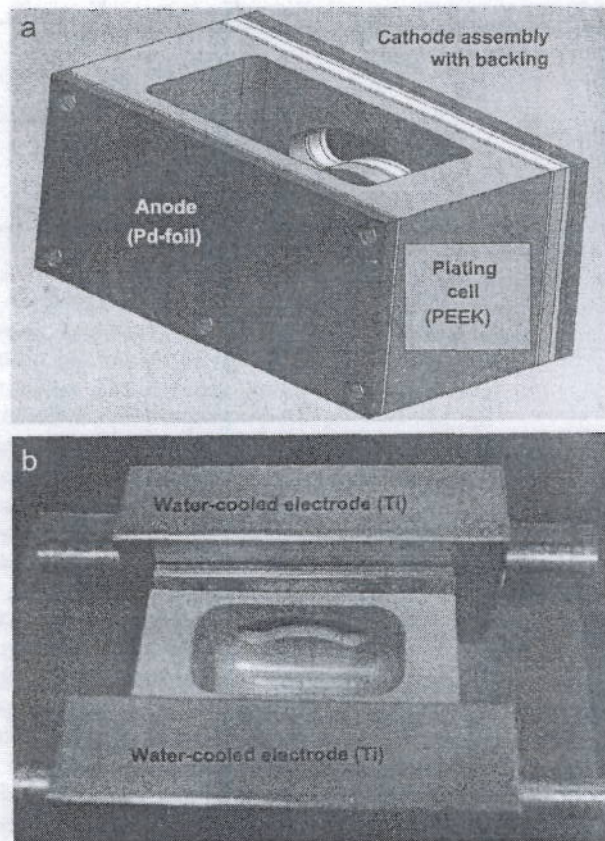


Fig. 4. Electrochemical cell used for the production of lanthanide and actinide targets by molecular plating. (a) Shows a schematic drawing of the cell made of PEEK with cathode assembly and Pd-anode foil. (b) Shows a picture of the set-up with attached water-cooled electrode blocks made of titanium.

Ti-blocks attached to the electrodes in order to keep their temperature at a constant value during deposition. A cell made of polyetheretherketone (PEEK) confines the area to be plated and acts as a container for the alcohol solution with a volume of about 16 ml. The backing foil with an active target area of 1.74 cm^2 is glued to an Al-frame and pre-mounted onto the Ti-cathode. A sealing plate made of

silicon and a cover plate made of plexiglas completes the cathode-assembly. The cathode is then screwed to the PEEK cell. A Pd-foil with a thickness of 50 μm serves as anode. The cathode assembly and the anode are water cooled in order to keep the temperature at a constant value during the deposition process. Furthermore, the solution is stirred by means of a magnetic stirrer operated at 1000 rpm.

Molecular plating is carried out by applying a voltage of 150 V, yielding a current density up to 1.5 mA/cm² under the described conditions. Normally, the deposition is performed on self-supporting foils with thicknesses ranging from 2 up to 10 μm . The foils should be pinhole-free and pre-cleaned with acetone, dilute nitric acid, water, and isopropanol or isobutanol prior to use. To avoid any cross contamination, for each actinide isotope a separate deposition cell is used. The new design offers several advantages compared to the previously used cell [32]. In particular, it allows precise temperature control of the electrodes and the organic solvent as well as stirring of the solution during the deposition process. The electrode geometry ensures homogeneity of the electric field inside the cell. With the new set-up, systematic investigations with

Table 2

Experimental conditions for the deposition of Sm-, Gd-, Ho-, and U-layers from isobutanol solution

Isotope	Backing	Voltage (V)	Plating time (h)	Amount (μg) ^a	Target density ($\mu\text{g}/\text{cm}^2$)
Sm (nat)	Al	150	4	2000	970
Sm (nat)	Al	150	4	1500	743
Sm (nat)	Ti	150	4	1500	814
Sm (nat)	Al	150	4	1000	501
Sm (nat)	Ti	150	4	1000	546
Sm (nat)	Al	150	4	500	265
Sm (nat)	Ti	150	4	500	280
Gd (nat)	Ti	150	5	1000	548
Ho (nat)	Al	150	5	2000	655
Ho (nat)	Ti	150	5	2000	625
Ho (nat)	Ti	150	5	1000	558
Sm (nat)/U-238	Ti	150	5	500/500	238/215

The target covers a total area of 1.74 cm².

^aListed here is the initially dissolved amount of the desired element in isobutanol.

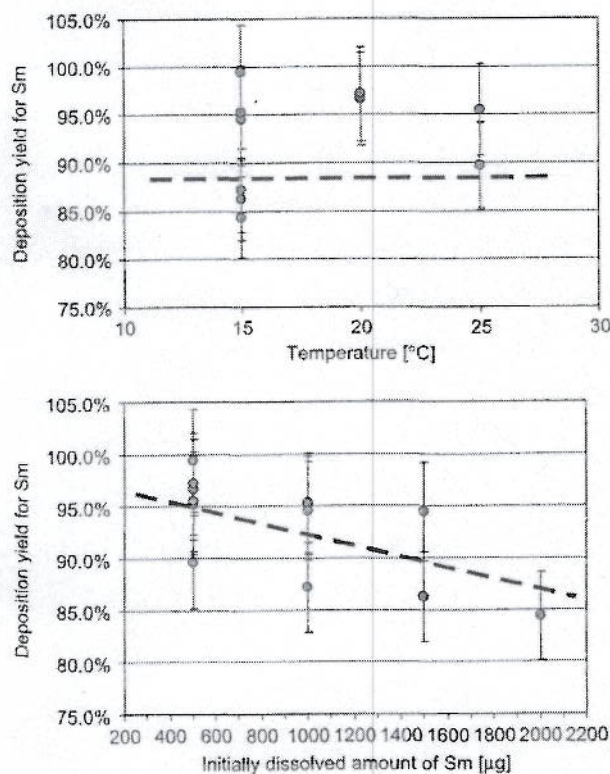


Fig. 5. Dependence of the deposition yield for Sm on the temperature of the organic solvent during the deposition process (upper diagram) and on the amount of initially dissolved Sm (lower diagram). The dashed lines are to guide the eye and do not result from any data fitting procedure. The diagrams comprise data as obtained with both Al- as well as Ti-backing foils.

Ho, Sm, and U have been performed in order to optimize the deposition process with respect to deposition yield and homogeneity of the deposited layer. For this, a total of 31 targets have been produced under different experimental conditions on Al- and Ti-backings. The deposition yield has been determined by neutron activation analysis of the supernatant solution subsequent to deposition [30]. Fig. 5 comprises the data as obtained for the deposition of Sm with variation of the temperature of the solvent and—in a separate investigation—the amount of initially dissolved material. As can be seen, deposition yields approaching 100% have been achieved at room temperature at a target thickness of about 500 $\mu\text{g}/\text{cm}^2$. Furthermore, Gd- and U-targets with thicknesses up to 500 $\mu\text{g}/\text{cm}^2$ on 2 μm thin Ti-backings have been produced with deposition yields of the order of 90%. These targets have already been used in the TASCA commissioning experiments. Table 2 comprises the experimental parameters used for molecular plating of Sm-, Gd-, Ho-, and U-layers on Al- and Ti-backings. Systematic investigations are under way to further optimize the deposition conditions for other lanthanide and actinide elements, in particular Pu, on thin Ti-backings.

6. Summary

The new gas-filled recoil separator TASCA at GSI is specially designed for advanced chemical investigations of the transactinide elements ($Z > 103$). For TASCA, a new target chamber was designed and built at GSI, which accommodates the rotating actinide target wheel assembly ARTESIA in an easily removable protective housing. Thin foils (2 μm) made of titanium or beryllium may serve as backing materials. Neutron-rich heavier actinide and transactinide elements with half-lives of at least a few seconds, as required for chemical studies, can only be

produced in hot fusion reactions with actinide targets. For this, layers of Th, U, Pu, or Cm with a thickness of about $500 \mu\text{g}/\text{cm}^2$ are suitable targets. At the University of Mainz, MP is applied for the deposition of lanthanide and actinide elements on thin ($2 \mu\text{m}$) Ti-backings. For this, an electrochemical cell is used, that ensures homogeneity of the electric field inside the cell and furthermore allows precise temperature control of the organic solvent as well as stirring of the solution. In systematic investigations with Sm, it could be demonstrated that deposition yields exceeding 90% are possible at a target areal density of $500 \mu\text{g}/\text{cm}^2$. Several targets made of Gd and U have also been produced for the TASCA commissioning experiments. Systematic investigations are currently being carried out to optimize the deposition conditions for Pu. In the near future, experiments with a ^{244}Pu -target wheel at TASCA are planned.

Acknowledgments

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