Extracts of the 2012 Annual Report of the (former) JGU "Institute of Nuclear Chemistry"

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is missing!

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The Superheavy Element Search Campaigns at TASCA

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Successful experiments on the synthesis of elements with Z=114-118 in ⁴⁸Ca-induced reactions with actinide targets were first performed at the DGFRS in Dubna [1]. Results for Z=114 (Fl) and Z=116 (Lv) nuclei have been later on confirmed by other groups [2-4].

Using doubly-magic ⁴⁸Ca for the synthesis of yet heavier elements is not possible due to the lack of sufficient amounts of target materials for elements beyond Cf. Thus, several attempts to produce element 120 in reactions with projectiles beyond ⁴⁸Ca have been carried out at DGFRS and SHIP [5-7]. The separator TASCA and its detection systems were significantly upgraded since the experiment on ^{288,289}Fl [3] was performed [8]. In the past two years, two experiments on the synthesis of elements beyond Z=118 have been undertaken at TASCA using the reactions $^{50}\text{Ti} + ^{249}\text{Bk} \rightarrow ^{299}119^*$ and $^{50}\text{Ti} + ^{249}\text{Cf} \rightarrow ^{299}120^*$. To verify the performance of the setup, element 117 was also synthesized.

The first attempt to form element 120 at TASCA was performed in August-October 2011. The search for element 119 was performed in two series from April to September 2012. The beam energies from the UNILAC, average initial target thicknesses (*d*) [9], and accumulated beam doses for each reaction are given in Table 1. These

values are **preliminary**. Beam doses were deduced from beam current measurements in front of the target. 85% of the beam doses were estimated to be on the target.

Subsequent to the months-long experiments on elements 119 and 120, an experiment on the synthesis of element 117 in reaction 48 Ca + 249 Bk \rightarrow 297 117* was successfully performed. In about one month of experiment time, the Bk target was bombarded by 48 Ca ions at three different beam energies. The final data analyses of all these experiments are currently ongoing.

We are grateful for support by the GSI directorate, ion source, accelerator, and experiment electronics staff.

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Table. 1. The parameters of the experiments. For details, see text

Beam	Target	CN	Date	Elab (MeV)	$d (\mu g/cm^2)$	Beam dose	Beam dose on target
⁵⁰ Ti	²⁴⁹ Cf	²⁹⁹ 120	25.08-12.10.2011	306	515	$1.1 \cdot 10^{19}$	$0.9 \cdot 10^{19}$
	. ²⁴⁹ Bk	²⁹⁹ 119	13.04-03.07.2012	300	440	4.2·10 ¹⁹	3.6·10 ¹⁹
			23.07-03.09.2012				
⁴⁸ Ca		²⁹⁷ 117	26.09-09.10.2012	270		$0.6 \cdot 10^{19}$	$0.5 \cdot 10^{19}$
			09.10-22.10.2012	274		$0.5 \cdot 10^{19}$	$0.4 \cdot 10^{19}$
			22.10-29.10.2012	268		$0.3 \cdot 10^{19}$	$0.2 \cdot 10^{19}$

Superheavy Element Flerovium is the Heaviest Volatile Metal

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Electron shells of superheavy elements (SHE), i.e., elements with atomic numbers $Z \geq 104$, are influenced by strong relativistic effects caused by the high value of Z. Early atomic calculations for element 112 (copernicium, Cn) and element 114 (flerovium, Fl) predicted them to have closed and quasi-closed electron shell configurations, respectively, and to be noble gas-like due to very strong relativistic effects [1]. Recent fully relativistic calculations studying Cn and Fl in different environments suggest them to be less reactive compared to their lighter homologues in the groups, but still exhibiting metallic character (see, e.g., [2]). Experimental gas-chromatography studies on Cn have, indeed, revealed a metal-metal bond formation with gold [3]. In contrast to this, for Fl, the formation of a weak physisorption bond with gold was inferred from first experiments [4].

A gas chromatography experiment on Fl at TASCA was conducted subsequently to the study of the reaction ²⁴⁴Pu(⁴⁸Ca; 3,4n)^{289,288}Fl [5]. The coupling of chemistry setups to a recoil separator promises extremely high sensitivity due to strong suppression of background from unwanted species. TASCA was operated in the Small Image Mode, focusing the products into a Recoil Transfer Chamber (RTC) of 29 cm³, from where they were flushed within 0.8 s to a detection setup (Fig. 1). Two COMPACT detectors [6] connected in series were used; each detector consisted of 32 pairs of 1x1 cm² PIN diodes covered with a 35 nm gold layer. The first detector was connected directly to the RTC exit and kept at room temperature.



Fig.1: First COMPACT with the attached RTC.

A negative temperature gradient from +20 to -162 °C (Fig. 2, panel a) was applied in the second detector channel placed downstream to the first one. The use of two detectors in series allowed the detection of species in a wide volatility range – from the non-volatile Pb, the nearest homolog of Fl in the group, to the noble gas Rn. Two decay chains, one

from ²⁸⁸Fl and one ²⁸⁹Fl were detected. Both decays from Fl isotopes occurred in the first detector channel at room temperature. The positions of decay chain members are shown in Fig. 2 (e) together with the Monte Carlo simulated deposition peak for ²⁸⁵Cn (dashed line). Distributions of Pb, Hg, and Rn (Fig. 2, b-d) are also shown for comparison. The observed behavior of Fl in the chromatography column is indicative of Fl being less reactive than Pb. The evaluated lower limit of the adsorption enthalpy $-\Delta H_{ads}^{Au} > 48kJ/mol$ reveals formation of a metal-metal bond with Au, which is at least as strong as that of Cn, and thus demonstrates the metallic character of Fl.

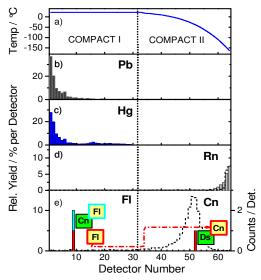


Fig. 2: The observed gas-chromatography behavior of Fl and Cn in COMPACT compared to those of Pb, Hg and Rn.

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Carbonyl Complex Formation of Short-Lived Ir and Re Isotopes*

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We recently reported on the in-situ carbonyl complex formation of short-lived group 6 and group 8 elements [1]. These elements are well known to form mononuclear, volatile carbonyl complexes. However, no mononuclear, binary complexes of group 7 and 9 elements are known from literature (see e.g. [2]). We studied the transport of short-lived Ir and Re isotopes in a He-CO atmosphere, which gives a hint at the formation of volatile carbonyl complexes of these elements.

In first experiments at the TRIGA reactor in Mainz, the transport of fission products of the neutron induced fission of ²⁴⁹Cf in a CO containing gas-stream was studied. The transported isotopes were collected on a charcoal filter and measured with a y detector. Volatile fission products as well as short-lived isotopes of the refractory elements Mo, Tc, Ru and Rh were identified. However, precursor effects prevent a clear assignment to the transported element. To get a better understanding, the homologs Re and Ir were produced in ²⁴Mg-induced fusion reactions with natEu and natTb targets at the gas-filled separator TASCA. The recoiling isotopes were separated from the primary beam and transfer products in TASCA and thermalized in the recoil transfer chamber in a He-CO atmosphere. The volatile species were transported in the gas stream through a 10-m long capillary to a charcoal filter which was monitored by a γ detector. This way the transport of $^{178-179}$ Ir and $^{170-172/172m}$ Re was observed. Both, Re and Ir, apparently form volatile complexes with CO under these experimental conditions.

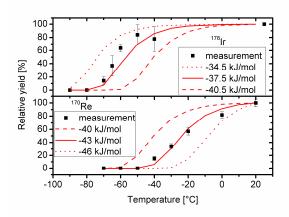


Figure 1: Transport yield of Re and Ir depending on the temperature of the isothermal chromatography column.

The lines show results of Monte Carlo Simulations.

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In further investigations the adsorption and decomposition of these complexes were studied. In the adsorptionstudies on a quartz surface, the gas-stream was guided through an isothermal quartz column. The temperature was varied from measurement to measurement and the transport yield through the column was determined for each temperature. These measurements were modeled with Monte Carlo Simulations, which yield values for the adsorption enthalpy of these complexes on the quartz surface. For the Re complexes the adsorption enthalpy was determined to be (-43 ± 3) kJ/mol, and for the Ir ones (-37.5 ± 3.0) kJ/mol. Figure 1 shows the breakthrough curves and the results of the Monte Carlo Simulations. The thermal stabilities of these compounds were studied by passing the gas stream through a quartz wool plug heated by an oven before reaching the charcoal filter. The transport yields were determined for various temperatures. (see Figure 2) The complexes start to decompose at temperatures around 300°C.

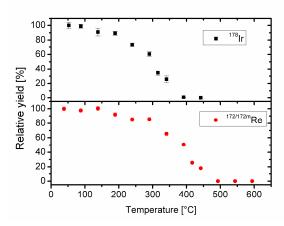


Figure 2: Transport yield of Re and Ir vs. the temperature of the quartz wool plug in the decomposition setup.

The experimental observation of the transport of Re and Ir in a CO-containing gas stream gives a hint on the formation of volatile complexes in CO. Further studies are planned to elucidate the nature of these complexes, which might open the door for bohrium and meitnerium chemistry experiments.

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Chemistry for Isobar Separation behind SHIP*

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Recoil separators are powerful instruments for the isoltation of desired nuclear reaction products. However, separation of ions of similar masses and isobaric nuclides is not possible at such a device without additional separation stages. One possibility for a second separation step is provided by chemistry. Two recoil separators at GSI are dedicated to superheavy element research – the velocity filter SHIP and the gas-filled separator TASCA. At gasfilled separators like TASCA, the coupling with chemistry setups is established[1]. In contrast, no vacuum separator has been used as a preseparator for chemical investigations so far. We have demonstrated that SHIP also can be combined with chemistry setups. Recently the in-situ formation of volatile metal carbonyl complexes was studied at TASCA [2,3]. Recoiling W, Re, Os, and Ir isotopes were thermalized in a CO containing atmosphere and formed volatile complexes. These complexes were transported in a gas jet over several meters to detection setups.

Short-lived Ta isotopes as well as Re and W isotopes were produced in the complete fusion reaction of $^{48}\mathrm{Ca}$ projectiles with a $^{133}\mathrm{Cs}^{127}\mathrm{I}$ target at SHIP, in order to investigate the behaviour of Ta under comparable conditions. The Recoil Transfer Chamber (RTC), which was originally built for experiments at TASCA in the small image mode, was attached to SHIP. The chamber was separated from the high vacuum of SHIP by a 5.8- μ m thick Mylar window supported by a honeycomb grid. The size of the RTC window was 3 cm \times 4 cm. The chamber was cylindrical with an inner diameter of 3 cm. The depth of the RTC was 3 cm.

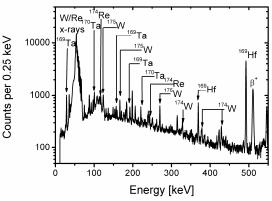


Figure 1: γ-spectrum of fusion products stopped in Alfoil behind SHIP.

In the first part of the experiment, an aluminium catcher foil was placed 5 mm behind the RTC window, where the recoiling ions were collected for 30 min. Within 2 min the * Work supported by the Helmholtz Institute Mainz, and the BMBF under contract No. 06MZ7165I, *j.even@gsi.de

foil was taken out of the RTC, placed in front of a γ detector, and measured for 10 min. Figure 1 shows a typical spectrum. γ-lines of ^{170,169}Ta produced in the reaction ¹²⁷I(⁴⁸Ca, 5-6n) and ¹⁷⁴Re produced in the reaction ¹³³Cs(⁴⁸Ca,7n) were observed. Furthermore ^{174,175}W and ^{169,170}Hf were identified. These are either produced in the (48Ca,p xn)- reactions, or are decay products of the Re and Ta isotopes. In the second part of the experiment the catcher foils were removed, and the RTC was flushed with either a He/CO mixture or pure CO. The gases were purified by passing oxysorb and hydrosorb cartidges. The pressure in the RTC was kept at 600 hPa. The recoiling ions were thermalized in the RTC and all volatile compounds were transported in the gas stream out of the RTC through a 566 cm long capillary to a filter of activated charcoal. The volatile compounds adsorbed on the charcoal filter, which was monitored with the y detector. Figure 2 shows a spectrum of the charcoal trap. Only W and Re isotopes were identified. No Ta and Hf isotopes were observed in the spectra.

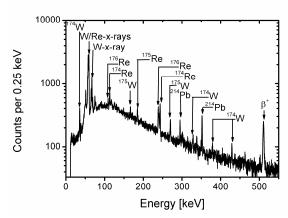


Figure 2: γ-spectrum of the charcoal filter. Pb γ-lines originate from the natural background.

This confirms former results that W and Re form volatile complexes in a CO atmosphere. However, Ta and Hf were not transported so that the formation of volatile complexes with CO was excluded. Gas phase carbonyl chemistry is therefore an appropriate tool to separate group 4 and 5 elements from group 6 and 7 elements and can thus be used for isobar separation.

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Ion Beam Cooling and Trapping at TRIGA-SPEC

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The TRIGA-SPEC setup is the prototype for mass spectrometry and laser spectroscopy at the low-energy branch at FAIR (MATS and LASPEC [1]). At the TRIGA Mainz research reactor this setup serves for the determination of nuclear ground state properties of neutron-rich fission products. Studies in the medium-mass region will yield information to benchmark mass models, nuclear structure calculations and investigate so far unexplored regions towards the r-process path of nuclear synthesis [2].

The TRIGA reactor provides a continuous neutron flux of 10^{11} n/(s·cm²) to induce nuclear fission in a 300 µg 235 U target. The target chamber is flushed with a nitrogen buffer gas and aerosol particles of potassium chloride. The fission products, adsorbed to the aerosol particles, are transported into a surface ion source that is held on high-voltage potential. Temperatures of about 2000° C lead to a release and ionization of the fission products. The ions are extracted against ground potential at 28 keV and are subsequently mass separated in a dipole magnet. A radio-frequency quadrupole RFQ cooler-buncher accumulates the ions in an axial potential well where they are cooled by collisions with the buffer gas (helium) and released as bunches to either the mass spectrometry TRIGATRAP or laser spectroscopy branch TRIGA-LASER.

In 2012, an ion beam was transported through all components of the TRIGA-TRAP and the TRIGA-LASER beam lines for the first time. For the mass spectrometry branch, a 50 pA ⁸⁵Rb⁺ ion beam was produced in the ion source, cooled and bunched in the RFQ. The transmission efficiency was estimated in the continuous-mode to about 25%. A pulsed drift tube decelerated the bunches to prepare them for injection into the double Penning trap mass spectrometer. The ⁸⁵Rb⁺ ions were purified in the preparation trap and then injected into the precision trap. A (time-of-flight) cyclotron resonance spectrum was recorded as depicted in Fig. 1.

For TRIGA-LASER, a ⁴⁰Ca⁺ ion beam was cooled. The RFQ was operated in the continuous-mode at a buffer gas pressure of about 10⁻⁴ mbar. The transport efficiency was measured using a Faraday cup at the end of the beam line to be about 15%. The very first observation of a

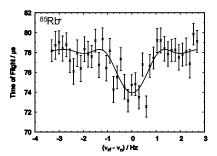


Figure 1: Time-of-flight ion cyclotron resonance spectrum of $^{85}Rb^{\scriptscriptstyle +}$ as a function of the irradiated radio frequency ν_{rf} .

resonance fluorescence spectrum as shown in Fig. 2 was made by collinearly superimposing the Ca beam with laser light of 397 nm. The Doppler-shifted laser frequency was tuned into resonance with the D1 transition applying an additional acceleration voltage to the optical detection region. Further investigations are now ongoing.

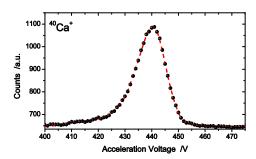


Figure 2: Resonance spectrum of the $2s_{1/2} \rightarrow 2p_{1/2}$ transition in $^{40}\text{Ca}^+$. The dashed line shows a multiple Voigt fit to take the asymmetry into account.

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Preparation of ²⁴⁹Bk targets

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In the year 2012 two long experiments to synthesize the elements 119 and 117 have been performed at the gas-filled recoil separator TASCA at GSI. In both experiments ²⁴⁹Bk was used as the target material, which was bombarded with ⁴⁸Ca for the synthesis of element 117 and ⁵⁰Ti to search for the new element 119.

For this, a ²⁴⁹Bk target-wheel was used [1]. 12.7 mg ²⁴⁹Bk in form of the nitrate were provided by Oak Ridge National Laboratory. The target segments were produced by Molecular Plating (MP) with high deposition yields at the Institute for Nuclear Chemistry at the University of Mainz [2,3].

The total amount of 249 Bk was delivered in four quartz vials covered with a Teflon septum. This septum was penetrated with a syringe containing 100 μ l 0.1 M HNO₃ to dissolve the 249 Bk nitrate prior to the MP. After complete dissolution, the solution was transferred into the electrochemical deposition cell (EDC). The quartz vial was washed with 1 ml isopropanol and this solution was also transferred to the EDC. Finally 51 ml isobutanol were added.

The EDC [3,4] is made from polyether-etherketone (PEEK). On one side of the EDC, the target backing - a thin (~2 µm) Ti-foil produced by cold rolling and glued on an Al frame at the GSI Target Laboratory - was mounted and acted as the cathode. As an anode, a Pd-foil in the same geometry as the target frame was used. The EDC was clamped between two water cooled Ti blocks. For the mixing of the $^{249}{\rm Bk}$ solution, an ultrasonic stirrer was applied. The deposition parameters for $^{249}{\rm Bk}$ were similar to those for $^{249}{\rm Cf}$ targets produced in 2011 [5], i.e., a current density of 0.3 mA/cm² was applied, resulting in voltages of 300 to 600 V. After deposition times of 3 to 4 hours target thicknesses of 354±18 – 508±25 µg/cm² with deposition yields exceeding 90 % were obtained.

The deposition yield and kinetics were determined by α -particle and γ -ray spectroscopy. Prior to the start of the deposition and then in one hour steps during the deposition, $10~\mu l$ aliquots of the supernatant solution were evaporated to dryness and measured by α -particle spectroscopy. Due to the rather short half-life of ^{249}Bk , its daughter, ^{249}Cf , is also present in the solution. Because both isotopes have α branches with significantly different α -particle energies, the deposition of both elements can be followed simultaneously in this way. The deposition kinetics of ^{249}Bk and ^{249}Cf are very similar. This allows the determination of the ^{249}Bk target thickness also via the γ -rays from ^{249}Cf , which is not possible for ^{249}Bk directly due to the absence of suitable γ -lines. For the

determination of the deposition yield by γ -ray spectroscopy, a thin ^{249}Bk target was used as reference sample. The results of the yield determination by α -particle and γ -ray spectroscopy are in good agreement with each other. The average thickness of the target on the day of production was $463\pm23~\mu\text{g/cm}^2$. The target segments were delivered to GSI and mounted on a targetwheel as shown in Fig. 1. The produced ^{249}Bk targets were able to resist a high total beam dose with beam intensities of over 4 particles microAmp for a long time.



Figure 1: ²⁴⁹Bk target-wheel

Acknowledgments:

We are very thankful to the Oak Ridge National Laboratory for providing ²⁴⁹Bk. The authors thank the Target Laboratory at GSI for providing the Ti-backing foils and the Mechanical Workshop of the Institute for Nuclear Chemistry in Mainz for the construction of the plating cells. This work was supported by the Helmholtz Institute Mainz.

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Smooth crack-free targets for nuclear applications produced by molecular plating*

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Within the TRAKULA project (**Tra**nsmutationsrelevante **k**ernphysikalische Untersuchungen langlebiger **A**ktinide) high-quality α sources are required for the precise half-life ($t_{1/2}$) measurement of the very long-lived low energy α -emitter ¹⁴⁴Nd ($t_{1/2} = (2.65 \pm 0.37) \cdot 10^{15}$ y, $E_{\alpha} = 1.85$ MeV).

Using a Nd salt, i.e., [Nd(NO₃)₃·6H₂O], as model electrolyte several constant current density molecular plating (MP) experiments were carried out to investigate the effects of parameters like the plating solvent (isopropanol and isobutanol mixed together -hereafter referred to as IP+IB- and N,N-dimethylformamide -DMF-) and the surface roughness of the deposition substrates (~13 and ~24 nm) on the quality of the produced layers. One of the deposition substrates (Ti-A, average roughness 24 \pm 7 nm) was a circular 50 μ mthick Ti foil cut from a bigger foil (Goodfellow), the other (Ti-B, average roughness 12.8 ± 0.7 nm) was produced by coating a 300-µm thick mechanicallypolished Si wafer onto which 100 nm of metallic Ti were sputtered. For the MP, 0.338 g of Nd(NO₃)₃·6H₂O were dissolved in 20 mL 0.1 M HNO₃. From the solution an aliquot of 100 µL was added to a mixture of 1 mL isopropanol (Fisher Chemical) and 34 mL isobutanol (Applichem), or to 35 mL DMF (Merck), finally yielding 0.22 mM solutions of Nd3+ in the deposition cell. The plating solutions were stirred by means of a quartz tip ultrasonic stirrer (Bandelin Sonopuls HD 2070) operated at 30% power pulse. MP experiments were always carried out for 3 h by applying a constant current of 6 mA, corresponding to a current density of 0.7 mA/cm^2 [1].

Gamma-ray spectroscopy was performed after MP of Nd solution containing radioactive ¹⁴⁷Nd tracer. The tracer was produced in a (n,γ) reaction on ^{146}Nd present in the used Nd of natural isotopic composition (nat Nd) by irradiating an aliquot of 100 µL of the stock solution with thermal neutrons in the TRIGA Mainz research reactor. A high-purity germanium detector (GEM series HPGe Detector Model No. GEM 23158 P-Plus, ORTEC Company) was used to determine the Nd deposition yield. To obtain quantitative data, reference sources with known amounts of the tracer were prepared. They consisted of filter papers with the same geometry as the targets, soaked with the tracer-containing solution. The yield values were always obtained as the average value calculated from three distinct y-ray measurements. The measurements always gave quantitative deposition yields: $(99.2 \pm 1.4)\%$ for targets produced on Ti-B using IP+IB, $(98.7 \pm 2.8)\%$ and $(99.1 \pm 2.2)\%$ for targets

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produced with DMF using Ti-A and Ti-B as deposition substrates, respectively.

The surface roughness of the inactive Nd targets was investigated by using an Atomic Force Microscope (AFM) (MFP 3D, Asylum Research) in tapping mode. Fig. 1a shows the AFM image of the Nd target produced on Ti-B using IP+IB (mean roughness 22 ± 13 nm). Figs. 1b and c show insted the images of the targets produced on Ti-A (b, mean roughness 130 ± 40 nm) and on Ti-B (c, mean roughness 18 ± 9 nm) using DMF. The smoothest deposition substrate grows the smoothest layers even if heavily cracked in the case of IP+IB. DMF is superior to IP+IB as there are fewer cracks present. They are even completely absent when the smoother Ti-B substrate is applied. Substrate roughness and plating solvent are thus key factors for the production of smooth, crack-free targets .

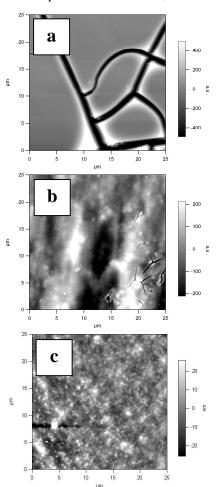


Figure 1: AFM pictures of Nd targets produced using Ti-B and IP+IB (a), Ti-A and DMF (b), and Ti-B and DMF (c).

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The heavy-ion reactions ²³⁸U+²³⁸U and ²³⁸U+²⁴⁸Cm revisited

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Recent theoretical work by Zagrebaev et al. [1,2] has renewed interest in radiochemically determined isotope distributions in reactions of 238 U projectiles with actinide targets that had previously been published only in parts [3,4]. These data have been revisited. The cross sections $\sigma(Z)$ below the uranium target have been determined as a function of incident energy in thick-target bombardments. These are compared to predictions by a diffusion model [5] whereby consistency with the experimental data is found in the energy intervals $7.65-8.30~{\rm MeV/u}$ and $6.06-7.50~{\rm MeV/u}$, see Fig. 1. In the energy interval $6.06-6.49~{\rm MeV/u}$, the experimental data are lower by a factor of 5 compared to the diffusion model prediction indicating a threshold behaviour for massive charge and mass transfer close to the barrier.

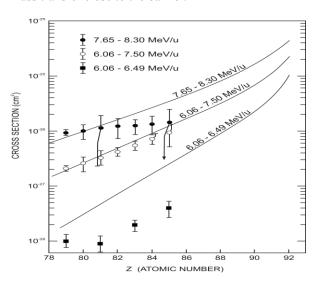


Fig. 1 Element yields P(Z) in the ²³⁸U+²³⁸U reaction in three different bins of laboratory energies defined by the incident projectile energy and the effective target thickness. For the higher energy bins and for the higher atomic numbers the cross sections are increasingly depleted by sequential fission. The solid lines are diffusion model predictions [5].

For the intermediate energy interval, the missing mass between the primary fragment masses deduced from the generalised Q_{gg} systematics including neutron pairbreaking corrections δn and the centroid of the experimental isotope distributions as a function of Z have been used to determine the average excitation energy as a function of Z. From this, the Z dependence of the average total kinetic-energy loss has been determined. This is compared to that measured in a thin-target counter experiment at 7.42 MeV/u [6]. For small charge transfers, the values of of this work are typically about 30 MeV

lower than in the thin-target experiment with the difference decreasing with increasing charge transfer developing into even slightly larger values in the thick-target experiment for the largest charge transfers. This is the expected behaviour which is also found in a comparison of the partial cross sections for quasi-elastic and deep-inelastic reactions in both experiments. The cross sections for surviving heavy actinides, e.g., 98Cf, 99Es, and 100Fm indicate that these are produced in the low-energy tails of the dissipated energy distributions, however, with a low-energy cutoff on the order of 35 MeV. A comparison of the survival probabilities

$$\prod_{i=1}^{x} \langle \Gamma_n / \Gamma_{tot} \rangle$$

of the target-like residues of equal charge and neutron transfers in the reactions of ²³⁸U projectiles with either ²³⁸U or ²⁴⁸Cm targets is consistent with this cutoff as evaporation calculations assign the surviving heavy actinides to the x=3 and/or x=4 neutron evaporation channels.

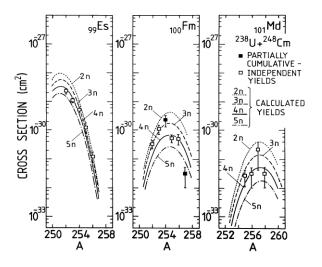


Fig. 2 Comparison of the measured (symbols [4]) and calculated (curves) for the isotope populations in the $^{238}U+^{248}Cm$ reaction at 7.4 $\,$ MeV/u

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