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TRIGA-TRAP: A new facility for high-precision mass measurements on neutron-rich fission products and actinoids

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Introduction: The nuclear mass is one of the most fundamental properties in nature since it reflects all interactions in the nucleus. Therefore, high-precision mass data has its application in many fields of physics, but especially in nuclear astrophysics and nuclear structure studies [1]. The natural abundances of the elements as we observe today can be explained by different nucleosynthesis processes. One of them is the rapid neutron capture process (r-process), which proceeds far away from the stable nuclides in the region of nuclei with neutron excess. Most of them are presently not accessible in experiments, which has triggered the planning of new radioactive beam facilities. The research reactor TRIGA Mainz with the recently installed Penning trap mass spectrometer TRIGA-TRAP [2] already now provides the possibility to extend the limit for high-precision mass measurements towards the r-process nuclides. In off-line experiments samples of actinoids from uranium to californium will be investigated as well. Recently, the masses of the three nobelium isotopes $^{252-254}$No have been measured at SHIPTRAP (GSI, Darmstadt). Masses of other nuclides above uranium are determined via alpha-decay chains but have not been measured directly. TRIGA-TRAP is besides SHIPTRAP (GSI, Darmstadt) the only facility world-wide, where direct mass measurements of transuranium elements are performed.

Status of the experiment: After initial development at the physics institute, the TRIGA-TRAP mass spectrometer has been relocated and commissioned at beam port B of the TRIGA Mainz. The cryogenic double-Penning trap system has been very precisely positioned in a 7 T superconducting magnet. Two off-line ion sources were brought into operation, which provide stable reference ions like alkalines or carbon clusters [3]. Certain actinoids can be ionized as well by one of these sources. The carbon cluster ions are used for absolute mass calibration, and to perform systematic tests of the Penning trap setup. First measurements have been carried out in the second half of 2008, where cyclotron resonances have been recorded for different cluster sizes up to C_{24}^+ (see Fig 1). Due to comparably high residual gas pressure in the measurement trap, the excitation time, and thus, the line width of the resonance presently limit the achievable relative mass uncertainty to about 10^{-7} using a conventional non-interrupted excitation scheme. By the implementation of time-separated oscillatory fields, which is known as the Ramsey technique [4], the uncertainty could be improved to about 5×10^{-8} for a single measurement sufficient for first mass measurements on actinoid elements. The feasibility of this approach has been demonstrated by performing measurements on several gadolinium isotopes and first data on $^{241}Am^{16}O^+$ ions have been recorded.

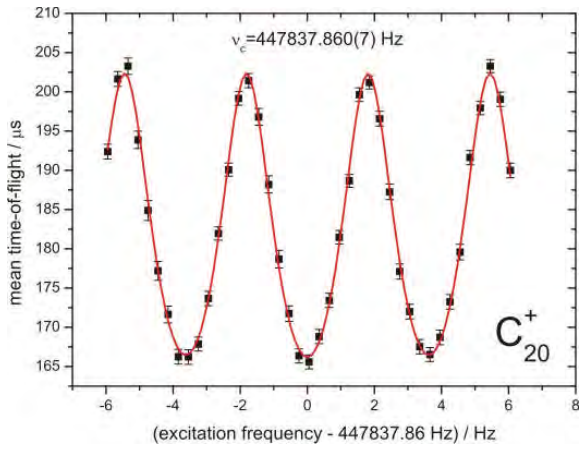

Figure 1. Excitation of C_{20}^+ carbon cluster ions with time-separated oscillatory fields including a fit of the theoretical line shape. The mass value is extracted from the centroid frequency.

Outlook: A new differential pumping-barrier assembly will be implemented in order to suppress the helium buffer gas flow between the preparation trap and measurement trap further. In addition, an ion getter pump will be installed. These improvements will enable extended observation times, thus line widths below 1 Hz could be achieved. Extensive work is undertaken to set-up an ECR source on a high-voltage platform in combination with a carbon aerosol gas jet arrangement in order to have access to fission products from the reactor [5].

We will focus on finalizing and commissioning the non-destructive ion detection system. Having two independent ion detection techniques implemented within the same Penning trap mass spectrometer, TRIGA-TRAP will be unique among the on-line Penning trap mass spectrometers for short-lived nuclides. Ultimately single-ion sensitivity will enable measurements on nuclides with extremely low production rate [6].

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Acknowledgement

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A carbon cluster ion source for absolute mass calibration at TRIGA-TRAP

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Introduction and motivation: Penning trap mass spectrometers are leading devices to determine one of the most fundamental property of a nucleus: its mass [1]. To this end, the cyclotron frequency $\nu_c = q\,B\,/\,(2\,\pi\,m)$ of a stored ion is measured, where q denotes the charge, m the mass of the ion, and B the magnetic field strength, which is obtained from a calibration with a reference ion of well-known mass. A new laser ablation ion source was developed for TRIGA-TRAP [2] to produce carbon cluster reference ions, facilitating absolute mass measurements, since carbon clusters are integer multiples of the atomic mass unit. Systematic studies of the accuracy of the mass spectrometer can be performed by carbon cluster cross-reference measurements [3], in which the well known frequency ratio of two different carbon cluster ions is determined. Furthermore, since a large variety of reference ions with well-known mass is provided, a reference ion species with a cyclotron frequency similar to the ion of interest can be selected in order to minimize systematic mass dependent uncertainties.

Experimental setup and results: Laser ablation is the common approach to generate carbon clusters for mass spectrometry purposes. At TRIGA-TRAP a pulse from a frequency-doubled Continuum Minilite Nd:YAG laser at a wavelength of λ = 532 nm, with a pulse width of τ = 5 ns, and a repetition rate below 1 Hz is guided into a vacuum chamber and focussed on a Sigradur® target with an energy density of 120 to 250 MW/cm^2. Thereby, carbon is removed from the target surface, and a plasma is formed, in which carbon clusters are created and ionized. Afterwards, they are extracted by an electrode of Pierce geometry, focussed by an Einzel lens and after a 90° deflection guided by conventional ion optics into the Penning traps. A detailed description of the ion source is found in [4, 5].

The laser ablation ion source produces an ion pulse composed of a mixture of ions of a broad mass range. In order to analyse the mass spectrum, the ions were stored in a Penning trap, where their initial energy spread is reduced by collisions with helium buffer gas. Subsequently, the cooled ions are ejected from the trap and detected with a channeltron-type electron multiplier detector. Ions of different masses are distinguished by their time of flight, as shown in figure 1. Clusters up to C_{24}^+ (mass 288 u) were successfully produced, thus the entire nuclide chart can be covered with reference ions. The desired cluster species is selected by applying a mass selective buffer-gas cooling technique in the preparation trap, so that a clean ion bunch is available for the actual measurement in the precision trap.

The same source has been used to produce a sufficient amount of gadolinium monoxide ions and americium monoxide ions for a first off-line mass measurement.

Conclusion and outlook: A laser ablation ion source for the production of carbon cluster ions was developed, tested and brought into operation at the TRIGA-TRAP facility. Reference ions for absolute mass calibration are thus available, and the stability of the source was demonstrated by 14 hours continuous operation without any maintenance. Mass measurements on lanthanoide and actinoide elements are currently in progress.

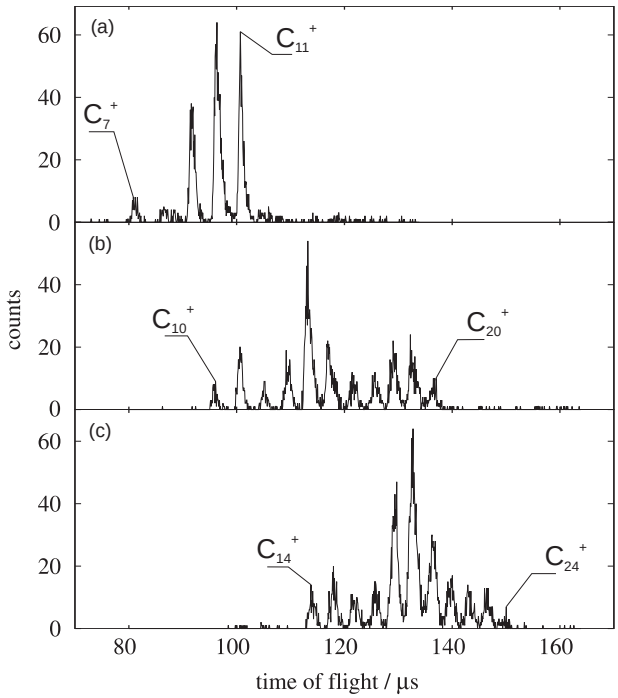

Figure 1. Time-of-flight spectra recorded after ejection of carbon clusters from the preparation Penning trap in order to identify the ion species. Different fractions of the mass spectrum produced by the ion source were sampled.

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Extraction of fission products for TRIGA-SPEC using a helium gas jet with carbon aerosols

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Introduction: High-precision measurements of nuclear ground-state properties like masses, magnetic moments, spin and charge radii reflect the interactions inside the nucleus and are therefore of fundamental importance, with applications in nuclear structure and nucleo-synthesis studies [1]. The TRIGA-SPEC experiment at the TRIGA Mainz reactor aims to investigate neutron-rich fission products and determine these properties in a model independent way by means of Penning trap mass spectrometry and laser spectroscopy [2]. A helium gas-jet arrangement in combination with a carbon aerosol generator has been implemented in order to transfer neutron-rich fission products from the reactor to the TRIGA-SPEC experiment. The gas-jet arrangement will be connected to a 30 cm^3 target chamber close to the reactor core containing a fissionable target, e.g. ^{235}U or ^{249}Cf. The thermal neutron induced fission products having energies of approximately 100 MeV thermalize in the helium gas at a pressure of about 2.5 bars. After thermalization they attach to the carbon aerosols that are added to the gas and can be flushed out of the target chamber in a laminar flow through a PE-capillary to a skimmer system where the helium gas is separated from the aerosols. In order to release the fission products from the aerosols and to ionize them, an ECR ion source is being installed on a high-voltage platform. After ionization the fission products will be mass separated in a 90° dipole magnet and finally transferred either towards the laser spectroscopy experiment or the Penning trap mass spectrometer [3].

Experimental setup and results: Carbon aerosols are produced by constantly charging a capacitor between two sharpened pure graphite electrodes placed in a helium gas flow. When the breakdown voltage of about 1.5 kV is reached, a spark discharge takes place and carbon evaporates. The repetition rate of the discharge can be easily adjusted through the charging current.
Using an electrostatic classifier we observed that the carbon vapour condenses to particles with a diameter of 10 nm to 1 µm lognormally distributed around a maximum at about 100 nm at a gas flow of 1.4 l/min. Despite the erosion of the electrodes, the continuous production of aerosols has been demonstrated over about 8 hours without maintenance, satisfying the requirements of the TRIGA-SPEC experiments.
The accessibility of radionuclides away from the valley of β−stability depends on their half-lives and production rates, thus a rapid and efficient transfer to the experiment is crucial. Thermalization in the target chamber and the transport to the ion source defines mainly the total transfer time since the ionization and the transport of ions are very fast. Hence, the transport time of the fission products has been determined by guiding the aerosols through a 7 m long capillary of 0.86 mm diameter to a filter placed in front of a γ-detector during a reactor pulse. As shown in Fig. 1 γ-radiation is emitted during the reactor pulse, which is observed by the detector. The peak shape as well as the neutron flux in the reactor can be approximated with a Gaussian distribution. After a few hundred milliseconds the fission products arrive and the activity increases first to a maximum and decreases afterwards again down to background level. The transport time is defined as the time difference between the maximum of the reactor pulse and the half of the maximum γ−activity and is determined to be about 400 ms.

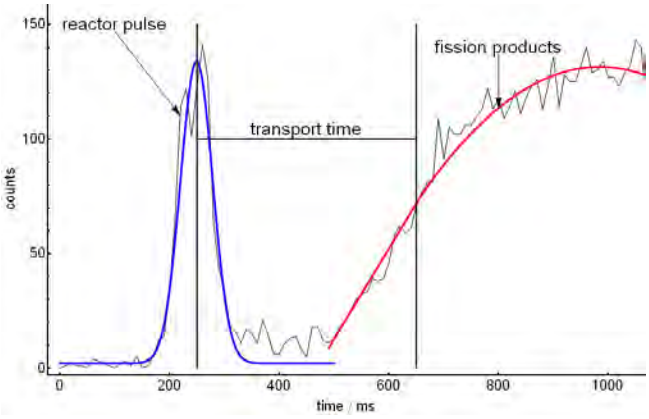

Figure 1: γ-activity recorded during and after a reactor pulse. The transport time is defined as the time difference between the maximum of the pulse and the half of the maximum activity that is caused by the arriving fission products.

Conclusions and outlook:
The extraction of fission products using carbon aerosols was demonstrated. The transport efficiency will be investigated in detail. Values up to 70% have been reached in first tests.
Next steps are to finalize the ECR ion source and connect it to the gas jet via a skimmer to study the ion production.

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[1] K. Blaum, Phys. Rep. 425 (2006) 1-78.
[2] J. Ketelaer et al., Nucl. Instr. Meth. A 594 (2008) 162-177.
[3] see reports to TRIGA-LASER and TRIGA-TRAP in this issue

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Financial support is acknowledged from the Helmholtz Association for National Research Centers (HGF) under contracts VH-NG-037 and VH-NG-148 as well as by the Stiftung Rheinland-Pfalz für Innovation under contract 854. Sz. Nagy acknowledges the support of the Alliance Program of the Helmholtz Association. We also thank the technical staff of the TRIGA Mainz reactor.

TRIGA-LASER: A collinear laser spectroscopy beamline at the TRIGA reactor Mainz

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Introduction: Collinear Laser spectroscopy of radioactive nuclei can provide fundamental information on the structure of radioactive nuclei. The dependencies of the hyperfine splitting and isotope shift on the nuclear moments and mean square nuclear charge radii are well known and the theoretical framework for the extraction of nuclear parameters is well established. We have started to set up the *TRIGA-LASER* experiment at the University Mainz. It will allow us to perform new measurements on neutron rich isotopes produced at the research reactor [1], but it will also serve as a development platform for the *LaSpec* experiment [2] at *FAIR*, *GSI*.

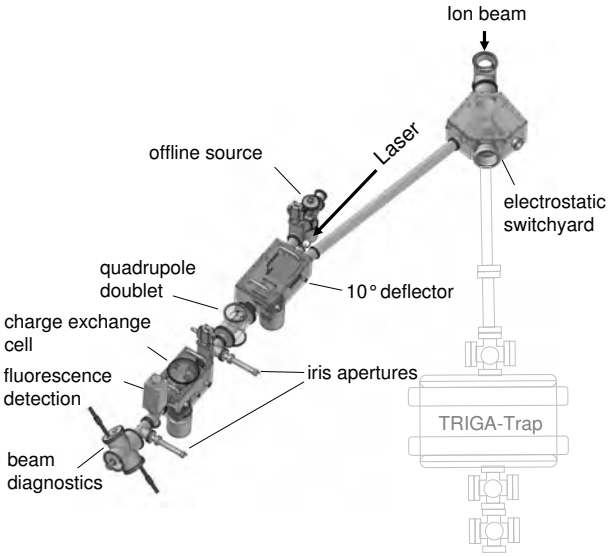

Figure 1: Drawing of the collinear beamline. All components except the switchyard and the detection unit are already installed.

Experimental: The vacuum system of the laser beamline was assembled after machining of the electrostatic deflection chamber and the chamber for the charge exchange cell according to our custom design. A vacuum pressure of 2×10^{-7} mbar could be reached without baking. In Fig. 1 a drawing of the complete laser beamline is shown. For commissioning purposes, an offline surface ion source based on an electro-thermally heated graphite oven was constructed. The source is operated in a HV cage and can be set to voltages up to 10 kV. For fast loading of the ionizer tube, the source chamber is separated from the laser vacuum system with a valve and can be pumped with a separate turbo molecular pump.

Results: First tests showed 70 % ion beam transmission with 3.5 nA current on the Faraday cup after the non-operating charge exchange cell. For ion beam profile monitoring a fork scanner system 5100 from Danfysik was installed to get a good control of the ion beam diameter, which is essential for a good overlap between the laser and ion beam. In Fig. 2 a plot of the beam profile at the end of the ion beamline in both, horizontal and vertical plane is shown.

First laser spectroscopy test measurements will be performed with Rb atoms after charge exchange. The transition at 780 nm wavelength can be excited with a diode laser which will be frequency locked to a HighFinesse WS7 wavemeter. This allows to install a compact laser system next to the beamline.

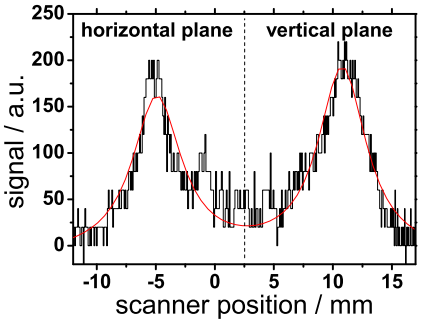

Figure 2: Beam profile of a 300 pA Li^+ beam at 2 keV energy in horizontal and vertical plane. The corresponding beam diameter was estimated to be 5 mm.

Acknowledgement

Work supported by HGF under contract VH-NG-148 and 'Stiftung Rheinland-Pfalz für Innovation' under contract 854.

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TASCA Commissioning Completed*

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The **T**rans**A**ctinide **S**eparator and **C**hemistry **A**pparatus (**TASCA**) project [1], which is focusing on the separation and investigation of neutron-rich transactinide nuclides produced in actinide-target based reactions, has successfully finished its commissioning; see [2] for an interim report. TASCA is ready for the envisioned research program which includes both chemical investigations of transactinide or superheavy elements (SHE) after preseparation with the gas-filled separator and nuclear structure and nuclear reaction studies.

The central device of TASCA is a gas-filled separator in a DQQ configuration operated either in the "High Transmission Mode" (HTM, DQ_hQ_v) or in the "Small Image Mode" (SIM, DQ_vQ_h) [1-5]. In the HTM, the unsurpassed transmission of TASCA - at a relatively low dispersion - is exploited. In contrast, the SIM provides unique possibilities due to its small spot size in the focal plane (< 3 cm diam.) at a still relatively high transmission; see Table 1.

Table 1: Important parameters of TASCA, calculated for the reaction ^{48}Ca(^{244}Pu,3n)^{289}114, in comparison with other gas-filled separators operated in SHE research.

Separator	Configuration	Transmission %	Dispersion mm/%	Bρ (max) Tm
DGFRS	DQ_hQ_v	35	7.5	3.1
GARIS	DQ_hQ_vD	40	9.7	2.16
BGS	Q_vD_hD	49-59	20	2.5
TASCA	DQ_hQ_v	60	9	2.3
TASCA	DQ_vQ_h	35	1	2.3

Table 2 provides a compilation of all nuclear reactions and reaction products applied and detected in the course of the commissioning program together with the mode TASCA was operated in and the fill gas. Also listed are experiments to test and optimize the recoil transfer chambers (RTC) [6], the gas-jet transport of preseparated products into our Rotating wheel On-line Multidetector Analyzer (ROMA), and its performance, and the coupling and use of aqueous chemistry set-ups behind TASCA.

Table 2: Nuclear reactions and their products used to commission TASCA; H=HTM, S=SIM, TSp=TASISpec, R=ROMA, C=chemistry, catch=catcher foils.

Product	xn	Beam	Target	Mode	Gas	RTC +R/C
^{30}Si	--	^{30}Si	--	H,S	Vac	
^{173,175}Os	7n	^{40}Ar	^{nat}Ce	H	He	C
$^{180-182}$Hg	2-4n	^{40}Ar	^{144}Sm	H,S	He	C
^{188}Pb	4n	^{48}Ca	^{144}Sm	H,S	He	
^{188}Pb	4n	^{40}Ar	^{152}Gd	H,S	He	
$^{194-196}$Pb	4-5n	^{40}Ar	^{nat}Gd	H,S	He	R
$^{198-199}$Bi	4-5n	^{22}Ne	^{181}Ta	H,S	He	catch
$^{195-196}$Po	4-5n	^{48}Ca	^{152}Gd,	H	He	R
^{200}At	3n	^{64}Ni	^{nat}La	TSp	He	
^{200}Fr	5n	^{64}Ni	^{141}Pr	TSp	He	
$^{205-206}$Fr	5-6n	^{30}Si	^{181}Ta	H	He	
$^{208-211}$Ra	3-4n	^{54}Cr	^{nat}Gd	H,S	He	
$^{208-211}$Ra	3-6n	^{64}Ni	^{150}Nd	TSp	He	
^{210}Ac	5n	^{40}Ar	^{nat}Lu	H,S	He,N_2	
^{215}Ac	4n	^{22}Ne	^{197}Au	H,S	He,H_2	
$^{218-x}$Th	xn	^{64}Ni	^{154}Sm	TSp	He	
$^{224-x}$U	xn	^{64}Ni	^{nat}Gd	TSp	He,H_2	
^{245}Fm	3n	^{40}Ar	^{208}Pb	H,S	He	R
^{252}No	2n	^{48}Ca	^{206}Pb	H,S, TSp	He	R
^{253}No	2n	^{48}Ca	^{207}Pb	H, TSp	He	
^{254}No	2n	^{48}Ca	^{208}Pb	H,S	He,H_2	
^{255}No	5n	^{22}Ne	^{238}U	H,S	He,H_2	R
^{256}No	4n	^{22}Ne	^{238}U	H	He	
^{260}Rf	6n	^{22}Ne	^{244}Pu	H	He,H_2	
^{261a,261b}Rf	5n	^{22}Ne	^{244}Pu	H	He	R,C
^{262}Rf	4n	^{22}Ne	^{244}Pu	H	He,H_2	

Extensive studies have been performed in the HTM and SIM to obtain optimized parameter sets for (i) the target thickness and stability, (ii) the gas pressure and the gas filling (He, H_2, and mixtures), (iii) the dipole setting (Bρ) and quadrupole focusing, (iv) the RTCs (window material and thickness, support structures, and size and shape of the chamber), (iv) gas-jet transport of pre-separated products, and (vi) the coupling and performance of devices

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like ROMA and the Automated Rapid Chemistry Apparatus (ARCA). Results of many of these parameter studies were compared with TASCA model calculations [7] and very good agreement was achieved. This agreement is of special importance as it allows for the selection of proper settings for magnetic rigidities (Bρ) in the dipole magnet and the quadrupole magnets for all nuclear reactions and for all gases and gas mixtures tested at various pressures. It is especially rewarding to see that not only Bρ values were properly chosen to centre product distributions on focal plane detectors (FPD) but also that the measured spatial distributions and, more importantly, the efficiencies were in very good agreement with model calculations. These results confidently demonstrate that we are able to perform trustworthy SHE experiments with TASCA. In the following, we mention a few concluding experiments, some of the highlights and new developments; see [2] for additional information on the parameter studies.

The first efficiency measurements with catcher foils behind the target and in the focal plane showed very good agreement with model calculations for the fairly asymmetric reaction ^{22}Ne(^{181}Ta,xn)^{198m,199}Bi [2]. To confirm this agreement in a more symmetric reaction, leading to a significantly heavier reaction product, and to obtain a standard reaction to test and check the TASCA performance, detailed studies were performed with well known reactions of ^{48}Ca with ≈ 0.5 mg/cm^2 thick targets of ^{206,207,208}Pb leading to ^{252,253,254}No. Assuming cross sections of 0.5 µb, 1.3 µb, and 2 µb [8] for the production of ^{252}No, ^{253}No, and ^{254}No, efficiencies of 54%, 56%, and 50%, respectively, were obtained for the HTM, using a He filling of 0.8 mbar, and a (80x36) mm^2 16-strip FPD. Taking into account uncertainties in cross sections and systematic errors of target thicknesses and beam current measurements, we observe an excellent agreement with model calculations [7] predicting 54%. Equally good is the agreement in the SIM, at a He pressure of 0.8 mbar, where a 30% efficiency was measured for the reaction ^{48}Ca(^{208}Pb,2n)^{254}No.

A new (140x40) mm^2 large, highly efficient FPD, consisting of double-sided silicon strip detectors (DSSSD) in the focal plane and SSSDs for the backward box detectors will further increase the TASCA efficiency; see [9] for details of the new detector.

As one of the crucial tests and one of the highlights finalizing the TASCA commissioning program, we studied the isotopes ^{260}Rf, ^{261a,261b}Rf, and ^{262}Rf synthesized in the very asymmetric reaction ^{22}Ne + ^{244}Pu; see [10] for details of the nuclear reactions, for TASCA parameters, and for the interesting nuclear decay results. In essence, the performance of TASCA was as anticipated; everything worked well, including the ^{244}Pu target wheel. Efficiencies and magnetic settings (Bρ=1.99 Tm at 0.4 mbar He, HTM) were as expected. As observed in previous experiments [2], it was again possible to reduce the background in the FPD by using a mixture of He and H_2. This part of the commissioning program showed clearly that TASCA can be applied efficiently for nuclear decay and nuclear reaction studies of neutron-rich nuclides of SHE synthesized in very asymmetric hot-fusion reactions. Rf isotopes were not only measured in the FPD but were also collected in an RTC and were transported either to ROMA for nuclear decay measurements [10] or to ARCA for chemical investigations [11].

An additional highlight of the experiment was the first transactinide chemistry behind TASCA designed as a proof-of-principle experiment. It was performed in ARCA with pre-separated 78-s ^{261a}Rf; details of the nuclear reaction and the Rf separation in TASCA are described in [10] while all chemical aspects are discussed in [11]. This successful experiment, which studied the formation of Rf-fluoride complexes and their adsorption behaviour on an anion-exchange resin, demonstrated that aqueous-phase transactinide chemistry behind TASCA can now be performed.

The new set-up termed *TA*sca *S*mall *I*mage mode *Spe*ctroscopy (TASISpec) [12] exploits advantages of the SIM, i.e. the fact that neutron-rich nuclides of SHE, produced in hot-fusion reactions, can be focused with high efficiency into an area of < 7 cm^2. This provides the unique possibility to build a compact Si-detector box for α-particle, electron, and fission-fragment measurements, and to pack composite Ge-detectors in very close geometry, resulting in an unprecedented, highly efficient set-up for multi-coincidence measurements with γ-rays and X-rays; see [12] for details. A prototype set-up has been commissioned successfully and first data have been collected for nuclides as heavy as ^{252,253}No.

In conclusion, the performance of TASCA as a separator is well understood and is perfectly under control. TASCA as a whole is presently the most versatile and highest efficient instrument in SHE research worldwide. It has entered the region of transactinides or superheavy elements, and is ready to explore the physics and chemistry of the "terra incognita" it was designed and built for.

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First Transactinide Chemistry Behind TASCA*

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The final phase of the TASCA commissioning program [1] included a series of experiments with ^{260}Rf, ^{261a,b}Rf and ^{262}Rf produced in the ^{244}Pu(^{22}Ne,xn) reaction. One of the highlights was a proof-of-principle transactinide chemistry experiment with 78-s ^{261a}Rf. The Automated Rapid Chemistry Apparatus (ARCA) [2] was used to study the formation of fluoride complexes of Rf in diluted HF-solution by anion-exchange chromatography.

TASCA was operated in the High Transmission Mode (HTM) [3] at a pressure of 0.4 mbar He. After passing a 14 cm x 4 cm large Mylar window of 1.2 µm thickness, evaporation residues were thermalized in He at 1200 mbar in a newly designed recoil transfer chamber (RTC) of 1.7 cm depth. The RTC had two funnel shaped inlets at the right and left hand side for the He/KCl jet and a central outlet in the cover plate. The Rf was transported to ARCA by a He/KCl jet through a 10 m long PE capillary of 2 mm i.d. at a gas flow rate of 2.9 L/min. To monitor the gas-jet yield, a ^{227}Ac emanation source was connected to the RTC. Ar with a flow rate of 20 mL/min was passed through the source and transported ^{219}Rn into the RTC. The yield of its decay product ^{211}Bi was compared to the respective yield in ROMA [4]. The ^{211}Bi yield in ARCA including collection, dissolution in $7x10^{-4}$ M HF solution, and evaporation on a Ta disc was 50% of that in ROMA.

For the anion-exchange chromatography in ARCA, the column magazines were filled with the resin MCI GEL CA08Y from Mitsubishi Chemical Corporation, particle size 22±5 µm, which was transferred into the hydroxide form as described in [5]. In each chromatography experiment, two Rf fractions were collected. The first one was $7x10^{-4}$ M HF, which was also used for column loading, and the second one was 5 M HNO_3. The latter was used to strip the remainder of the Rf from the column. After around 18 h experiment, the concentration of the first solution was changed to $1x10^{-3}$ M HF and the experiments were continued for another 25 h.

The KCl clusters were collected in ARCA for 90 s. Within this time, the column for the next experiment was preconditioned for 65 s with the HF solution. After the collection, the products were dissolved in 200 µl of $7x10^{-4}$ M HF solution and were subsequently fed onto the anion-exchange column at a flow rate of 1.0 mL/min. The effluent of the column was collected on a Ta disk as fraction 1. The fraction of the products adsorbed on the resin was eluted with 250 µL of 5 M HNO_3 and collected on a second Ta disk. Both fractions were evaporated to dryness by infrared light and a hot helium stream. The two Ta discs were then subjected to α-spectroscopy. Counting of the first fraction started 60 s after the end of the collection interval, counting of the second fraction started 65 s after the end of the collection.

In total, seven α-events were detected which we attribute to 78-s ^{261a}Rf based on the measured α-energy and lifetime. All of them were observed in the HNO_3 fraction. Two of these events were detected during the experiments with $7x10^{-4}$ M HF, the other five events were detected while using $1x10^{-3}$ M HF. As no events were observed in the HF fractions, it is only possible to give a lower limit for the %ads value. As in Poisson statistics zero observed events are compatible with three events at 95% confidence level, 3 events were assumed for the first fraction and the remainder in the second fraction resulting in %ads ≥ 62.5 % in $7x10^{-4}$ M HF and %ads ≥ 72.5 % in $1x10^{-3}$ M HF. The sum of the α events is shown in Figure 1 indicating that the α spectra were very clean, also thanks to preseparation in TASCA.

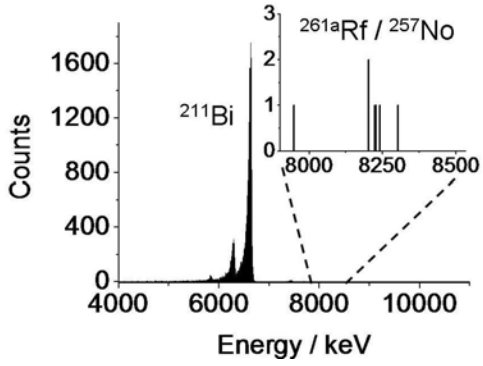

Figure 1: Sum spectrum of the α-particle events of ^{261a}Rf.

From a comparison with the number of ^{261a}Rf events in ROMA, we have to conclude that the chemical yield in ARCA was low, on the order of 30% only. This is in line with earlier observations indicating some sorption of transactinides from HF solutions on the Kel-F slider in ARCA.

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Die Herstellung von Plutoniumtargets für TASCA durch Elektrodeposition*

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Am **T**rans**A**ctinide **S**eparator and **C**hemistry **A**pparatus TASCA bei der GSI können durch Fusionsreaktionen mit Aktinidentargets Transaktinidenelemente (Z>103) hergestellt werden. An TASCA wird dazu ein aus drei bananenförmigen Segmenten mit einer aktiven Fläche von je 1,74 cm² bestehendes, rotierendes Targetrad benutzt. Eine 2 µm dicke Ti-Folie dient als Backing für die Elektrodeposition des Targetmaterials.

Kürzlich ist gezeigt worden, dass an TASCA die Produktion von Rf in der Reaktion ^{244}Pu(^{22}Ne,4-6n)$^{260-262}$Rf möglich ist [1]. In naher Zukunft soll das Element 114 durch die Reaktion ^{244}Pu(^{48}Ca,4n)^{288}114 produziert und seine chemischen Eigenschaften untersucht werden.

Da ^{244}Pu nur in sehr begrenzten Mengen erhältlich ist, muss die Targetherstellung mit hohen Ausbeuten erfolgen. Zur Optimierung der Parameter für die elektrochemische Abscheidung von Pu auf einer Ti-Oberfläche, werden die Experimente zunächst mit dem in größeren Mengen vorhandenen Isotop ^{239}Pu durchgeführt.

In einem Teflonbecher wird 1,5 ml einer ^{239}Pu-Lösung (1,25 mg Pu in 8 M HCl) zur Trockene eingeengt. Der Rückstand wird mit 100 µl warmer 0,5 M HNO_3 aufgenommen und mit 100 µl 0,1 M HNO_3 in die Platingzelle überführt. Mit 800 µl Isopropanol wird der Teflonbecher gespült und die Lösung ebenfalls in die Platingzelle überführt. Diese wird dann mit 15 ml Isobutanol auf insgesamt 16 ml aufgefüllt.

Die Platingzeit beträgt 6 Stunden. Dabei wird die Stromdichte auf 1,15 mA/cm² begrenzt (entsprechend einer Stromstärke von 2 mA). Versuche haben gezeigt, dass bei höheren Stromdichten die Pu-Schicht nicht fest auf der Ti-Unterlage haftet. Die Spannung beträgt am Anfang 160 V und am Ende der Platingzeit bis zu 370 V.

Während der Deposition werden in einem Abstand von einer Stunde 10 µl der Lösung entnommen und daraus ein α-Präparat hergestellt. Dadurch kann die Pu-Konzentration in der Lösung verfolgt werden.

Nach dem oben beschriebenen Schema kann man eine Ausbeute von 68% bis 90% erreichen. Die Massenbelegung der so hergestellten Targets wird mithilfe einer α-spektroskopischen Messung bestimmt und beträgt 350 bis 500 µg/cm². Bei einer Massenbelegung von mehr als 600 µg/cm², löst sich die entstandene Pu-Schicht nach dem Trocknen des Targets wieder von der Ti-Folie ab.

Die Homogenität der Schicht wird mittels Radiographie untersucht [2]. Dazu wird das Gerät FLA 7000 der FUJIFILM Corporation benutzt. Es konnte so gezeigt werden, dass bei Targetdicken bis zu ca. 500 µg/cm² das Targetmaterial sehr homogen auf dem Backing verteilt ist. Abb. 1 zeigt, dass die Abweichungen im Bezug auf die Massenbelegung ≤ 10 % betragen.

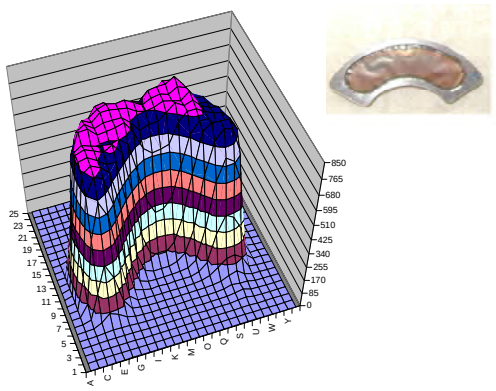

Abb.1: Radiographie und Photographie eines Targets mit 500 µg/cm²

Das zur Abscheidung verwendete ^{244}Pu enthält u.a. auch geringe Mengen ^{241}Pu und damit auch ^{241}Am, welches durch seine hohe spez. Aktivität wesentlich zur Gesamtaktivität des Targets beiträgt. Zur Untersuchung des Abscheideverhaltens von Am neben Pu wurde vor der Abscheidung ^{241}Am-Tracer zur ^{239}Pu-Lösung zugegeben. Es konnte bestätigt werden, dass die Deposition von ^{241}Am und ^{239}Pu parallel stattfindet, siehe Abb. 2.

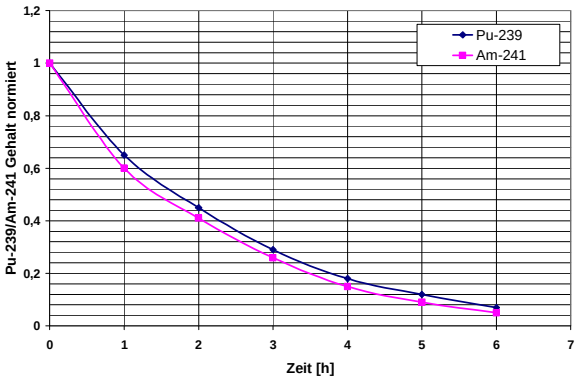

Abb.2: Gleichzeitige Abscheidung von ^{239}Pu und ^{241}Am

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Measurements of $^{260\text{-}262}$Rf produced in ^{22}Ne + ^{244}Pu fusion reaction at TASCA*

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As a final experiment in the commissioning phase of TASCA the transactinides (Z ≥ 104) were reached. Production and decay of ^{260}Rf, ^{261a,b}Rf and ^{262}Rf [1,2,3], produced in the asymmetric nuclear fusion reaction ^{244}Pu(^{22}Ne,xn) was studied. Separated reaction products were guided to a Focal Plane Detector (FPD) or into a Recoil Transfer Chamber (RTC), where they were available for transport to either the Rotating wheel On-line Multidetector Analyzer (ROMA) or to the Automated Rapid Chemistry Apparatus (ARCA) for chemical experiments [4].

TASCA was operated in the High Transmission Mode (HTM) [5]. The ^{22}Ne ion beam (average intensity: 0.8 μA_{part}) impinged on a rotating target wheel with 0.4 mg/cm^2 ^{244}PuO2 targets on 2.2 µm Ti backings. Three beam energies in the center of the target, $E_{c.o.t.}$, of 109 MeV, 116 MeV and 125 MeV, were used for the production of ^{262}Rf, ^{261}Rf and ^{260}Rf, respectively. The transmission of Rf has been optimized in He filling gas. The optimal pressure was 0.4 mbar. The magnetic rigidity, Bρ, was determined to be 1.99 T·m. To increase suppression of unwanted products, a He/H_2 (2:1) filling gas at a pressure of 1.5 mbar was used in experiments with the FPDs. Evaporation residues were implanted into a (80 x 36) mm^2 16-strip Position-Sensitive silicon Detector (PSD) or a (58 x 58) mm^2 Double-Sided Silicon Strip Detector (DSSSD). In other experiments, ^{261a,b}Rf passed a 1.2 µm thick (140 x 40) mm^2 Mylar window and was thermalized in 1.2 bar He in the RTC (depth: 17 mm). Rf atoms were then transported to ROMA by an He/KCl jet (gas flow rate: 3.45 L/min) through a 4 m long polyethylene capillary (inner diameter: 2 mm).

The measurement of ^{260}Rf, produced in the 6n evaporation channel at $E_{c.o.t.}$= 125 MeV yielded 15 time (Δt ≤ 200 ms) correlated EVR-SF events in the PSD. The correlation time analysis yielded a half-life of $21^{+7.3}_{-4.3}$ ms (errors are within the 68% confidence interval). A search for ^{262}Rf decays at $E_{c.o.t.}$ = 109 MeV 7 position and time correlated EVR-SF events observed in the DSSSD, with EVR energies of 0.8 to 3.3 MeV and SF fragment energies of > 100 MeV. The measured $T_{1/2}$ for ^{262}Rf is 210^{+128}_{-58} ms (Fig. 1a), in contradiction with values from [1,2]. In addition, 9 short EVR-SF correlations were registered with Δt ≤ 1.5 ms and EVR energies of 7.5 ± 5.0 MeV. They were attributed to the decay of ^{244mf}Am ($T_{1/2}$ = 0.9 ms). Because of a relatively high counting rate of EVR-like events in the DSSSD a random event analysis was performed for EVR-SF correlations within a Δt of 1 s. The random event number, n_b, was calculated individually for each observed event. It varies between 0.035 and 0.11 and depends on the event position in the DSSSD.

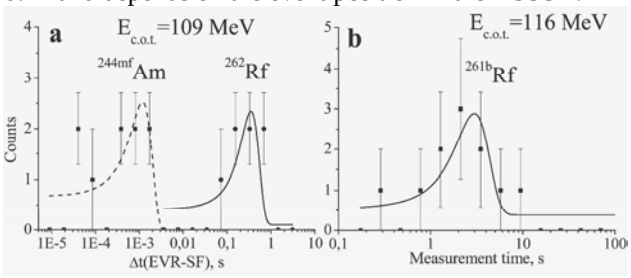

Figure 1: Time distributions of a) EVR-SF correlations from the DSSSD and b) SF decays from ROMA.

^{261}Rf was produced in the 5n channel at $E_{c.o.t}$ = 116 MeV and was detected in ROMA. Stepping time of 35 s (for ^{261a}Rf) were used. 149 single α-particles ($E_α$ = 7.8 – 8.5 MeV) from ^{261a}Rf and ^{257}No were registered; among these 28 α-α correlations. Also, 11 SF-events were registered and are attributed to ^{261b}Rf based on the measured $T_{1/2}$ of $2.2^{+0.9}_{-0.5}$ s. The SF activity assigned in [2] to ^{262}Rf likely originated from then unknown ^{261b}Rf.

From our results and cross section of 4.4 nb [6], a transmission of Rf through TASCA to a 140 x 40 mm^2 large area in the focal plane of 10% follows. For ^{261b}Rf, a cross section of $1.8^{+0.8}_{-0.4}$ nb was calculated, respecting decay during transport. With an estimated transmission of 6% to the area of the FPDs, preliminary cross sections for ^{260}Rf and ^{262}Rf of ≈ 1.2 nb and ≈ 250 pb, respectively, follows ^{261b}Rf was observed for the first time as an EVR. The production ratio of ^{261a}Rf to ^{261b}Rf is about 2.5:1. The data analysis is still in progress.

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A new TASCA focal plane detector*

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The new, highly efficient, gas-filled TransActinide Separator and Chemistry Apparatus (TASCA) has been recently put into operation at GSI with the aim to study chemical and physical properties of superheavy elements with Z≥104. Based on the results of magnetic field model calculations of the dipole and the quadrupoles, two modes of operation of TASCA, the "High Transmission Mode" and the "Small Image Mode" have been realized [1]. Two types of focal plane detector setups (FPD) were used during the TASCA commissioning phase: a (80 x 36) mm^2 16-strip position-sensitive silicon detector (PSD) and a (58 x 58) mm^2 double-sided silicon strip detector (DSSSD) as a prototype for a new TASCA focal plane detector. The test experiments showed that the image size in the HTM is larger than the detector size of both detector types, as it was expected according to ion transport calculations and Monte-Carlo simulations [1,2]. The nominal vertical position resolution of the PSD is ± 0.2 mm. However, a search for position correlated decay chain members usually occurs within ± 1 mm limits, corresponding to a pixel size area of (5 x 2) mm^2. The relatively large pixel size of the PSD negatively affects search limits for rare decay chains from long-lived isotopes of superheavy elements. The TASCA FPD working group has decided to build a detector setup based on a DSSSD as a stop detector and a backward array consisting of single-sided silicon strip detectors (SSSD). A *veto* detector for light fast ions that penetrate the DSSSD will be mounted behind the stop detector. The new setup will feature a (144 x 489 mm^2 large detector, which will accept > 90% of all evaporation residues reaching the focal plane. The geometrical detection efficiency for α-particles emitted from implanted nuclei will be > 70%. As compromise between a pixel size as small as possible and the number of spectrometric electronic channels as small as possible, a pitch width of 1 mm on the front and back side of the DSSSD has been chosen. To reduce the necessary number of ADCs the number of strips on each side of the DSSSD and on the SSSD should be a multiple of 8 when using 32-channel preamplifiers and 8 or 16-channel amplifiers with integrated multiplexers. A DSSSD structure with an active area of (72 x 48) mm^2 has 72 strips on the front side and 48 strips on the back side. The strip width and the interstrip distance on the both sides of the DSSSD are 900 µm and 100 µm, respectively. Two adjacent DSSSDs form the stop detector with an active area of (144 x 48) mm^2. A SSSD structure with an area of (72 x 48) mm^2 has 8 strips, which are 72 mm long. The strip width and the interstrip distance of the DSSSD are 5.65 mm and 100 µm, respectively. 8 SSSD detectors form the backward array with a depth of 72 mm. Two similar SSSD detectors are used as *veto* detector. Silicon wafer thicknesses of 300 µm and 500 µm have been chosen for the DSSSDs and SSSDs, respectively. These thicknesses will allow for the detection of conversion electrons in the backward array. A schematic design of the new TASCA detector array and first results from source measurements are shown in Fig. 1.

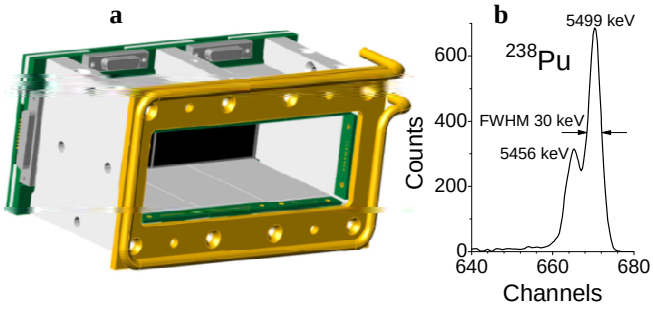

Figure 1: A drawing of the new TASCA FPD array (a) and an alpha particle spectrum from a DSSSD strip (b).

In total, the new TASCA detector array has to process the signals of 320 strips. The necessary spectrometric and data readout electronics, which are compatible with TASISpec [3], as well as the associated software, should be ready for experiments in spring 2009.

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TASISpec - A new twist on spectroscopy of superheavy elements.

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A new nuclear spectroscopy set-up called TASISpec (*TA*sca *S*mall *I*mage mode *Spec*troscopy) has been designed and commissioned. It exploits TASCA's specific small image focal mode, i.e. the fact that superheavy elements (SHE) produced in fusion-evaporation reactions can be focused into an area of less than 3 cm in diameter. This provides the world-unique possibility to pack composite Ge-detectors in very close geometry, resulting in an unprecedented, highly efficient detection of γ-rays and X-rays in coincidence with implanted SHE.

The test set-up as used in 2008 is illustrated in Fig. 1. It comprised four single sided silicon strip detectors (SSSD), one double sided silicon strip detector (DSSSD) and two Ge detectors; one cluster (7 crystals) downstream and a large VEGA clover (4 crystals) on one side. The final set-up foresees additional three clover detectors, while their implementation requires a dedicated holding structure to be built in 2009.

Conversion electron, γ-ray, and α sources as well as parasitic beams inducing the reactions ^{206}Pb(^{48}Ca, $2n$)^{252}No, ^{207}Pb(^{48}Ca, $2n$)^{253}No, ^{244}Pu(^{48}Ca, $4n$)^{288}114, and ^{150}Nd(^{64}Ni, xn)$^{214-x}$Ra, ^{154}Sm(^{64}Ni, yn)$^{218-y}$Th, ^{nat}Gd(^{64}Ni, zn)$^{224-z}$U, have been used to start to characterise the performance of TASISpec. Detection efficiencies amount to some measured 80% for emitted α particles and extrapolated more than 40% (absolute scale) for γ rays at an energy around 250 keV. In addition, energy thresholds of particle- and γ-ray detectors, dead times, and data rates of a first combined VME (up to 224 channels for Si-strip detector processing) and XIA-DGF (11 channels sampling the Ge-detector signals) data acquisition system were successfully tested as well as TASCA-SIM transmissions determined at different magnet settings.

In 2009 it is planned to establish the extraordinary $\gamma\gamma$-, γ-EC-, EC-EC, or even $\gamma\gamma$-EC-, multi-coincidence capabilities of the both segmented and compact TASISpec set-up by settling the decay scheme of K-isomeric states in ^{253}No [1, 2, 3, 4]. Despite of the underlying amount of data many question marks still remain regarding the structure of ^{253}No.

Following upon this ideal starting point to show the proof-of-principle of TASISpec, K isomers expected in neutron-rich SHE around ^{270}Hs [5] will be stepwise approached via, e.g., spectroscopic studies of ^{255,257}No, ^{261}Rf, and ^{265}Sg. Such a programme employs also the unique facets of both high UNILAC beam intensities, use of radioactive actinide targets, and high transmission of TASCA for rather asymmetric reactions. Last but not least, the unprecedented γ-efficiency of TASISpec may allow to discriminate SHE by means of characteristic X-rays.

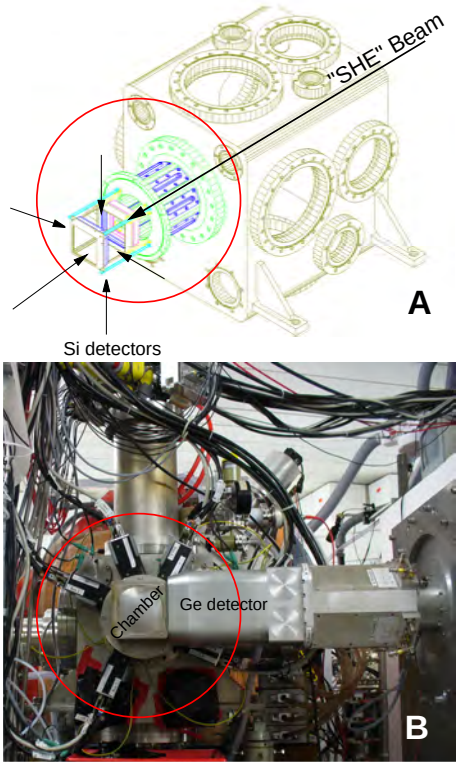

Figure 1: The TASISpec set-up installed at the focal plane of TASCA. Panel (A) illustrates a drawing of the end of the TASCA separator. The TASISpec structure is encircled. Five silicon strip detectors are placed in a cube-like shape. Panel (B) shows a photograph of the commissioning set-up. To the right a Ge-VEGA-clover detector is placed.

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Observation of ^{270}Hs in the complete fusion reaction ^{36}S+^{238}U*

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Superheavy elements ($Z \geq 104$) exist only due to nuclear shell effects. Deformed shell closures at Z=108 and at N=162 have been predicted theoretically [1] and observed in recent experiments [2]. Three hassium isotopes, $^{269-271}$Hs, have been produced in the complete fusion reaction ^{248}Cm(^{26}Mg, xn)$^{274-x}$Hs at the linear accelerator UNILAC at GSI [2,3]. The excitation function measurement at five different beam energies resulted in maximum cross sections of the 3n, 4n, 5n exit channels of a few pb.

Recent theoretical studies, analysing the formation of ^{270}Hs in the 4n channel, using a two-parameter Smoluchowski equation, predict even higher cross sections for the reaction ^{238}U(^{36}S,4n)^{270}Hs and ^{226}Ra(^{48}Ca,4n)^{270}Hs in the order of a few tens of pb [4]. Due to a lower reaction Q-value, these calculations predict a maximum cross section of about 24 pb for the ^{238}U(^{36}S,4n)^{270}Hs reaction, while the HIVAP code [5] predicts a maximum cross section of 3.5 pb.

Here, we report on first results of an experiment aimed to study the nuclear fusion reaction ^{36}S + ^{238}U, leading to the compound nucleus ^{274}Hs*. For the experiment we used our highly efficient chemical separation and detection system COMPACT which was connected to a Recoil Chamber (RC) installed behind the ARTESIA target wheel in cave X1 [2]. A beam of $^{36}S^{5+}$ ions was accelerated by the UNILAC up to 7.13 MeV/u and impinged on a rotating ^{238}U target wheel. During the experiment, we used two different sets of ^{238}U targets. The beam passed through a 20.2 µm Be vacuum window, 6 mm of He/O_2 gas mixture (He:O_2 = 9/1), a 12.6 µm Be target backing before entering the ^{238}U target (3 segments of 1.8 mg/cm^2, 1.5 mg/cm^2 and 1.6 mg/cm^2). The second target set had a Be backing of 8.2 µm and 3 segments containing 1.0 mg/cm^2 of ^{238}U each. We assumed that only Hs nuclei with a minimum residual range after exiting the target of 10 mm in gas could be transported to the detection system, corresponding to an active layer of the target of 1.0 mg/cm^2. The energy of the ^{36}S ions was in the range of 175.0 MeV to 181.2 MeV within the first set of target and in the range of 190.4 MeV to 196.1 MeV in the second set [6], corresponding to excitation energies, E*, of 38±3 MeV and 51±3 MeV [7], near the predicted maxima of the 4n and 5n evaporation channel, respectively. Starting at E*= 38 MeV we irradiated the first target set with a beam dose of $5.74 \cdot 10^{17}$ ions. The second target set was irradiated with a beam dose of $1.03 \cdot 10^{18}$ ions at E*= 51 MeV.

The data analysis revealed one correlated chain at the higher E* of 51 MeV. A 9.02±0.05 MeV α-particle was observed in bottom detector #24 followed after 23 ms by one 41 MeV spontaneous fission fragment measured in the top detector #24. Due to the measured properties we attributed this decay chain to the decay of ^{270}Hs produced in the 4n evaporation channel [2].

Because of background from α-decay and SF of heavy nuclides (A>200) from (multi) nucleon transfer pseudo correlated chains can be found with nonzero probability.

We have searched for possible random decay chains α·α·α·α, α·α·SF and α·SF, using subsequent time windows of 300 s length each. α-particles in an energy window $8.0 \leq E_\alpha \leq 9.5$ MeV and SF-like events with at least one fragment above a threshold of 15 MeV were considered. 351 and 771 α-particles have been registered in the first and the second run, respectively. Mainly this background was attributed to α-decays of ^{212}Po. In addition, 6 SF-like events were registered in the first run and 13 events in the second run. None of these high energy events were correlated in time and position to each other. The results are shown in Table 1.

Table 1: Random rates for different decay chains

decay chain	E* = 38 MeV	E* = 51 MeV
α·α·α·α	$7.08 \cdot 10^{-4}$	$3.12 \cdot 10^{-3}$
α·α·SF	$4.61 \cdot 10^{-3}$	$1.67 \cdot 10^{-3}$
α·SF	$1.6 \cdot 10^{-2}$	$4.6 \cdot 10^{-2}$

At E*= 38 MeV the cross section limit for both channels is 2.9 pb. The cross section for the 4n channel at E*= 51 MeV based on the one event is $0.8_{-0.7}{}^{+2.6}$ pb and the cross section limit for the 5n channel is 1.5 pb. Errors and limits correspond to 68% confidence level. The measured cross section and cross section limits are lower than for the reaction ^{248}Cm(^{26}Mg,xn)$^{274-x}$Hs [3], especially for 5n evaporation channel, in contrast to calculations made in [4]. We plan to continue these measurements in the near future.

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Liquid – liquid – extraction with the MicroSISAK – system

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In previous experiments it could be demonstrated that continuous liquid – liquid – extractions on a µl scale are possible with the MicroSISAK device [1]. Recently the IMM developed an improved version of this so called microreactor with two different mixer designs and up to three consecutive separation stages. A scheme of the setup for the experiments is shown in figure 1.

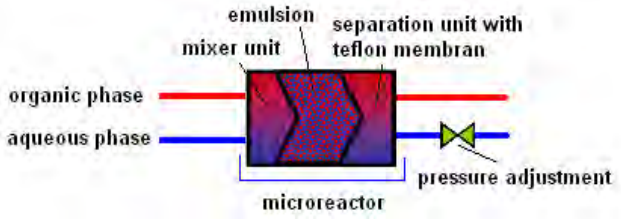

Figure 1: Scheme of the MicroSISAK setup

A static interdigital mixer is used for the formation of small droplets [2]. The formed emulsion is than separated by using a hydrophilic teflon membrane. A backpressure at the outlet of the aqueous phase corresponds to the internal pressure in the MicroSISAK device. This has an effect on the separation at the membrane. The inner volume of the used so far needle valve is much larger than the volume of the mixer, thus it is not possible to adjust a definite and reproducible backpressure in the separation unit.

Therefore a new apparatus was designed to set backpressure on the aqueous outlet of the microreactor by a gas volume. A standard vial made of polyethylene-terephtalate is equipped with three holes in the upper sealing cap: one as inlet for the aqueous phase coming from the reactor, and two for gas in and out. One hole in the bottom serves as liquid outlet (Fig. 2).

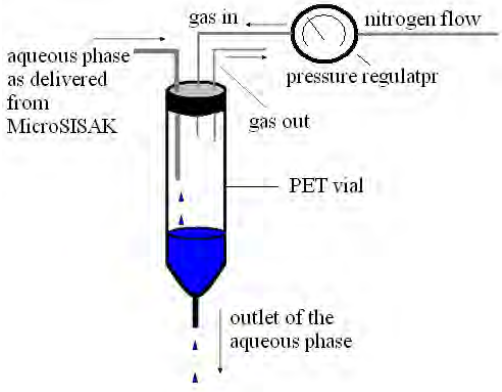

Figure 2: Scheme of the new pressure regulation apparatus.

The pressure in the vial is controlled with a high precision pressure regulator. It is reproducible with an accuracy of 5 mbar.

For studies on the extraction yield for hafnium – the lighter homologue of rutherfordium (Rf) Z = 104 – we have chosen an extraction system, that has successfully been applied in chemical investigations of Rf with SISAK [3]: Extraction of hafnium (Hf) with trioctylamine (TOA) in toluene from dilute sulphuric acid. While keeping the TOA concentration constant, the concentration of the acid was varied from 0.05 mol/l to 2 mol/ml. In order to measure the extraction yield Hf-181 is used as a tracer. Its activity in the organic phase is compared to the total activity in both phases. From this one can calculate the extraction efficiency. The main results of the batch experiments are shown in the upper line of figure 3. The results of the extraction experiments with the microreactor (lower part of figure 3) compare to the one in the batch experiments relating to the concentration of the two solutions.

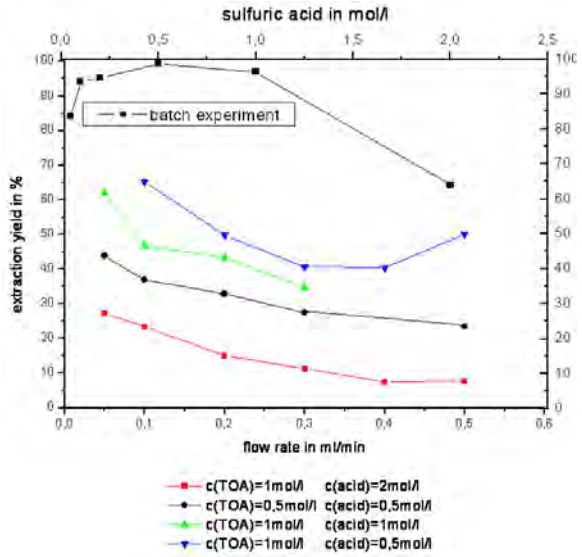

Figure 3: Extraction of Hf-181 with TOA: The upper line shows the extraction yield by varying the concentration of the sulphuric acid and a constant concentration of TOA (1 mol/l). The lower part: the extraction yield depends on the flow rate for different concentrations of the TOA and sulphuric acid.

One can observe a decrease of the extraction yield with increasing flow rates. At flow rates below 1 ml/min phase mixing is inefficient, thus extraction yield is low and mainly depends on the hold-up time in the mixer. At flow rates higher then 1 ml/min, smaller droplets are formed in the mixer. Thereby an emulsion is created and a much higher extraction yield can be expected. Corresponding experiments are currently carried out.

Acknowledgement:
I would like to thank the mechanical workshop of our institute for the realisation of the new pressure regulation apparatus.

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