Thesis for the Master’s degree in chemistry
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Investigations into new degasser technology for SISAK

60 study points

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Preface

This work was started in January 2008 under the supervision of Professor Jon Petter Omtvedt of the University of Oslo. The subject was fascinating as it combined cutting edge research in fields of chemistry, physics and engineering.

I would like to thank my supervisor for his support during this work, and for all help with the conclusion of this work which you now hold in your hands.

Special thanks to my fellow students Frøydis Schultz and Fereshteh Samadani for helping me with experiments and tirelessly listening to my ranting throughout the last two years. The discussions and inputs I got from the two of you have been of utmost importance for me and enabled me to finish this thesis.

Without the cooperation of Dr. Klaus Eberhardt and Daniel Hild from the Mainz University, most of this thesis would never have seen the light. Thank you for inviting me to your institute and for accommodating me for several months. Special thanks to the students at the nuclear chemistry department in Mainz, with Daniel leading the way, for showing me Mainz by night.

I would also like to express my gratitude to Professor Emeritus Jorolf Alstad for all the nice discussions and help with concluding this work. Thanks also to Professor Tor Bjørnstad for inputs for the development of the new degassers.

I am very grateful to the cyclotron operators in Oslo, Eivind Olsen and Andrey Semchenkov, not only for managing to keep the cyclotron running when the laws of physics disagreed, but also for lending me all their tools for my experiments.

Special thanks are also in order for the everyone at the workshop in the department of chemistry in Oslo, for all their help in improving my sketches and making the devices I needed for this work. Gratitude is also expressed to PALL Norway and Millipore Norway for making teflon membranes available for testing.

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Johannes Nilssen
Blindern, October 2009
Abstract

The prototype of a new liquid-liquid extraction system to be applied for small liquid volumes (microSISAK), in tracer studies of chemical properties of short-lived nuclides, has been tested and applied to longer-lived nuclides. The studies were performed at the institute for nuclear chemistry in Mainz, Germany, in cooperation with the staff and students there.

Development and testing of two new gas-liquid separation devices were performed at the University of Oslo. Two principles of gas separation were studied; separation of KCl clusters from the gas jet prior to transfer to the liquid phase, and separation of gas from liquid phase by membrane filtration.

Pre-separation of KCl clusters from the gas jet was successful, but the development of a method of transferring the clusters to aqueous phase could not be concluded within the timeframe of this work.

Development and testing of a static degassing unit, based on teflon filter separation, was successfully accomplished. The degasser has not yet been tested in connection with SISAK, but the principle of separation of gas from liquid by membrane filtration was proved.

A full scale SISAK experiment was performed at GSI, which gave important knowledge on how a transactinide experiment can be conducted. The results from the experiment showed how important fast transfer through the system is.
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1. Introduction

1.1. The periodic table of elements

The periodic table of elements was proposed by Dmitri Mendeleev in 1869 [1]. By arranging the elements in accordance to proton number he found that there was a periodicity in the properties of the elements. The predictive power of Mendeleev Periodic Table was demonstrated by the discovery of elements which at the time were unknown, but were found due to "vacant" places in the table. However, not all elements in the periodic table are found in nature (on Earth). The reason for this is that they do not have any isotopes which are either stable or long lived enough to have survived since they were created more than five billion years ago (in stars, before the earth was formed). Such elements, like technetium (Z=43), must be synthetically produced in nuclear reactions.

The heaviest naturally occurring element is uranium (Z=92). Heavier elements than U are known as transuranium elements and have only been available for studies during the last 60 years or so, as they must be produced using neutrons from nuclear reactors or intense charged-particle beams from cyclotrons and other types of particle accelerators.

Today, many of the synthetically made elements are comparatively easily available and have commercial uses, e.g. $^{243}$Am was commonly used in ordinary smoke detectors. However, the further away from the stable elements a given element is, the more difficult it is to produce. Thus, for the heaviest elements man has been able to produce, only a single atom at a time are available for study, as the life time of each atom is much shorter (seconds or less) than the production rate (a few per hour or days or even weeks). This is generally true for the so called super heavy elements, which is the popular name for the elements heavier than the actinides, i.e. with > 103. These elements are also referred to as transactinide elements (UIPAC recommends the terms lanthanoid, actinoid and transactinoid, but the research field commonly uses lanthanide, actinide and transactinide).
1.2. Chemical properties and Quantum Mechanics

Ever since the Schrödinger equation was introduced in 1926, quantum mechanical models have been used to calculate electron configuration in atomic structure, and also to explain the connection between outer electron structure and properties of the atom.

Early on in the transuranium research, Glenn T. Seaborg proposed the introduction of the actinide series in the periodic table of elements, analogous to the lanthanide series [2], which changed the look of the periodic table, but also helped explain why the transuranium elements didn’t show the properties one expected from the old periodic table.

The same motivation applies to today’s transactinide research: Do a “new” element, i.e. one which has been synthesized for the first time, follow the trends discovered by Mendeleev? Can current quantum mechanical models and calculations explain its chemical behavior? Such fundamental questions are highly interesting and provide the driving force for the big and expensive experiments currently undertaken to probe into physical and chemical properties of elements further and further away from the stable elements.

Figure 1.1 shows the modern periodic table of elements with the transactinides in purple (element 117 has yet to be discovered).
1.3. Fusion Reactions

Production of the heaviest elements is done by fusion reactions (Fig. 1.2). Fusion reactions of heavy target materials and beams have made it possible to produce nuclei with proton number as high as 118 (which is the heaviest element observed).

Production rate of trans-actinides is very low due to the small cross-section of forming the desired product, and it decreases when the atomic number of the product nuclei increases [3].

The properties of the heaviest elements are more or less unknown due to the short half-lives and low production rate causing experiments to be time consuming and expensive.

1.4. Investigation of chemical properties

Determining the properties of the heaviest elements by experimentation will give important knowledge on the extent to which relativistic effects [4] have an influence on the outer electron orbitals. There are different ways of investigating these elements, but due to the fact that their half-life is shorter than the production rate, experiments must be performed in "single atom at a time" mode. Experimental systems involved in investigation of these elements thus have to be very quick. In addition the experiments have to be performed over long time periods in order to produce enough data. A lot of undesired reaction products are formed in the nuclear reactions as well, so the system has to be efficient in separating the desired reaction products from the rest.

Most of the available knowledge on chemical properties of the transactinides comes from gas-phase chemistry studies. Gas-phase chemistry is easier and faster than liquid chemistry, but many properties (such as complex formation and solubility) can only be investigated by means of liquid chemistry experiments.

Solvent extraction chemistry on one atom at a time means that conventional distribution ratios determined from the concentration of a solute in two phases cannot be determined, as one atom is restricted to one phase at any given time. A solute will however undergo diffusion even at low concentrations, and by conducting the experiment over a long time it is possible to achieve statistical data that can be used to determine the distribution ratio.
SISAK\textsuperscript{1} is a fast continuous solvent extraction system designed for studying short-lived isotopes with half-lives down to \(~\text{1s}\) \cite{5}. It consists of a series of mixers and centrifuges for fast mixing and separation of different phases. The system has successfully been applied for studies on transactinides element 104, rutherfordium \cite{6}. Benefits of the system include fast extraction, good separation and good efficiency in detection. Drawbacks of the system are mainly large consumption of solvents and poor energy resolution in detection.

MicroSISAK is a minimization of the existing SISAK system. It drastically reduces the problems concerned with solvent consumption, and opens for the possibility of utilizing surface detectors with better energy resolution. Drawbacks of the system include the requirement of very pure and stable flows, which prove to be hard to meet due to the low flow rates, and longer hold up times in the system. The current system has not been tested for online studies, much due to the lack of a suitable degasser.

The current degasser for SISAK is not able to deliver stable flows in the low flow regime required by microSISAK, and the only degasser device that has been connected with previous microSISAK devices is not able to deliver a continuous flow. Two new types of degassers have been developed and tested in the current work, but not in connection with SISAK or microSISAK.

\begin{footnote}{
\textsuperscript{1} SISAK is an acronym for “Short-lived Isotopes Studied by the AKufve method”, where AKUFVE is a Swedish acronym for “\textit{\`{A}nordning f"{o}r \text{"{O}nders"{o}kning av F"{o}rdelningsj"{a}mvikter vid V"{a}tske Extraktion}”, i.e. “arrangement for continuous investigation of distribution ratios in liquid extraction”.
}
\end{footnote}
2. Background

2.1. Transactinides

As for the actinide series, for the transactinides it is expected that the relativistic part of the wave function will strongly determine the valence shell electron orbital. How strong the relativistic effects are compared to the non-relativistic effects has to some extent been determined by theoretical approach, but not validated by experiments to sufficient degree. Different methods of calculation lead to different results, and the only way to determine how strong these effects really are, is by measuring the properties of these elements.

2.2. Solvent extraction

Solvent extraction, also known as liquid-liquid distribution, involves the distribution of a solute, or solutes, between two immiscible liquids in contact with each other. The two liquids are generally one aqueous phase, and one organic phase. A solute A originally dissolved in one phase, eventually will distribute between the two phases depending on the strength of the solutes interactions with the different solvents.

Concentration of solute A in the aqueous phase is \([A]_{aq}\) and \([A]_{org}\) in the organic phase. The distribution ratio of the solute (formula 1.1) is “the total analytical concentration of the substance in the organic phase to its total analytical concentration in the aqueous phase, usually measured at equilibrium” [7].

Distribution ratio:

\[ D = \frac{[A]_{org}}{[A]_{aq}} \]  

(1.1)

Extraction of a solute is dependent on that solute’s interactions with the extract (the separate phase containing the solute extracted from another phase). These interactions include chemical reaction kinetics and diffusion. The diffusion of an atom from one phase to the other is only possible if that atom can be extracted. This again requires the atom to be on an appropriate chemical form (i.e. atomic, complex, ion etc.). The rate of extraction is consequently dependent on the chemical reaction kinetics and the rate of diffusion, and the overall extraction kinetics is dependent on the slowest of these [8]. Diffusion is the process where matter moves from one region with higher concentration, to one with lower concentration.

Fick’s first law (equation 2.1) states that the diffusive flux (number of moles crossing each square meter per second) is proportional to the concentration gradient;

\[ \text{Flux} \left( \frac{\text{mol}}{m^2 \cdot s} \right) \equiv J = -D \frac{\partial c}{\partial x} \]  

(2.1)
where $J$ is the diffusion flux, $D$ is the diffusion coefficient, $c$ is the concentration of the solute and $x$ is the distance of diffusion.

Fick’s second law (formula 2.2) predicts how concentration changes with time due to diffusion:

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2}$$

(2.2)

this formula can be rearranged to give [9];

$$t \sim \frac{d^2}{D}$$

(2.3)

where $t$ is the mixing time, $d$ is the lamellae width (the liquid streams can be split into many narrow streams (lamellae) to increase mixing) and $D$ is the diffusion coefficient.

Equation 2.3 shows that the mixing time can be reduced simply by reducing the volume of the liquid streams mixed, and thus increasing the contact area (Fig. 2.1). This is achieved by fast and rigorous mixing.

Figure 2.1: Increased contact area by splitting of liquid streams.
2.3. **SISAK**

The solvent extraction system SISAK (Fig. 2.2) was designed for studying short-lived isotopes from complex nuclear reaction production mixtures [10]. It was later applied to the field of superheavy element research.

![Diagram of SISAK system](image)

**Figure 2.2:** SISAK online experimental setup with one extraction stage (figure from [10]).

SISAK consists of static mixers, small centrifuges with high rotation speed for separation of liquid and gas phases and a detection system (Fig. 2.2). For homologue studies the detection system consists of two gamma detectors, one for the organic phase and one for the aqueous phase. For transactinide experiments the detection system consists of an array of liquid scintillation counters (usually two for each phase).

The system has been improved several times over the years, most notably the size reduction of the system, and the change from Ti to PEEK\(^2\) as construction material [11] has made the system faster and more resistant to different solvents. The current generation is SISAK 3.

SISAK has successfully been applied to investigation of transactinide element 104, rutherfordium [6], and proven that it can be applied to chemical investigations of superheavy elements.

---

\(^2\) Poly Ether Ether Ketone
2.4. MicroSISAK

The microSISAK prototype version 3 (Fig. 2.3) device has been developed by the Institut für Microtechnik Mainz (IMM) as a co-operation between IMM, Chalmers University and the nuclear chemistry group at the University of Mainz. It is as the name implies a minimized version of SISAK. With much lower flow rates, the microSISAK device can solve many of the problems concerning SISAK, such as the low resolution of LS counting and the large solvent consumption. By reducing the flow from 0.5 mL/s to 0.5 mL/min it could be possible to vaporize the solvents from the outlets, and directly adsorb it on surface detectors. This gives a relatively good counting geometry and better resolution than LS counting offers.

Another advantage is that microSISAK utilizes static separation, which ensures fast and stable separation of the two phases. This especially applies to the organic phase, as the aqueous phase is unable to pass over the membrane, thus eradicates contamination of the organic phase. This makes the system more reliable (unless the mixer or filter clogs) and easier to handle.

Early versions of the device has been successfully applied for online extraction of $^{104}$Te [12] in test experiments.

2.5. Alpha detection

There are different ways of detecting $\alpha$ decay, but for chemical investigation using SISAK, liquid scintillation counting has been the preferred method as the detection yield for $\alpha$ particles is very high, and it can be run continuously in connection with SISAK. Scintillation detectors make use of the fact that certain materials emit light (i.e. scintillation) when struck by a nuclear particle or radiation. Scintillation occurs when radiation (in form of photons or particles) passes through the scintillating material and excites the atoms and molecules in the scintillation liquid, which in turn emits light as it de-excites.
The big benefit of using liquid scintillation detectors for α detection is that due to the short range of energy deposition of α particles, nearly every disintegration is detected, giving a counting efficiency close to 100% [13].

A problem with liquid scintillation detectors is quenching of the light emitted in the scintillation liquid. Quenching includes all aspects that might decrease the fluorescence intensity of the scintillator, such as impurities in the scintillator causing absorption of the light emitted, energy transfer which either results in no light output, or light output of a different wavelength etc.. This makes liquid scintillation detectors extremely sensitive to any impurities in the solvent.

To avoid quenching the organic phase mixed with the scintillation liquid has to be extremely pure to avoid water, the mix also has to be flushed by an inert gas to avoid oxygen in the scintillator, both known for their immense quenching abilities.

Another way of detecting α particles is the use of surface detectors. These detectors have better energy resolution than scintillation detectors, and are therefore a better tool for distinguishing between alpha peaks that are close together in energy. Since the range of α particles is so short, the particles entering the detector will most likely deposit the energy in the detector. However, since a flat surface detector can maximum detect 50% of alpha events (due to geometry), the efficiency will be lower than that of liquid scintillation detectors. The efficiency of surface detectors can be raised by coupling two or more detectors together in a “sandwich”, but there will always be problem regions such as gaps between detectors and detector edges.

Surface detectors are sensitive to surface impurities, such as particles or liquid films, which will cause energy absorption of particles. They are also sensitive to light and have to be mounted in a light tight enclosure.
3. Methods and instrumentation

3.1. Production of radioactive nuclides:

For experiments with radioactive hafnium in Germany, the TRIGA (Training, Research, Isotopes, General Atomic) research nuclear reactor at the University of Mainz was used for thermal neutron activation of natural Hf and natural Na:

\[
^{180}\text{Hf}(n,\gamma)^{181}\text{Hf} \quad (t_{1/2} = 42.39 \text{ d}) \quad (3.1)
\]

\[
^{23}\text{Na}(n,\gamma)^{24}\text{Na} \quad (t_{1/2} = 14.96 \text{ h}) \quad (3.2)
\]

The production rate (R) of activated Hf is given by the formula:

\[
R = \sigma \phi N_0 \quad (3.3)
\]

For online experiments in Oslo, the cyclotron at the Oslo Cyclotron Laboratory (OCL) was used. The cyclotron at OCL is a Scanditronix MC-35 cyclotron, with a radius of 51 cm that has been in use since 1978. Beams available from OCL are listed in Table 3.1.

<table>
<thead>
<tr>
<th>Particle</th>
<th>p</th>
<th>d</th>
<th>(^3\text{He})</th>
<th>(^4\text{He})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Energy</td>
<td>2-35 MeV</td>
<td>4-18 MeV</td>
<td>10-47 MeV</td>
<td>7-35 MeV</td>
</tr>
</tbody>
</table>

Table 3.1

One way to produce radioactive nuclides with a particle accelerator, which is widely applied in the field of chemical investigations on transactinides, is by irradiating a target made of a thin metal with a beam of accelerated particles (Fig. 3.1). When the accelerated particles hit the target material nuclear reactions occur, producing new isotopes.

![Figure 3.1: Target chamber at OCL](image)

For experiments performed in the present work, radionuclides were produced by irradiating \(^{nat}\text{W}\) and \(^{nat}\text{Zr}/^{nat}\text{Hf}\) targets with a \(^3\text{He}\) (45 MeV, 0.5-1 \(\mu\text{A}_{\text{ran}}\)) beam:
The recoil products are transferred from the target chamber to the experimental set-up by a He gas-jet, with KCl aerosol particles to catch the recoil products, through a capillary tube.

### 3.2. Liquid-liquid extraction:

#### 3.2.1 SISAK mixer:

In order to transfer the species formed in the recoil transfer chamber from the gas phase to the liquid phase the two phases must be mixed rigorously and fast before separation to transfer as much of the species as possible to the liquid phase before separation. To achieve good mixing a large contact surface between the two phases is important. This is a problem as the flow of gas to that of the liquid in the mixer is about 50:1 for SISAK and 1000:1 for microSISAK. This means that highly efficient mixers are necessary.

The most widely used mixer in the SISAK group in Oslo is a PEEK tube mixer, made of PEEK and filled with PEEK wool (Fig. 3.2) in order to increase the contact area of immiscible phases. The mixers can be customized with regards to length, diameter and density of PEEK wool to individual experimental requirements.

![Figure 3.2: SISAK PEEK wool tube mixer.](image)

The same mixers as for gas and liquid are used for two liquid phases.

#### 3.2.2 SISAK liquid centrifuge

The SISAK 3 centrifuges (Fig. 3.3) currently used for studies of super-heavy elements are small centrifuges with high speed of rotation (16 000 - 28 000 rpm) to ensure fast separation of liquid phases with flow rates in the range 0.1 - 0.8 mL/s.
Separation of the two phases is done by centrifugal force. The center of the centrifuge rotates and the liquids are separated since the one with highest density will be located furthest away from the centre. Outlets placed closer and farther away from the centre will extract the light and heavy phase, respectively. Throttle valves are fitted on the outlets for adjusting the counter pressure and thus the position of the phase boundary in the chamber. Clean phases (less than 1% entrainment) are achieved when the phase boundary is adjusted to be midway between the two outlets.

With an inner volume of 0.3 mL the SISAK centrifuges achieve hold up times of less than one second when the highest possible flow rates (about 0.5 mL/s for each phase) are used.

3.2.3 MicroSISAK

The microSISAK prototype version 3 device consists of a stack of titanium disks with micro-scale mixer and separation units machined into the surface of the disks, all stacked together in a Ti housing. The following units were produced for testing: Two mixer units with interdigital channels (interlacing flow channels, in the fashion of a zipper), one with 16+16 channels (Fig. 3.4) and one with 1+2 channels [12].

Mixing is achieved by sending the two phases as counter-flows through 2 mm long and 20 µm broad interdigital channels. The laminated flow leaves the mixer perpendicular to the feed flow directions through a small slit and fast mixing takes place (Fig. 3.5).
Figure 3.4: mixing unit for microSISAK prototype version 3 with 16+16 channels.

Figure 3.5: Multilamination (interlacing of several layers of sheet-like flows) of liquid streams in interdigital channel mixer ensures fast transfer through diffusion (figure from [14]).

The micromixer has been tested as a separate unit in many applications [15], giving results for droplet size as function of flow rate (Fig. 3.6). The droplet size determines the contact area of the two phases, and thus the diffusion length. In general, diffusion between two phases is determined by the diffusion length and the contact time of the two phases.
The separation units, specially constructed for liquid-liquid extraction by the SISAK group at Mainz University in cooperation with IMM [17], consist of a stretched TEFLOW membrane between two Ti-disks, with grooved channels on each side (Fig. 3.7). In order to force the organic phase through the membrane, a small differential pressure (10-50 mbar) is applied over the membrane.

The microSISAK prototype version 3 device can be set up with either one or three separation units (Fig. 3.8 and Fig. 3.9).

With a total inner volume (including mixer, separation units, flow channels and inlets and outlets) of ~0.02 mL the microSISAK device is considerably smaller than the SISAK system. The system was designed to provide a hold up time of maximum 1 s for a flow rate of 0.02 mL/s.

So far the microSISAK prototype version 3 device has only been tested off-line.
3.3. Gas-liquid extraction

3.3.1 SISAK centrifuge degasser

The centrifuge degasser (Fig. 3.10) that is currently used for SISAK experiments separates the gas-phase from the aqueous phase by centrifugal force. To ensure good separation the rotating speed of the degasser must be high, between 17 000 and 20 000 rpm.
The SISAK degasser has an inner volume of 0.1mL which results in a hold up time, using the highest possible flow rates (~0.5 mL/s per liquid phase), of less than a second.

### 3.3.2 ALOHA

![ALOHA sideview schematic drawing](image)

The ALOHA (Automated Liquid Online Heavy element Apparatus) degasser device (Fig. 3.11) is a semi-continuous gas pre-separator. A tantalum disk with 4 positions (Fig. 3.12) ensures transfer of aerosol particles from the gas jet to aqueous phase in a stepwise manner. First the aerosol particles are deposited on the disk (position 1), then after a preset collection time, the disk is rotated 90° to position 2 where the deposited particles are dissolved and flushed into the liquid chemistry system (e.g. microSISAK) (step 2). Position 3 and 4 in ALOHA are for washing and drying the disk respectively.

![Ta-disk for ALOHA](image)

The ALOHA device has been successfully tested with microSISAK prototype version 2 for extraction of radioactive Tc produced in a nuclear reactor [12].

### 3.3.3 Nozzle-based degasser

One option for achieving good mixing, with reduced liquid flow, is to reduce the volume of gas mixed with the liquid. The main problem with this is that the gas-flow has to be high in order to transport the recoil products from the target chamber to the SISAK set-up fast and efficiently, i.e. the gas-flow
cannot be reduced. In order to solve this problem, methods of pre-mixing separation of gas from the KCl clusters are sought. Part of the work of this thesis has been focused on investigating such possibilities.

One method, investigated in the current work, is to lead the gas-jet through a small nozzle. The momentum of the KCl clusters will keep them moving in a forward direction for a certain distance, but the gas will disperse and can be pumped out of the chamber.

![Diagram](image)

Figure 3.13: nozzle-based degasser schematic drawing

A prototype degasser for "proof-of-principle" testing with two compartments (Fig. 3.13), one for gas-separation (A), and the other for mixing KCl clusters with liquid phase (B), was made by the workshop at the department for chemistry in Oslo. The gas-jet is led through a small nozzle in the first compartment, where the gas is sucked out through 1-4 gas-outlet points, all connected to a vacuum pump. The distance from the nozzle to the filter can be adjusted in order to focus or disperse the KCl clusters on the filter.

Within the scope of the current thesis it was not possible to develop the mixing section of the degasser, this is work for a future project.
3.3.4 Membrane degasser

The degasser consists of a PEEK housing (Fig. 3.14), with a compartment divided by a stretched TEFLOM membrane (Fig. 3.15) that ensures quick separation of aqueous phase from gas phase as well as organic phase (i.e. the degasser principle can be applied to liquid-liquid separation). Separation occurs due to the different polarity properties of the phases; the membrane allows gas and non-polar liquids (e.g. toluene) to pass, but not polar liquids (e.g. water, acids etc.).

The degasser has an inner volume of ~0.8 mL, which gives it a hold up time of several seconds with low flow rates.
4. Experiments

The Master's degree work consists of several different types of experiments. The overall aim was to look into possibilities for developing a more efficient and preferentially simpler (with respect to use and reliability) degasser for SISAK. Initially a period was spent at Mainz University to learn about and participate in the development of the microSISAK system and its membrane technology. Then several prototype devices (based on the nozzle and membrane separation) were built in Oslo and tested. Finally, participation in a full-scale SISAK experiment at GSI were done, which gave invaluable insight into the requirements of how a real-life transactinide experiment must be performed and a much better understanding of degasser requirements.

4.1. MicroSISAK

The microSISAK setup consists of two syringe pumps; one for aqueous phase and the other for organic phase, see the schematic drawing in Fig. 4.1. The interconnections are by PFA tubes with 0.15 mm inner diameter.

![Figure 4.1: microSISAK experimental setup.](imageurl)

The microSISAK device was tested with a well known extraction system for rutherfordium: TOA as extraction agent dissolved in toluene as organic phase, and dilute sulfuric acid as aqueous phase.

An aqueous solution with 20 mg/mL hafnium and 25 mg/mL sodium was neutron activated in the TRIGA reactor before being loaded in the syringe pump. The amount of activity per mL was:

\[
^{180}\text{Hf}(n,\gamma)^{181}\text{Hf}
\]

\[
D = 13 \cdot 10^{-24} \text{cm}^2 \cdot \frac{0.7 \cdot 10^{15} n}{\text{cm}^2 \cdot \text{s}} \cdot \frac{0.02 \text{ g} \cdot \text{mol}}{180 \text{ g} \cdot \text{ml}} \cdot N_A \cdot \left(1 - e^{-1.287 \cdot 10^{-5} \text{ s}^{-1} \cdot 21600 \text{ s}}\right) \approx 2.5 \text{ MBq} \text{ ml}^{-1}
\]
\[
\frac{^{23}Na(n, \gamma)^{24}Na}{D = 0.53 \cdot 10^{-24} \text{cm}^2 \cdot \frac{0.7 \cdot 10^{12} n}{\text{cm}^2 \cdot s} \cdot \frac{0.025 g \cdot mol}{23 g \cdot ml} \cdot N_A \cdot \left(1 - e^{-1.89 \cdot 10^{-7} s^{-1} \cdot 21600 s}\right) \approx \frac{58.9 MBq}{ml}}
\]

Sodium was added to make sure that the system was exclusively extracting Hf, which it did.

Problems with the pumps due to backpressure from the microSISAK device as well as clogging of the mixer unit and the separation filters occurred several times, and illustrated some of the major drawbacks of the system.

### 4.2. Nozzle-based degasser

For online studies of the degasser, the OCL cyclotron was used, with a 45 MeV \(^3\)He-beam, with intensities ranging from 0.5-1.0 \(\mu A\), on a \(^{nat}\)W target, and a mixed \(^{nat}\)Zr/\(^{nat}\)Hf target, producing various radioisotopes of osmium, or molybdenum and tungsten, respectively.

A helium gas-jet with KCl aerosol particles was used to transport the recoil products from the target chamber in the cyclotron hall to the degasser.

The 145 keV peak in the osmium gamma spectra, and the 100, 116, 122, 141 and 257 keV peaks from the tungsten/molybdenum spectra were used for investigating the scattering of the aerosol particles.

The focusing of the KCl clusters over various distances between the nozzle opening and a catcher disk, different nozzle openings, and varying gas flow were tested. The first tests were performed with two filters to collect the activity: One (filter A) was placed in front of the nozzle opening, and the other (filter B) was placed in front of gas outlet 3 (see Fig. 4.2).

During experiments on the 29th of May, the gas connection to the KCl cluster oven developed a leak due to high pressure. This led to uncertainties about the actual pressure and flow in subsequent experiments, and was not discovered and corrected until 14th of September. Unfortunately, due to all the problems with operations of the OCL cyclotron, resulting in very little available beam time and frequent unscheduled down periods, it was not possible to redo these experiments within the framework of the current thesis. The results of the experiments are included here, but should of course be repeated when beam time is available.
For focusing tests, one filter was placed in front of the nozzle opening, then divided in a center part (A1) with a diameter of 20 mm and a periphery part (A2) after collection (Fig. 4.3), before being placed in front of the detectors. To get the best possible counting statistics, the filters were placed in front of alternating detectors (i.e. filter A1 in front of detector 8 in run 1,3,5 etc. and in front of detector 4 in run 2,4,6 etc, and the opposite for filter A2). In this way, both filters could be counted simultaneously, but the individual detector efficiencies will cancel out.

Figure 4.3 Center cutout (A1) and periphery part (A2) of filter in filter holder A.
Problems with the cyclotron caused several stops and fluctuation of the beam, resulting in difficulties producing valid data for the experiments, as already described above.

4.3. Membrane degasser

For preliminary tests, the degasser was set up with a 5 cm standard PEEK tube mixer, where purified water and He gas-jet was mixed and run through the degasser. A PALL [REF or specs!] stretched TEFLOON membrane with 0.45µm pore size was used. For the first runs, there was no vacuum connected on the gas outlet (this turned out to be because of a gas leak from the KCl oven, which severely reduced the gas flow through the system). For higher flow-rates of the He gas-jet a vacuum pump was connected to force the excess gas through the membrane. A needle valve was also fitted on the vacuum hose to prevent damage to the membrane itself due to pressure differences.

PALL teflon membrane with 0.45 µm pore size (serial numberTF-450) was successfully tested as separation filter in the degasser. Millipore teflon membranes (serial number FGLP04700, FHLP04700 and FALP04700) were also tested, but suffered from bad separation as water penetrated the membrane for all pore sizes (0.22-1.0 µm).

The same radioisotope production setup was used for online studies of the membrane degasser as for the nozzle-based degasser, with exception of the natW target. The gas-jet was transported to a SISAK mixer, where the gas-jet and purified water was mixed to transfer the KCl particles to aqueous phase before separation of the gas-phase in the degasser.

The direct catch (DC) filters were placed in the same type of bottles as the samples from the degasser, but not dissolved. This could lead to some uncertainties in the counting, due to placement error, as well as effecting the efficiency of the detector due to distance to the sample.

Again, problems with the cyclotron caused several stops and fluctuation of the beam, resulting in difficulties producing valid data for the experiments.
4.4. TASCA SISAK experiment

Figure 4.4: The schematic set-up of the SISAK system for measuring of distribution of $^{172,174}$OsO$_4$ between NaOH solutions and toluene as a pilot experiment for chemical investigation of Hs experiment [19].

Fereshteh Samadani et al.’s SISAK extraction system for hassium [19] was performed full scale on osmium at GSI (Gesselschaft für Schwerionenfororschung) in Darmstadt, Germany, in April 2009. The system takes advantage of the fact that group VIII elements form volatile tetraoxides, which can be transferred in the gas jet without KCl clusters. This also results in a selective transport from RTC to SISAK where only group VIII elements will be transferred.

The purpose of the experiment was to determine whether SISAK could be successfully coupled with TASCA (Fig. 4.4), a gas-filled separator for pre-separation of unwanted reaction products, and to establish whether the system could be performed if applied for hassium.

SISAK was equipped with liquid scintillation $\alpha$-detectors to see whether the system was fast enough to detect $\alpha$ particles.
In transactinide experiments the system has to be very fast in all aspects, and fast transport from the RTC (Recoil Transfer Chamber) to the chemical setup is very important. For investigations on the transport time from RTC to SISAK, the beam was irradiating the target in preset pulses of 10 s, and the time from irradiation to detection was used to establish the transport time from RTC to degasser (Fig. 4.5). Comparison of the activity collected from the degasser to the activity collected on Al catcher foils in the RTC was used to establish the transport time from RTC to SISAK and the efficiency of the degasser.

All the pumps for liquid phases were tested for stability and accuracy to see whether they would deliver stable flows for this experiment and also to see the stability for different flow rates for future experiments. The flow rate of the pumps is adjusted by a feed voltage to the pumps, i.e. by increasing the voltage the flow increases. For experimental setup the pumps are connected to flow meters which monitor the flow from the pumps and gives feedback to the pump control system for increased stability of the flow rate. The feedback to the pump control system was switched off for investigating the fluctuations in flow (monitored by the flow meters) from each pump at a set voltage. The liquid volume after a 10 min collection time was compared to the volume of 10 min collection of the pumps with feedback control.
5. Results and discussion

For all measurements with the nozzle-based and the membrane degasser the number of counts in each peak was divided by the beam-integral and multiplied by 2000 to give counts normalized for a beam integral of 2000. The beam integral is proportional to the amount of current passing through the target. For the experiments performed in this work the absolute value of the integral is of no consequence, since it is only used for comparisons of series of experiments. Thus, the beam integral is in arbitrary units.

5.1. MicroSISAK

![Graph showing the distribution ratio of Hf extraction as a function of H2SO4 concentration.]

Figure 5.1: batch experiments with extraction of Hf to the organic phase as a function of H2SO4 concentration, from Daniel Hild [18].

The extraction of Hf by TOA in toluene is dependent on the concentration of H2SO4 in the aqueous solution as shown in figure 5.1.
Comparison of SISAK and microSISAK for the same H2SO4 concentration dependence (Fig. 5.2 and 5.3) shows that SISAK gives higher extraction yields than microSISAK, even with lower extractant concentration. This is most likely due to poor mixing, a recurring problem with microSISAK.
As explained in section 2.2, the extraction of elements from one phase to the other is dependent on contact area and contact time. In figure 5.4 the extraction decreases with increasing flow rate due to shortened contact time of the two phases.

In order to increase extraction, either the contact area has to be increased, or the flow rate has to decrease. As lowering the flow rate will result in unacceptable hold up time in the system, the contact area between the two phases must be increased. This can be done by further increasing the flow rate, and thus force the liquids to form smaller droplets in the emulsion. To increase the contact area, higher flow rates were tested, but the generated back pressure from microSISAK caused the syringe pumps to fail. The contact time can also be increased by increasing the length of the channels where the emulsion flows within the device.

Developing a chemical system with faster kinetics may also be a solution, but since the tested system is among the fastest systems ever developed for SISAK, this is probably not practical.

Figure 5.4: Extraction with microSISAK as a function of flow rate.
The backpressure generated from microSISAK as a function of flow rate can be seen in figure 5.5. As previously mentioned in chapter 3.2.3 good mixing of two immiscible phases in the IMM interdigital mixer requires relative high flow rates (to achieve a droplet size of 60µm the flow rate has to be 5 mL/min and for a droplet size of 20µm the flow rate has to be 20 mL/min). This flow regime will result in a backpressure of more than 2 bar for microSISAK, which will result in problems for the pumps. At such high flow rates, the mixing time would also be very short as the hold up time for microSISAK as a whole would be less than 100 ms, which is too short to achieve good mixing.

5.2. Nozzle-based degasser

From setup 4.3.1 the results show that $96.3^{\pm 7}$% of the activity measured is collected on filter A. It means that the particles to high extent move in a straight forward direction and are not sucked out with the gas through outlet 3.
Experiments investigating the focus of the aerosol particles show that increasing gas flow results in better focus of the particles (Fig. 5.7, and Fig. 5.8). The uncertainties in figure 5.6 are...
probably higher for low flow rates as the gas flow was more or less unknown due to problems with leakage from the KCl cluster oven.

The studies show that the KCl clusters can be separated from the gas jet prior to transfer to the aqueous phase.

5.3. Membrane degasser

Off-line experiments performed with the membrane degasser showed that the degasser would separate gas from liquid phase, without sub-pressure applied over the membrane. It later turned out to be mainly caused by leakage in the gas jet giving lower flow rates than read out on the flow meter.

![Graph showing activity transferred from gas jet to aqueous phase in membrane degasser compared with activity in direct catch filters.](image)

Figure 5.8: Activity transferred from gas jet to aqueous phase in membrane degasser compared with activity in direct catch filters.

The amount of transferred activity from the target chamber to the membrane degasser and to the direct catch filter are comparable (Fig. 5.8) for different gas flows.
Figure 5.9: Comparison of activity in direct catch filter (DC), SISAK centrifuge degasser (SDG) and membrane degasser (MDG).

As shown in figure 5.9, the extraction yields are comparable within the uncertainties for the SISAK centrifuge degasser and the membrane degasser.

Figure 5.10: Comparison of membrane degasser (MDG) activity to that of direct catch filter (DC).

Even with samples DC filters with undissolved activity (i.e. they should give a higher efficiency for the detector) measurements of the samples from the degasser show that the
extraction yield is more or less equivalent to that of the activity collected on the DC filters (Fig. 5.9 and Fig. 5.10).

The long hold-up time in the degasser would cause problems when investigating nuclides with very short half-lives unless the flow rate was increased to the order of mL/s.

![Graph showing variation of DC activity for various runs using the measurement of the for 100 keV γ-peak.](image)

As shown in figure 5.11 the amount of particles that is transferred from the RTC varies a lot, this gives further uncertainties for all results.
5.4. TASCA

Results from main experiment:

Figure 5.5: Comparison of the results from the experiment at GSI with α-emitting Os to the batch and SISAK experiment at OCL using γ-emitting Os. The distribution of OsO₄ between NaOH solutions and pure toluene was studied (figure from [19]).

As shown in figure 5.12 the results from the TASCA experiments are comparable to the experiments performed in Oslo. This proves that the system is fully functional when connected to TASCA, and that transactinide experiments with SISAK are feasible at GSI.

Time measurements from RTC through the SISAK degasser:

The sum of activity in the aqueous phase gathered from the degasser and the sum of the Al catcher foils corrected for beam integral are listed in table 5.1.

<table>
<thead>
<tr>
<th></th>
<th>174Re</th>
<th>173W</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sum NaOH</td>
<td>601,4</td>
<td>64,4</td>
</tr>
<tr>
<td>Sum Al-foil</td>
<td>1519,3</td>
<td>111,9</td>
</tr>
</tbody>
</table>

Table 5.1

The transport time from the RTC to the degasser could be determined by comparing the fraction of transferred activity to the degasser for the two isotopes, which should be equal to each other, adjusted for disintegration (174Os t₁/₂ = 45s and 173Os t₁/₂ = 22,4s):

\[
\frac{601,4}{792,3} = \frac{1519,3 \cdot 0,5^{t/45s}}{3801,6 \cdot 0,5^{t/22As}}
\]

By solving the equation with respect to \( t \), a transport time of 41.28 s was found.
The efficiency of the system was calculated by inserting the correct transport time:

\[
\frac{601,4}{1519,3 \cdot 0,5^{\frac{41,28}{45s}}} \approx 0,75
\]

giving a result of 75% efficiency for the RTC, gas jet, mixer and degasser combined.

The long transport time from RTC to SISAK is mainly caused by the RTC itself. For the pilot experiment the efficiency of transfer from the RTC was of higher priority than a fast transfer, as the nuclides investigated have relatively long half-lives. For a transactinide experiment the transfer time will have to be much shorter, and a smaller RTC should be applied (and a shorter capillary). The smaller RTC provides much faster transfer, but suffers from higher yield loss in TASCA due to the different focusing arrangement used to create the smaller image size required for a small-area RTC.

**Pump tests:**

![Graphs showing flow rate stability for different pump flows measured by flow meter.](image)

Figure 5.6: Flow rate stability for different pump flows measured by flow meter.

In general, pump 5 seem to deliver more stable flows than the other pumps (the voltage of the pumps were set manually, thus some differences in mean value for the different pumps).
Pump 5 (Micropump, model I-Drive) is a new generation of pumps where a magnetic field drives the pump wheel instead of a motor, resulting in more stable flows.

Measurements with and without flow feedback of the pumps gave similar results for all pumps in terms of volume collected after a 10 min collection time. The pumps were not connected to SISAK, and flow correction was thus not required. For experiment setup, flow correction is crucial as fluctuation in liquid flows greatly affects mixing and extraction.

One of the biggest problems concerning microSISAK is that the pumps are not able to push liquids through the device due to the high counter-pressure generated by microSISAK. The I-Drive pump type could be able to force the liquids through the device, even at higher counter-pressures.
6. Conclusion

6.1. MicroSISAK

The current version of the microSISAK device does not operate at, or at least close to, equilibrium conditions. Thus, it is not yet suitable for performing transactinide experiments. This is most likely due to poor mixing in the device, and improvement of the internal mixer or adding an external mixer unit should be considered.

From experience with operation under real-life conditions, it seems that microSISAK is too miniscule, and thus generates too much counter-pressure at the necessary flow rates for a transactinide experiment.

One solution is to apply stronger pumps to force liquids through the device, but this could lead to problems with perforation of the teflon membranes.

By increasing the size of microSISAK, the backpressure generated should drop, and the system would be operative with current pumps and degassers.

Applying static separation of the different phases seem to work quite well for gas-liquid as well as liquid-liquid separation, and with minor adjustments microSISAK should be able to meet the requirements set by transactinide investigations.

6.2. ALOHA

Although ALOHA has been tested and run for several real-life experiments its main flaw is that it delivers a semi-continuous flow where one wants continuous flow. This makes it absolutely impractical for liquid-liquid extraction, which requires very stable liquid flows. In addition, it is not very reliable and it produces too much electrical noise to be put in close proximity of sensitive detectors.

For low liquid flow rates combined with high gas flows the principle of the ALOHA is very good. If it could be improved with regards to continuity of liquid output it could very well be applied to transactinide experiments.

6.3. Nozzle-based degasser

The nozzle-based degasser lacks it’s most important feature; the ability to transfer activity from the gas-jet to liquid phase. As there will always be a sub-pressure in the degassing compartment of the device, it will be very hard to prevent liquid phase from being sucked through the small hole in the separating wall. One option might be to have a “running film” of liquid in the mixing department, where hydrogen bonds forces the liquid phase to stay in place as the KCl clusters are shot directly in the liquid phase.
For use with microSISAK this would be the better option as the low liquid flow rates will result in a gas to liquid volume ratio of ~1000:1 which will make good mixing difficult. It has not been time enough within the current project to look into such solutions, but will certainly be interesting to follow-up in future projects.

6.4. Membrane degasser

The membrane degasser seems to work quite well with a pore size of 0.45µm.

By reducing the inner volume of the degasser, the hold-up time in the degasser should be comparable to that of the SISAK degasser or better, and if the membrane proves reliable in terms of lifetime (i.e. it doesn’t clog up or tear apart) the membrane degasser can replace the SISAK degasser in future experiments, providing an important simplification.

In order for the membrane degasser to work with the flow regime required for microSISAK, a new geometry with bigger surface (e.g. a stretched TEFLON tube or similar) for the degasser should be considered. There are membrane degassers commercially available with tubular TEFLON membranes, but most are designed for much higher flow rates or longer hold up times than required by SISAK/microSISAK.
7. Future perspectives

The microSISAK system has to reduce the counter-pressure generated by the device; this can either be done by enlargement of the device, or by applying sub-pressure after the outlets and thus sucking all liquids through it. Improved extraction through better mixing, either by improving the internal mixer or by applying external mixing units also needs attention.

For the nozzle-based degasser the challenging point will be the development of transfer to liquid section for the degasser. Its further development will be very important, as the miniaturization of the liquid-liquid extraction system, either as microSISAK or downscaling of the current SISAK system, inevitably must lead to lower flow rates. It is known that high gas-jet transfer yields require high gas-flows (up to as much as 5 L/min). Therefore, the ratio between the volume of gas and liquid becomes very unfavorable for a good transfer of the activity. In this work it was proved that a high reduction in gas volume can be achieved by linear momentum separation. Thus, the first step towards developing a degasser where only the aerosol particles and not the gas are mixed with the liquid has been achieved.

From the current experiments and developments, it is also clear that a membrane type degasser is both practical and feasible to develop quickly. More detailed experiments is needed to decide if it is efficient enough to be used even for high gas-flow and low liquid-flow rates, but regardless of this it will be very useful as a substitute of the booster degasser stages used in SISAK transactinide experiments for removing the flushing gas needed to expel oxygen from the scintillator solutions.

Before the membrane degasser can replace the centrifuge degasser, it has to be tested that it can deliver a stable output flow. To ensure good mixing of the liquid phases and appropriate D-values, the liquid flows going in the mixer has to be extremely stable. If it can deliver a stable output flow, it should also be tested with the microSISAK device. Another requirement which has not been tested but needs verification is if the pressure behind the outgoing liquid flow is high enough to feed the SISAK liquid-liquid mixer/centrifuge.

Since the microSISAK device operates with miniscule flow rates, the degasser will probably need to be miniaturized as well in order for the nuclides under investigation not to disintegrate due to long transport times. Testing how much gas/liquid ratio the degasser can handle will be crucial for developing a smaller version.

The "lifetime", i.e. the time it takes before it needs to be replaced, of the teflon membranes should also be tested to see if it is worthwhile changing from rotating to static separation. If the service interval is too short, it will cause too many interruptions during experiments.
Appendix

A.1 Acronyms and abbreviations

<table>
<thead>
<tr>
<th>Acronym</th>
<th>Definition</th>
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<tbody>
<tr>
<td>SISAK</td>
<td>Short lived Isotopes Studied by the AKufve method</td>
</tr>
<tr>
<td>AKUFVE</td>
<td>Anordning för Kontinuerlig Undersökning av Fordelningsjämvikter vid Vätske Extraktion (arrangement for continuous investigations of distribution ratios)</td>
</tr>
<tr>
<td>ALOHA</td>
<td>Automated Liquid Online Heavy element Apparatus</td>
</tr>
<tr>
<td>TASCA</td>
<td>TransActinide Separator and Chemistry Apparatus</td>
</tr>
<tr>
<td>PEEK</td>
<td>Poly Ether Ether Ketone</td>
</tr>
<tr>
<td>OCL</td>
<td>Oslo Cyclotron Laboratory</td>
</tr>
<tr>
<td>RTC</td>
<td>Recoil Transfer Chamber</td>
</tr>
<tr>
<td>TOA</td>
<td>Tri-n-octylamine, (C₈H₁₇)₃N</td>
</tr>
</tbody>
</table>

A.2 Formulas and calculations

Mean values of repetitive measurements are given by the formula:

$$ x = \frac{\sum x_i}{n} $$

where $x$ is the mean value, $x_i$ is the result of each individual measurement, and $n$ is the number of measurements.

A.3 Uncertainty

Where several measurements have been performed, and added together to give a mean result, the standard deviation ($s$) is given by the formula:

$$ s = \sqrt{\frac{\sum (x_i - \bar{x})^2}{n - 1}} $$

For some measurements, the results have been added to produce better statistical data, the uncertainties are then given by the law of propagation of error, which gives the formula for uncertainty as:

$$ s = \sqrt{(s_1)^2 + (s_2)^2 + \cdots + (s_n)^2} $$

Counting uncertainties are given by the formula:

$$ \sigma = \sqrt{N} \quad \text{where} \ N = \text{number of detected counts in the peak.} $$
References


12. Eberhardt, K., Separation of Tc from fission products with the extraction system MicroSISAK.


17. Eberhardt, K., MICROSI SAK – A NEW DEVICE FOR FAST AND CONTINUOUS LIQUID-LIQUID EXTRACTIONS ON A MICROLITER SCALE.


19. Samadani, F.e.a., A SISAK extraction system for chemical study of element 108 Hs. to be published.