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Chemical properties of the transactinide elements studied in liquid phase with SISAK

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Abstract. This article starts with a review of the current SISAK liquid-liquid extraction system, as used after the physical preseparator BGS at LBNL for chemical studies of transactinide elements. Emphasis will be on new additions and developments. Then the possibilities offered by the new TASCA separator at GSI and the use of actinide targets at both GSI and LBNL are discussed with respect to future SISAK transactinide experiments. Finally, current and future liquid-liquid extraction systems for studying elements Rf up to Hs are discussed.

PACS. 23.60.+e Alpha decay – 27.90.+b $220 \leq A$ – 29.40.Mc Scintillation detectors

1 Introduction

The on-line liquid-liquid extraction system SISAK[1] has been adapted for studies of the chemical properties of the transactinide elements. SISAK [1,2] was developed to investigate short-lived nuclei recoiling out of an irradiated target and transported in a gas jet. The system is based on small centrifuges with a continuous feed and output and is one of the fastest systems available for liquid-phase studies of transactinide elements. For transactinide experiments, liquid scintillation (LS) [3,4] is used to detect their nuclear decay. The LS detectors have high detection efficiency, but limited energy resolution. Therefore, transactinide experiments with SISAK are performed with a physical preseparator between the target and SISAK to suppress unwanted reaction products. Until now, the Berkeley Gas-filled Separator (BGS) at Lawrence Berkeley National Laboratory (LBNL) has been used, see Stavsetra et al. [5, 6]. Recently, a new separator, named TransActinide Separator and Chemistry Apparatus (TASCA) [7], at the Gesellschaft für Schwerionenforschung (GSI) in Darmstadt, entered the commissioning phase. Thus it will soon be possible to also perform SISAK experiments at this facility.

2 Status of the SISAK system

2.1 Double detector setup

To achieve a sufficient energy resolution in the LS detectors, only the LS occurring in the organic phase can be measured. The aqueous phase cannot be measured due to severe quenching of the LS process. For the same reason, only carefully selected organic solvents and solutes can be used [3,6]. Liquid-liquid distribution ratios of the element under study between the two phases could therefore only be determined indirectly. The amount in the aqueous phase had to be deduced from the measured activity that

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[1] SISAK = **S**hort-lived **I**sotopes **S**tudied by the **AK**UFVE-technique, where AKUFVE is a Swedish acronym for an arrangement for continuous investigations of distribution ratios in liquid-liquid extraction.

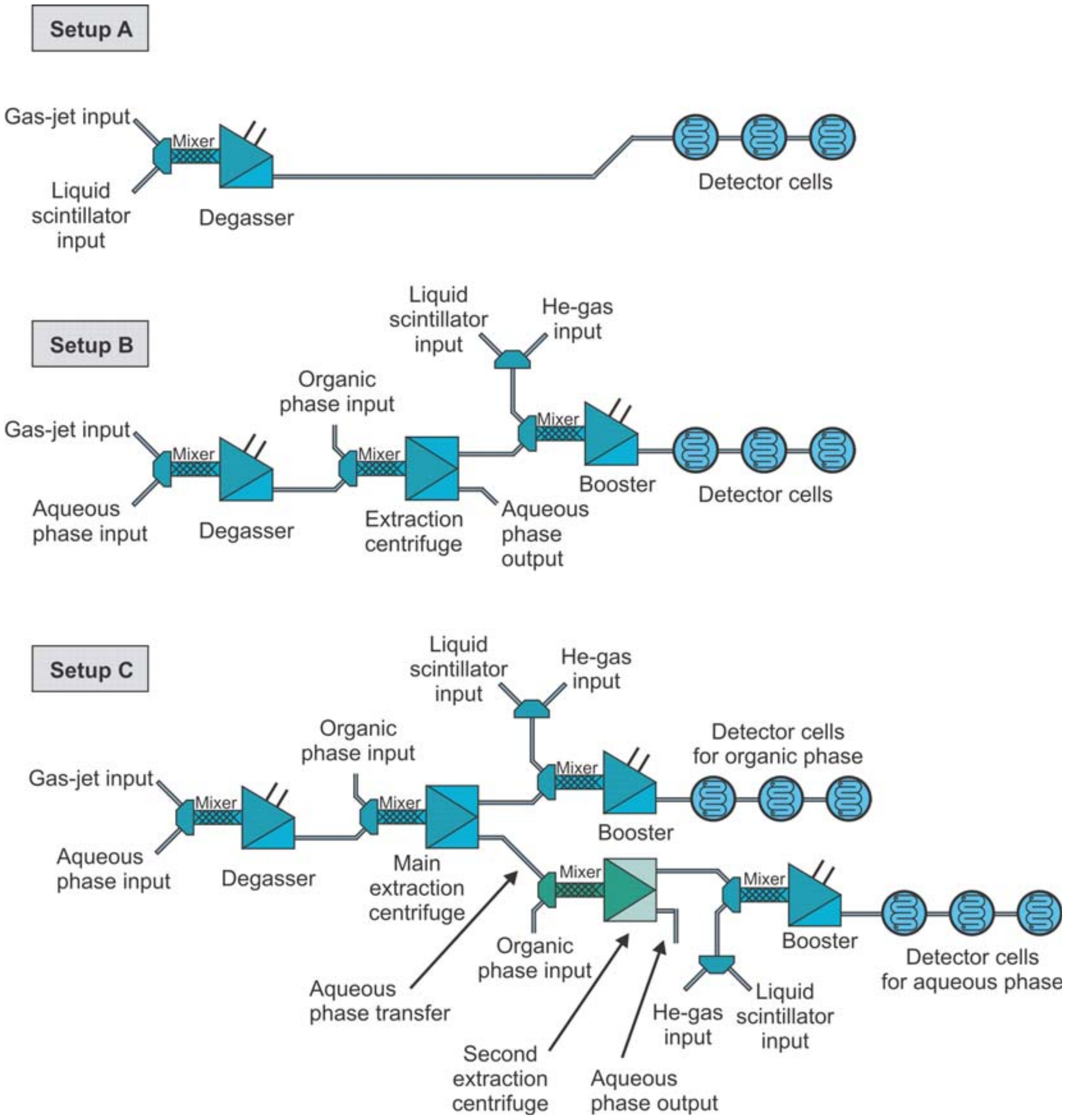

Fig. 1. (Color online) Schematic drawings of SISAK setups used in investigations of transactinide elements. Setup A is used to measure the amount of activity entering the system. The gas-jet carried activity is dissolved directly in a liquid scintillator cocktail and measured. No chemical separation is performed. Setup B is used to measure the amount of activity in the organic phase after liquid-liquid extraction. The amount of activity in the aqueous phase is not measured but must be deduced from a separate experiment with setup A. Setup C enables the measurement of activity in both phases using a second extraction step and a second array of liquid-scintillator detectors.

entered the extraction centrifuge and the amount in the organic output phase. Since these two measurements cannot be done simultaneously, the experiment takes twice as much time and is susceptible to variations in the gas-jet transfer yield between the two measurements. In order to overcome this disadvantage, the SISAK system was recently extended to include a second extraction step in which part or all of the activity remaining in the aqueous phase after the first extraction stage is transferred to a second organic phase [8]. This way the amount of radionuclides in the two phases exiting the first extraction stage can be measured simultaneously, allowing direct determination of the distribution ratio. See Figure 1 for a schematic drawing of the different setups which have been used for investigations of transactinide elements with SISAK. The relationship between the distribution ratio in the first extraction stage and the amount of activity in the first and second organic phase is explained in [8].

The new system with a second extraction stage to transfer the activity in the aqueous phase from the first stage into a second organic phase, was used for the first time in an rutherfordium experiment at LBNL in April 2005 [9]. See Section 3.1 for further discussion of this experiment.

2.2 Preseparator limitations

So far, only heavy-ion induced cold-fusion reactions with targets of ^{208}Pb and ^{209}Bi have been possible at LBNL if the BGS is used as a preseparator for chemistry

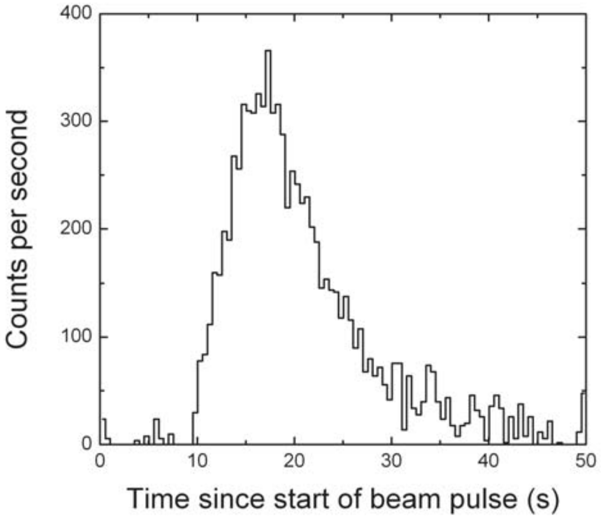

Fig. 2. Measured transport time from target to the first detector through a SISAK setup with one extraction stage. The flow rate was 0.8 mL/s. The average hold-up time is about 18.5 s.

experiments. This is mainly due to the required momentum of the evaporation residues (EVR), as explained below. The only transactinide elements available for liquid-phase chemistry studies after physical preseparation were Rf and Db by using 4.7-s ^{257}Rf and 4.4-s ^{258}Db. Other transactinide nuclei available from cold fusion reactions do not have long enough half lives for chemistry studies with SISAK. The ^{257}Rf and ^{258}Db nuclei have much shorter half lives compared to the more commonly used 78-s ^{261}Rf and 34-s and 27-s ^{262,263}Db nuclei produced in hot-fusion reactions. Hot fusion reaction products cannot be used in the current preseparation setups due to the low momentum of the evaporation residues (EVR) produced in the asymmetric reactions with beams of e.g. ^{18}O or ^{22}Ne. This is because the EVRs will not be able to pass through the vacuum window separating the low-pressure (∼1 mBar) environment of the separator and into the high-pressure (1.5–2.2 Bar) gas-jet recoil transfer chamber (RTC). Alternative reactions suitable for preseparators are being sought, see Section 3 below.

The restrictions to 4–5 s nuclei impose very stringent limits on the transport time that the EVRs need to travel from the RTC to the detectors. Figure 2 shows a transport-time measurement made in an experiment with a setup equal to setup B in Figure 1 [10]. The transport time was measured by using $^{164-166}$Hf activity produced in a ^{120}Sn(^{50}Ti, xn)$^{170-x}$Hf reaction with a pulsed beam (3 s on and 87 s off). The $^{164-166}$Hf nuclei have half lives equal to 114 s, 76 s, and 6.8 m, respectively. The Hf activity carried with the gas-jet was dissolved in a liquid scintillation solution in the degasser stage and pumped through the remaining apparatus and detection cells. The scintillation solution was pumped in a closed loop at 0.8 mL/s and changed when needed. Using the data shown in Figure 2 and a Monte-Carlo simulation, the number of ^{257}Rf atoms surviving long enough to be detected in cell 1 was estimated. For this estimation a 50% gas-jet, 67% gas to liquid transfer, and 80% extraction yield were used. This simulation showed that only 7 out 1000 ^{257}Rf nuclei reached the detector [11].

One bottleneck is the liquid transport through the chemistry apparatus (SISAK). Thus, a method was developed to enable rapid and precise measurements of the transport time through the SISAK system without the use of pulsed beams or radioactive tracers. In this way flow-through time measurements can be done easily and quickly in connection with tests and developments to reduce the transit time. This system uses coloured solutions and light transmittance transducers (LTT) to detect the presence of the coloured solution [10]. A schematic drawing of the setup for measuring the transport time through the degasser is shown in Figure 3. An example of the use of such measurements is given in Figure 4. Here, the hold-up time of different lengths of mixers for flow rates of 0.4 mL/s and 0.8 mL/s is shown. The results clearly show the difference in flow-through time for the two flow rates measured. Further measurements show that the average transport-time for a centrifuge with an attached mixer at the inlet is 2.5 ± 0.2 s at typical flow-rates for transactinide experiments (0.4–0.5 mL/s of each liquid phase). The same transport time was measured for a degasser unit, including the gas/liquid phase mixer. This implies that each stage in a SISAK setup adds about 2.5 s to the overall hold-up time. Taking into account the tubing and connectors coupling the whole system together, it can be estimated that the average hold-up time for setup B and the first extraction stage in setup C in Figure 1 is about 10 s. From Figure 2 it can then be deduced that about 55% of the hold-up time is due to the SISAK apparatus and about 45% due to the RTC and gas-jet transfer.

3 Target and RTC development

Work to develop thinner vacuum windows between the separator and the gas-jet transfer chamber is currently in progress both at LBNL and GSI [12]. This, in combination with the use of actinide targets like U or Pu, will allow the production of a broader range of nuclei since the recoil energy of these hot fusion reaction products will be high enough to pass through such an improved window. Therefore, in the future it will be possible to perform SISAK experiments with elements heavier than rutherfordium.

3.1 New possibilities with Pu targets

Work is in progress at the BGS at LBNL and at TASCA at GSI to enable the use of Pu targets with the separators. This has previously not been possible at the BGS due to concerns regarding safety and maintaining a low-level detection environment. At GSI, the TASCA separator is being designed from the start with such targets in mind. Thus, a wider range of reactions will soon become

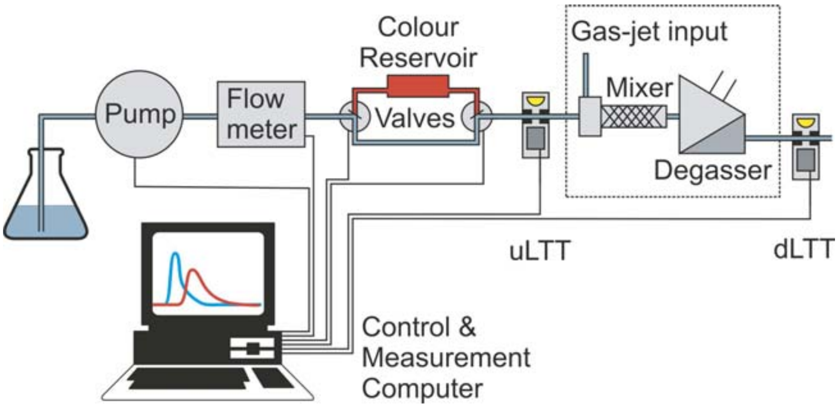

Fig. 3. (Color online) Setup for measuring the flow-through time using a coloured solution and detection by light transmittance transducers (LTT). The flow-through time is measured for the equipment placed within the dashed square. Different parts or sections of the SISAK setup can be selected for flow-through time measurement. uLTT and dLTT indicate upstream and downstream LTT, respectively.

Table 1. Likely reactions with ^{244}Pu targets for producing suitable isotopes for SISAK experiments on the transactinide elements Rf to Hs. The EVR recoil energy and the corresponding range in the mylar RTC window is shown for each reaction. For comparison, the reaction between ^{50}Ti and ^{208}Pb to produce ^{257}Rf is included. The decay loss is calculated on the assumption that the average transport time is 18.5 s, as the measurement shown in Figure 2 indicates. The relative yields in the last column are based on the decay loss column.

Reaction	Product $T_{1/2}$ (s)	EVR recoil energy (MeV)	Range in mylar (μm)	Decay loss in SISAK	Relative yield
^{244}Pu(^{22}Ne, 5n)^{261}Rf	78	9.5	1.7	15%	12
^{244}Pu(^{23}Na, 5n)^{262}Db	33	10.8	1.9	32%	9.7
^{244}Pu(^{26}Mg, 5n)^{265}Sg	7	13.5	2.4	84%	2.3
^{244}Pu(^{27}Al, 4n)^{267}Bh	15	14.4	2.6	57%	6.1
^{244}Pu(^{30}Si, 5n)^{269}Hs	14	18.1	2.9	60%	7.7
^{208}Pb(^{50}Ti, 1n)^{257}Rf	4.7	45.5	6	93%	1

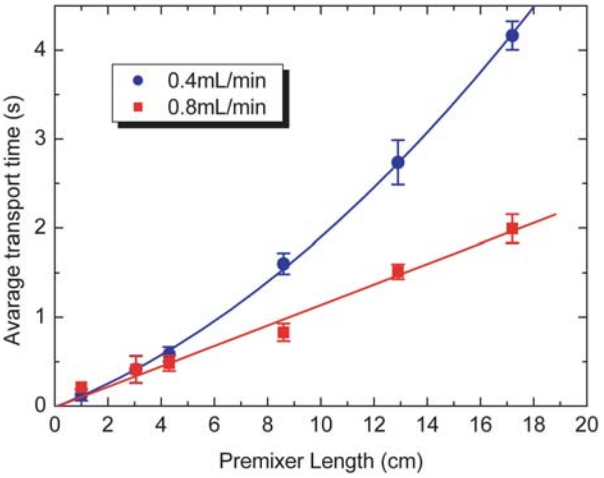

Fig. 4. (Color online) Hold-up time in mixers of different lengths for flow rates 0.4 mL/s and 0.8 mL/s measured by using coloured solutions and light transmittance transducers as explained in the text.

available for chemistry experiments behind physical separators. Some of the most relevant reactions for producing the elements from Rf through Hs are listed in Table 1.

It can be clearly seen from the table that the EVR recoil energy and corresponding range in the mylar foil used for the RTC window is much smaller than for the ^{50}Ti on ^{208}Pb reaction used so far. However, if thin enough windows can be constructed (see Sect. 3.2 below), much longer-lived isotopes become available. Considering the large decay-loss during the transport encountered in ^{257}Rf experiments, the smaller cross-sections for the indicated reactions will to a certain extent be compensated by the reduced transport-time loss, as indicated in the column for relative yield in the table. The gains will be particularly important for elements Rf and Db, which also are produced with the largest cross sections.

3.2 Small-volume RTC

The measurement of the average transport time encountered in SISAK experiments at LBNL indicate that the transport time from the RTC to SISAK is about 8–9 s. A 20-m long capillary with 1.56 mm i.d. is used to transport the EVRs attached to KCl aerosol particles with He gas flowing at 1.2 L/min. Most of the delay is due to the large volume of the BGS RTC chamber, which is necessary due to the large image size in the BGS focal plane (140 mm × 57 mm). The RTC depth is 40 mm, thus the chamber volume is 320 mL. Necessarily, it takes more time to flush the EVRs out of such a large chamber than it would take to flush out a small chamber. With this in mind, the TASCA separator can be operated in two modes, small-image and high-transmission mode [13]. The high-transmission mode has, as the name implies, maximum transmission through the separator. The penalty is a relatively large focal-plane image of 140 mm × 40 mm. The small-image mode focuses the EVRs into a much smaller focal plane image of only 30 mm × 40 mm. Simulations [14,15] done for ^{261}Rf EVRs produced in the ^{22}Ne on ^{244}Pu reaction indicate that the transmission in the small-image mode is 40% less than the high-transmission mode. Thus, for short-lived nuclei, the advantage of a shorter average transport-time out of a small-volume RTC will most likely out-weigh the disadvantage of the transmission loss in the separator. Furthermore, it seems reasonable to assume that the yield of a small RTC will be higher than for a large RTC, since the EVRs will spend less time in the chamber and thus be less likely to be adsorbed on the walls, etc. Two RTCs, one for each operating mode of TASCA, are being constructed and tests to investigate yields and transport times will be performed in the near future.

A new RTC-window design is being developed for TASCA [12] which will make it possible to use much thinner windows than what have been used at the BGS so far. A similar window construction should also be possible for the BGS. Thus, chemistry studies with EVRs from Pu-targets should be possible at both separators.

4 Transactinide elements available for SISAK

In Oslo, at the Centre for Accelerator Based Research and Energy Physics (SAFE), work has been undertaken to develop suitable extraction systems for the study of dubnium, seaborgium and hassium. This work is in preparation for the new possibilities emerging from the addition of using Pu targets with the BGS at LBNL and TASCA at GSI, as outlined above in Section 3.1. A brief description of the systems developed or being developed is given below.

4.1 Rutherfordium

A system to distinguish between Zr- and Hf-like behaviour was developed by Polakova et al. [16]. The group-4 elements are extracted with tri-octylamine from sulphuric acid solution into toluene. Two Rf experiments have been performed at LBNL with this system, one in 2003 and one in 2005. It was shown that Rf extracts to a lesser extent than Zr, and probably also slightly less than Hf. Preliminary extraction curves are shown in Figure 5. Full results and details can be found in [8,9,16]. The results are in accordance with recent theoretical calculations, which predict the extraction sequence Zr > Hf > Rf [17] under the given conditions.

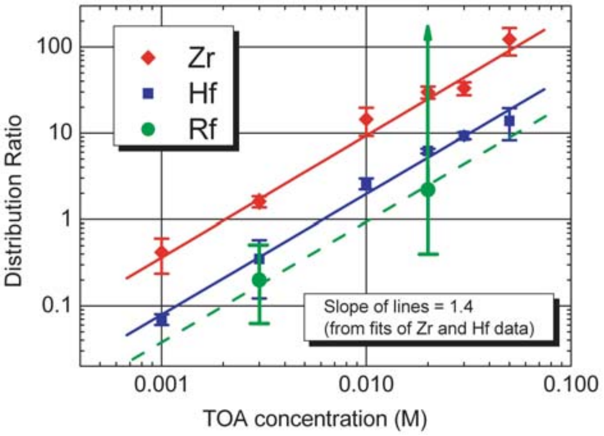

Fig. 5. (Color online) Extraction of group-4 elements from sulphuric acid using tri-octylamine in toluene with SISAK.

4.2 Dubnium

After Rf, Db is the transactinide element which can be made with the highest production rates. If Pu-targets can be used with the preseparators, then the comparatively long half-life of ^{262}Db also enhances the experimental conditions for such experiments. Zheng et al. are working on developing an extraction system which clearly distinguishes between Nb and Ta behaviour. The system is based upon extraction from sulphate solutions, in a similar way as Rf is extracted. A suitable system is expected to be ready in 2008 [18].

4.3 Seaborgium

Early work on adopting SISAK for the study of transactinide chemistry included the development of several separation systems for Sg [19–22]. These systems were not designed with preseparated reaction products in mind, as currently being used by the SISAK system, but focused on very high selectivity for group-6 elements with respect to all other elements. Today, when the input contains very little by-products, the focus is shifted toward constructing a system which yields as much chemical information as possible. One approach is to seek a liquid-liquid separation system which clearly distinguishes between the extraction, and thus e.g. complexation or redox properties, of the studied element's homologues. This approach is currently being followed by the SISAK Oslo group in a research project performed by Schultz and Opel. The aim is to develop a new separation system for the study of seaborgium, based on the experience gained in from the previous Sg separation systems and from the Rf experiments mentioned in Section 4.1. A suitable separation system is expected to be ready when preseparated Sg will become available from the TASCA and BGS separators (within 2–3 years).

4.4 Bohrium

As for the case of Sg, several separation systems were developed for Bh [23,24]. None of these systems were ever tried in Bh experiments, but extensive test with homologues were carried out. Currently, no work to develop a system that takes advantage of preseparated activity is performed. No such project is planned until results from experiments with Db and Sg are available.

4.5 Hassium

Due to the high volatility of the Hs tetroxide, it appears possible to perform experiments with Hs with low enough background without using preseparated isotopes. This has been demonstrated in several gas-phase Hs-experiments, pioneered by Düllmann et al. [25]. In such experiments an aerosol-free gas-jet is used. Hence, only volatile compounds are transported to the chemistry apparatus and a high suppression of all non-volatile elements and/or compounds is achieved. Samadani [26] has developed a SISAK system which explores the properties of HsO_4 and can be used to study the formation of $[HsO_4(OH)]^-$ and $[HsO_4(OH)_2]^{2-}$ in aqueous solutions at pH in the range 11–13 (0.001 M to 0.1 M NaOH solutions). Model experiments using on-line Os-activity and 10% O_2 addition to the He gas-jet, fit well to the relationship between the distribution ratio D and the equilibrium constants K' and K'' defined in Figure 6. The relationship, as explained in [26] is:

$$D = \frac{K_D}{1 + K'[\mathrm{OH}^-] + K'K''[\mathrm{OH}^-]^2}$$

where K_D is the distribution constant for OsO_4 (not to be confused with D, the distribution ratio D of *all* Os species). The experimental data is shown in Figure 7. If Hs behaves similarly to Os in this system, it should be possible to prove this by measuring distribution values in the range between 0.1–10.

5 Conclusions

The SISAK collaboration has performed a number of successful Rf experiments through the last years. At the same time the collaboration has pioneered the use of preseparators for transactinide chemistry experiments and enhanced the SISAK liquid-liquid extraction system and liquid-scintillation α-detectors. The experience gained during these experiments, the new TASCA separator, the use of Pu targets, and thinner RTC windows indicate that heavier elements than Rf are within reach of the SISAK system in the near future.

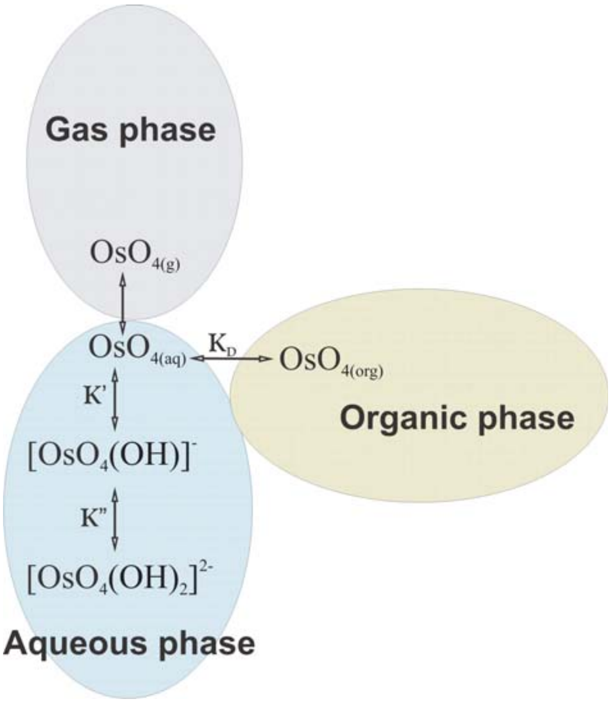

Fig. 6. A schematic drawing of the reactions that take place in the SISAK system during liquid-liquid extraction of OsO4.

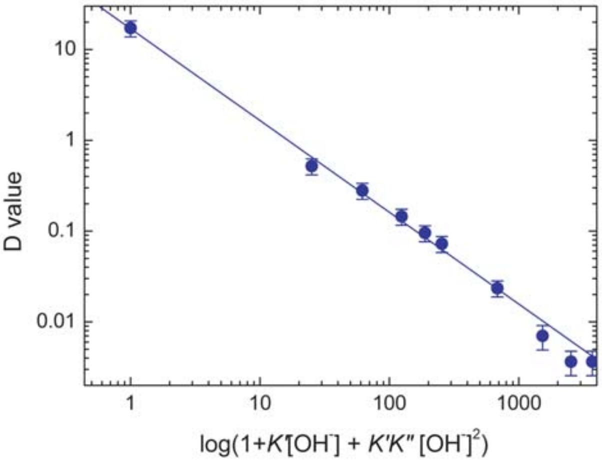

Fig. 7. Distribution ratio, D, vs. $1 + K'[\mathrm{OH}^-] + K'K''[\mathrm{OH}^-]^2$ measured for on-line SISAK extraction of OsO4 into toluene from NaOH solutions up to 0.1 M in concentration. The ionic strength was kept approximately constant by keeping all solutions 1 M in NaCl.

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