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Review of the SISAK system in transactinide research recent developments and future prospects

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Abstract

The performance of the SISAK[1] liquid–liquid extraction system applied in transactinide experiments has been improved with respect to the equipment itself and the way it is operated. The improvements were checked in on-line experiments, under conditions similar to those during transactinide experiments. As a result, the yield of the separation system was increased by a factor >5. Furthermore, a cleaner organic scintillation phase was obtained due to a better phase separation. This reduced the β-background, which disturbs the α-measurements. The sensitivity of the SISAK apparatus, including the gasjet and the detection system has been improved by more than one order of magnitude. © 1998 Elsevier Science S.A.

Keywords: Transactinides; SISAK; Liquid–liquid extraction; Rutherfordium; Chemical properties

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1. Introduction

The chemical properties of transactinides have been a matter of considerable interest in recent years. Due to the short half-lives and low production cross-sections of the transactinide elements, the methods used to study their chemical properties need to be both fast and efficient. Several systems have been developed which meet these criteria, e.g., the ARCA and OLGA systems (see [1,2] and references therein for a general discussion of the requirements and descriptions of the currently available methods). With these systems successful experiments have been performed with the elements Rf ($Z=104$), Db ($Z=105$) and Sg ($Z=106$) [1–3]. Preparations are now being made to extend the investigations to the elements Bh ($Z=107$) and Hs ($Z=108$).

SISAK is a fast liquid–liquid extraction system constructed to investigate short-lived nuclei in on-line measurments [4–6]. In this system the activity is continuously transferred from a gasjet to an aqueous phase (the mixer/degasser step), then the element of interest is selectively extracted into an organic phase (the extraction step) which is measured with suitable detectors. Phase separation is accomplished by using specially designed centrifuges. If necessary, additional purification steps can be inserted to wash the organic phase or to strip coextracted elements, which otherwise would disturb the measurement.

The small volume of the centrifuges, about 0.3 ml, and flow rates up to 2 ml s^{-1} ensure that SISAK is a very fast system. The minimum transport time from the mixer inlet to the detector is about 1 s. The continuous and fast operation makes SISAK well suited for chemical studies of transactinide elements involving nuclei with half-lives below 1 min produced with a rather low cross-section.

For the investigation of transactinide elements with SISAK various chemical separation systems have been worked out [7]. Parallel to this work, a detection system based on liquid scintillation counting has been developed for on-line detection of time-correlated α-decays in the organic phase [8]. To reduce the β-background in the α-spectra, pulse-shape discrimination is applied [9].

Several successful on-line tests were done with the homologs of the transactinides to optimize the operation of the adopted chemical separation systems. Then experiments with Rf, Db, and Sg were performed. Unfortunately,

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[1] *S*hort-lived *I*sotopes *S*tudied by the *AK*UFVE technique.

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these experiments did not identify unambiguously the transactinide elements [10–12].

Due to the low production rate and a rather high background, observation of mother–daughter time-correlations is needed to claim the detection of a transactinide element. In experiments performed at the 88″ cyclotron at the Lawrence Berkeley National Laboratory it was shown that the SISAK system, using the chemistry developed for Db, allows the measurement of the 0.85 s α-emitter ^{224}Pa [12] (Pa is a ‘‘pseudohomolog’’ of Db). This indicates that SISAK in connection with the detection unit and the analyzing technique is working. However, this experiment also showed that the overall yield was much lower than expected.

Therefore, a thorough examination of the complete system was undertaken to find ways to increase the yield and/or the sensitivity. The results obtained are reported below.

2. Enhancements of the SISAK performance

The transactinide experiments with SISAK pointed out several problems:

1. The phase separation was not working satisfactorily all the time: frequently the aqueous phase formed a milky-white emulsion. In the worst case up to 50% of the organic phase was lost this way, but usually the loss was between 20 and 30%.
2. The bad phase separation also affected the organic phase, resulting in ‘‘quenching’’ and a high β/γ-background due to contaminations.
3. The distribution coefficient (‘‘*D*-value’’) for various elements was lower than previously observed.
4. The back pressure in the α-detection cells was too high. This resulted in overflow in the centrifuge in the last stage, and thus loss of some organic phase. To reduce this effect, the rotation speed of the centrifuge had to be increased in order to increase the pumping capacity. Unfortunately, this enhanced the formation of the emulsion mentioned above.
5. The transfer of the activity attached to the KCl-cluster in the gasjet into the aqueous phase was not efficient enough.

To solve these problems, a series of on-line experiments were performed at the TRIGA reactor in Mainz and at the Oslo Cyclotron. Using Zr- (Mainz) and Hf-activity (Oslo) it could be shown that with an extractive scintillator (0.25 M dibutylphosphate (DBP), 2.5 g l^{-1} dimethyl-POPOP, and 80 g l^{-1} naphthalene dissolved in toluene) the distribution coefficient decreases and bad phase separation occurs. This is illustrated in Fig. 1 where the extraction of Hf from 2 M HNO$_3$ with 0.25M DBP in toluene is compared with the extraction using an ‘‘extractive scintillator’’. Furthermore, without the scintillation cocktail the milky-white emulsion was no longer observed and the phases were clean. Therefore, changes were made to the SISAK setup which allow the scintillation ingredients to be added after the last extraction step (see Fig. 2). In order not to increase the volume of the outgoing organic phase more than necessary, this must be done with a flow as low as possible. Because the solubility of naphthalene is only 200 g l^{-1} toluene, 1-methyl-naphthalene was used instead. Tests have shown that the scintillation yield and pulse shape discrimination are not influenced by this change. A mixing ratio of 0.4 ml s^{-1} organic phase and 0.1 ml s^{-1} scintillator demands 3.1 M 1-methyl-naphthalene in toluene to obtain equal properties as with 80 g l^{-1} (0.62 M) naphthalene solution. In summary, the problems 1, 2, and 3 were solved completely by adding the scintillation ingredients after the extraction step.

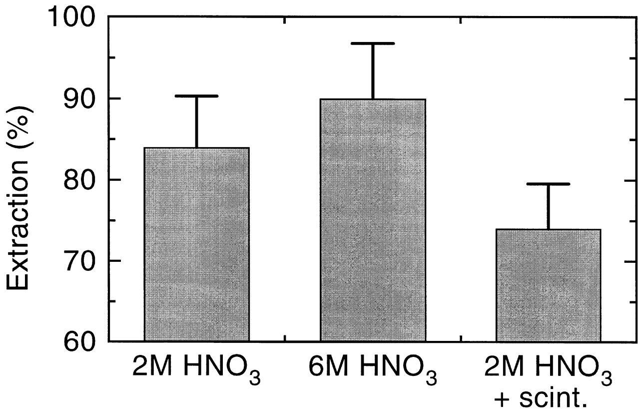

Fig. 1. On-line extraction of Hf from nitric acid solution into 0.25 M DBP dissolved in toluene. The bar on the right shows the yield from 2 M HNO$_3$ when the scintillation components are present in the organic phase.

An extra ‘‘booster’’ centrifuge was inserted after the last extraction step to solve problem 4. This centrifuge is not working as a separation centrifuge and therefore it can be operated at a much higher rotational speed, which in-

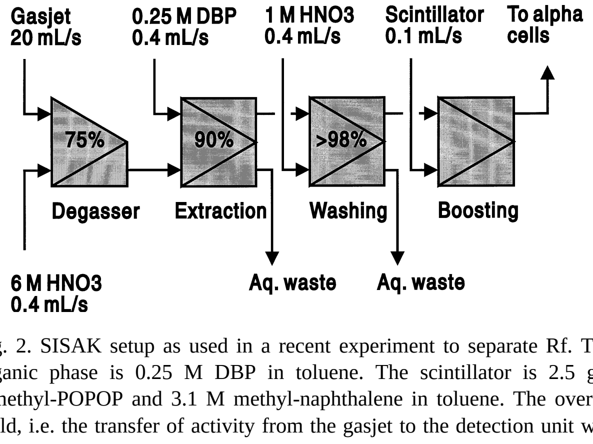

Fig. 2. SISAK setup as used in a recent experiment to separate Rf. The organic phase is 0.25 M DBP in toluene. The scintillator is 2.5 g/l dimethyl-POPOP and 3.1 M methyl-naphthalene in toluene. The overall yield, i.e. the transfer of activity from the gasjet to the detection unit was measured with Hf-activity to be better than 66%. The yield in percent for each step is indicated.

creases the pump capacity enough to overcome the back pressure in the measuring cells. It also serves as a mixer for the organic phase and the scintillator.

The on-line extraction of group IV elements increases with increasing HNO_3 concentration (see Fig. 3). The Zr data in Fig. 3 is taken from Ref. [13] and confirmed by the recent measurements. Unfortunately, HNO_3 extracted into the organic phase gives a strong color quenching. Since the amount of HNO_3 in the organic phase increases with the acid concentration in the aqueous phase, the need for optimum scintillation performance prevents extraction at high acid concentrations. However, it was observed that though the *D*-value for DBP extraction of the Rf homologs Zr and Hf from 2 M HNO_3 is lower than for 6 M HNO_3 (Fig. 3), the back extraction is kinetically too slow to cause any activity loss from the organic phase. Thus, if the organic phase already contains the activity of interest, practically nothing of it will be transferred to the aqueous phase during the less than 100 ms contact time in the washing step. Therefore, the organic phase can be washed with 1–2 M HNO_3, although the extraction is done from 6 M HNO_3.

These results led to the SISAK setup for the chemical separation of Rf as shown in Fig. 2. Here selective extraction of the group IV elements is performed from 6 M HNO_3 with 0.25 M DBP in toluene, then the organic phase is washed with 1 M HNO_3 and finally it is mixed with the scintillator in the ‘‘booster’’ centrifuge. Together with the scintillator ingredients a small amount (10 ml l^{-1}) of trioctylamine (TOA) is added. This addition removes the slightly yellow colour in the mixed scintillator/organic phase and prevents color quenching.

Very recently this scheme was used in an experiment at the Paul Scherrer Institute (PSI) in Villigen, Switzerland to isolate Rf produced in the reaction $^{248}Cm(^{18}O,5n)^{261}Rf$. The data has not yet been analyzed completely but preliminary results indicate that Rf may have been detected.

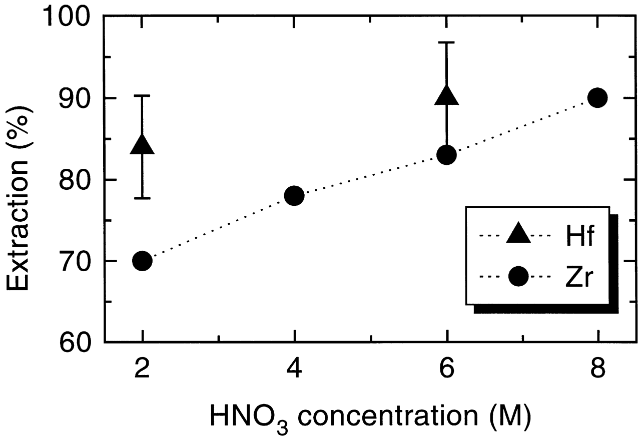

Fig. 3. Extraction of Zr and Hf from nitric acid solution into 0.25 M DBP in toluene.

A new mixer was constructed to improve the gasjet/aqueous phase mixer yield. Experiments with this mixer at the Oslo cyclotron gave a 75±8% yield for an aqueous phase with a flow rate of 0.5 ml s^{-1} and gasjet flow of 20 ml s^{-1}. If the gasjet flow was increased to 60 ml s^{-1}, the yield decreased to about 60±6%. Work is in progress to modify the mixer to enable higher gasjet flow rates.

3. Conclusion

By adding the scintillator components after the last extraction centrifuge, and by improving the yield of the gasjet mixer, the overall yield, i.e. the amount of group IV activity transferred from the gasjet to the detectors, has increased by at least a factor of five. This, together with a reduced β-background in the α-spectra, resulting from a more efficient phase separation, has increased the sensitivity of the SISAK system by a factor >10. This should make it possible to investigate the chemical properties of Rf and Db with the SISAK system. Further improvements, especially to the detection system, should enable the use of SISAK in experiments on even heavier elements. This is a task well suited for the SISAK system, as it is fast and easy to operate continuously over a long period.

Acknowledgements

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