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.

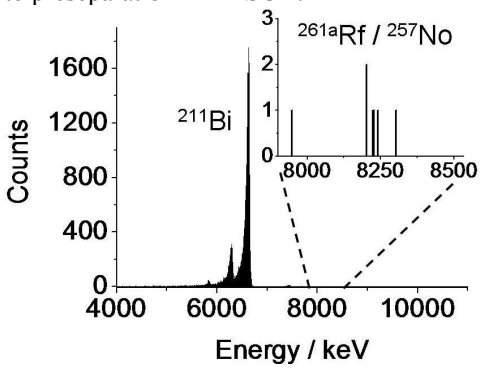

Figure 1: Sum spectrum of the α-particle events of ^{261a}Rf.

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.

References

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