

K_d -Value Determination for ^{261}Rf (Element 104) with the Multi-Column Technique

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After online experiments with ^{165}Hf ($t_{1/2} = 76$ s, decaying into ^{165}Lu) produced in the reaction $^{158}\text{Dy}(^{12}\text{C}, 5n)$ at UNILAC at GSI [1], we attempted the first transactinide experiment using the multi-column-technique [2] in the system HF/0.1 M HNO_3 . In this technique, the KCl aerosol particles transporting the activities from the target chamber in a gas-jet are continuously dissolved in a degasser unit in an aqueous solution which is continuously pumped through three consecutive columns. The achieved degasser yield is about 80%.

The solution from the degasser unit is pumped with a chemically inert HPLC-pump through the three columns with resin in HPLC quality. The transport time between the degasser unit and the first column through capillary of 0.3 mm inner diameter and a distance of 64 cm is about 6 sec. The hold-up time in the degasser is about 2 sec.

The first column (F), filled with a cation exchange resin (Dowex 50WX8, -400 mesh), acts as a filter for all decay products of rutherfordium-261 (see decay scheme in Fig. 1), while the latter passes through. The second column (C), filled with an anion exchange resin (Dowex 1X8, -400 mesh), is the true chromatographic column where Rf experiences a retention time, t_R , being of the order of its nuclear half life, $t_{1/2}$. The t_R can be manipulated by changing the mass of the resin. The daughter nuclides that are formed during the retention time of Rf on column C are strongly retained on column D (daughter activity, A_D), filled with a cation exchange resin (Dowex 50WX8, -400 mesh). The part of Rf that passes C is also passing the third column D without notable retention. The final eluate (E) is sampled in order to collect the residual atoms as well as its decay products (daughter activity A_E).

After the experiment, the activities of the decay products on column D and in the final eluate E are measured and the retention time is obtained according to (1). The distribution coefficient, K_d , of rutherfordium in column C may then be obtained from (2)

$$t_R = \frac{t_{1/2}}{\ln 2} \ln \left(\frac{A_D + A_E}{A_E} \right) \quad (1); \quad K_d = (t_R - t_0) \frac{V}{m} \quad (2)$$

where

t_0 = column hold-up time due to its dead volume [s]

V = flow rate of the mobile phase [$\text{ml} \cdot \text{s}^{-1}$]

m = mass of the stationary phase [g].

The preparation of the samples is done by the evaporation of the eluate E and the activity A_B stripped from the Columns D with 0.5 M α -HIB (at pH 3.8). For the determination of the chemical yield, ^{241}Am ($E_\alpha=5.5$ MeV) is used. The samples are prepared by molecular plating on thin Ti-foils ($420 \mu\text{g}/\text{cm}^2$) with about 80% chemical yield (700V, 2-3 mA/cm^2 , pH 2-3, isopropanol). The α -decay of ^{253}Es ($E_\alpha=6.6$ MeV) can be measured by 4π alpha-spectrometry. The background count rate

in the range of 6.4 to 6.8 MeV is 1.5 events per week with our detectors.

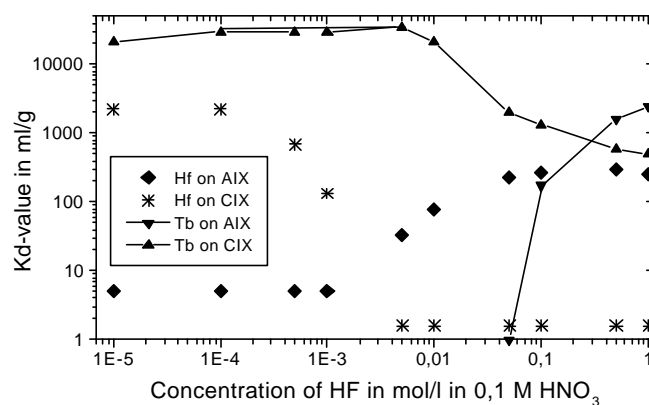
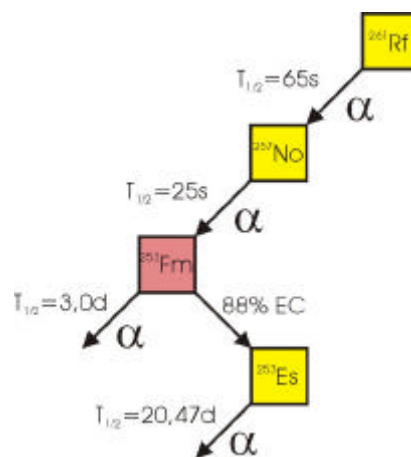


Fig. 1 Decay scheme for ^{261}Rf

Fig. 2 K_d -values of Hf and Tb in variable HF/0.1 mol/l HNO_3 on DOWEX 1X8 (AIX) und DOWEX 50WX8 (CIX)

As shown in Fig.2, the K_d -values of trivalent metal ions (Tb^{3+}) are in excess of 10000 in 0.1 M HNO_3 for HF concentrations up to 10^{-2} M and decrease for higher HF concentrations due to the formation of fluoride complexes (3500 in 0.05 M HF and 700 in 0.5 M HF). The K_d -value in 0.5 M HF/0.1 M HNO_3 was also verified for ^{250}Fm produced on-line in the $^{238}\text{U}(^{18}\text{O}, 6n)$ reaction at PSI. This decrease of the K_d -values was ignored in [3]. For ^{261}Rf , K_d -values on an AIX were determined with ARCA to be 7 ± 5 ml/g in 0.5 M HF/0.1 M HNO_3 and 112 ± 84 ml/g in 0.05 M HF/0.01 M HNO_3 [4]. This difference is tentatively attributed to the competition of the counter ion NO_3^- for the binding sites on the AIX. The Rf experiments with the multi-column technique will test this interpretation.

References

- [1] A. Kronenberg et al.: GSI annual report 1998, p. 171
- [2] A. Kronenberg et al.: Jahresbericht Mainz, 1998, p. 8

- [3] G. Pfrepper et al., Radiochim. Acta 80, 7-12 (1998)
- [4] E. Strub, J.V. Kratz, A. Kronenberg et al.,
Radiochim. Acta, submitted