A10

Fluoride Complexation of Rutherfordium (Rf, Element 104)

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It had been shown experimentally [1,2] that 261 Rf is not eluted from the cation exchange (CIX) column under the condition of previously performed seaborgium (Sg, element 106) experiments [3,4] and can only appear in the Sg sample as a result of the decay of 265 Sg.

The distribution coefficient (K_d) of ²⁶¹Rf on cation and anion exchange resins at various HF concentrations had been studied [1,2]. Rf was eluted at higher HF concentrations from CIX than Zr and Hf and rather resembled Th. Up to 0.05 M HF, Rf was not retained on the AIX.

Now, the K_d values of Rf at HF concentrations > 0.05 M (in 0.1 M HNO₃) have been determined to establish at which concentration the K_d value on AIX is increasing indicating that anionic complexes of Rf are being formed.

²⁶¹Rf was produced in the ²⁴⁸Cm(¹⁸O,5n) reaction at the PSI Philips Cyclotron. A 730 μ g/cm² ²⁴⁸Cm target was bombarded with a 0.5 μ A_{part} ¹⁸O⁵⁺ beam. The target contained 10% Gd thus producing simultaneously short-lived Hf isotopes. Rf and Hf were transported by a He(KCI) gas jet and collected for 90 s by impaction on a slider in the Automatic Rapid Chemistry Apparatus ARCA II. The residue was dissolved in 200 μ I 0.1 M HNO₃/x M HF (x variable) and fed onto the AIX. The effluent was evaporated to dryness as sample 1. In order to elute remaining Rf from the column, a second fraction (200 µl) was collected which is known to elute aroup 4 elements from the column (5 M HNO₃/0.01 M HF). This fraction was prepared as sample 2. 78-s ^{261}Rf was detected by $\alpha\text{-spectroscopy.}$ The counting time was 12 min. Every 8th pair of samples was monitored by additional γ -spectroscopy to determine the distribution of Hf.

The data (see figures) show that Rf behaves different from Zr and Hf and seems to resemble Th on both AIX and CIX (dotted lines). It is remarkable, that the Hf online data is in agreement with the Hf offline data while the Rf behaviour is clearly different.

On the CIX, the four elements are strongly retained as cations below 10^{-3} HF. For Zr and Hf, the fall of the K_d values due to the formation of fluoride complexes occurs between 10^{-3} M HF and 10^{-2} M HF. For Rf and Th, this fall is observed at one order of magnitude higher HF concentrations. On the AIX, for Zr and Hf, a rise of the K_d values due to the formation of anionic fluoride complexes is observed in the same range of HF concentrations where the fall on the CIX is observed, yielding a consistent picture. For Rf and Th, on the AIX, no rise of the K_d values is observed even if the HF concentration is increased up to 1 M.

By varying the concentration of the counter ion NO_3^- which is competing for the binding sites on the AIX resin, it could be shown, nevertheless, that Rf does form anionic fluoride complexes (i.e. the K_d value varies with the nitrate concentration). This is in agreement with reported results [5,6,7].

We conclude that the observerved behaviour of Rf is a result of a specific competition with NO_3^- on the AIX.



Fig.1: Sorption of Zr, Hf, Th and Rf in 0.1 M HNO₃ on a CIX resin (Aminex A6) at various HF concentrations.



Fig.2: Sorption of Zr, Hf, Th and Rf in 0.1 M HNO₃ on a AIX resin (Riedel de Haen) at various HF concentrations.

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