

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

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After online experiments with ¹⁶⁵Hf ($t_{1/2} = 76$ s, decaying into ¹⁶⁵Lu) at GSI [1] we performed two experiments with the multi-column-technique [2] with Rutherfordium.

²⁶¹Rf was produced in the ²⁴⁸Cm(¹⁸O, 5n) reaction at the PSI Philips Cyclotron. A 730 $\mu\text{g}/\text{cm}^2$ ²⁴⁸Cm target was bombarded with a 0.5 $\mu\text{A}_{\text{part}}$ ¹⁸O⁵⁺ beam. The KCl aerosols transporting the activities was continuously dissolved in a degasser with a yield > 80% at a tolerable pressure in the target chamber of 1.7 bar at a gas-flow rate of 1.6 – 1.7 L/min and a distance between the target chamber and the degasser of approximately 50 m. The hold-up time of the activity in the degasser was about 2 s. A solution-level of 50 to 150 μL was kept at the bottom of the degasser through control by a light barrier which caused, if activated, an adjustment of the flow rate from the HPLC pump that pumped the solution into the degasser. The active solution leaving the degasser was pumped by a double-piston HPLC pump at a flow rate of 1 mL/min through a capillary of 0.3 mm inner diameter to three columns connected sequentially: A cation filter followed by the true chromatographic column filled with an anion-exchange resin, the latter followed by another cation filter.

Two experiments were performed, one using 0.5 M HF/0.1 M HNO₃ as the mobile phase, another one using 0.01 M HF (without any HNO₃). The running time was 24 hours in both cases. In the first case, the cation-exchange columns were filled with 330 mg of DOWEX 50WX8, minus 400 mesh. These filters were used for 3 hours and were then replaced by new ones. This ensured that there was no break-through by any of the descendants of ²⁶¹Rf, i.e. ²⁵⁷No, ²⁵³Fm, and ²⁵³Es. The anion-exchange column was filled with 50 mg of DOWEX 1X8, minus 400 mesh in the nitrate form.

17 mg of DOWEX 1X8 in the fluoride form was used in the experiment with 0.01 M HF. In this experiment, 68 mg of DOWEX50WX8 were used as cation filters for running times of 4 hours. The effluent from the final cation filter was collected in a reservoir (activity AE). After the end of the experiment, the final cation filter was stripped with 5 M HNO₃ (activity AD). The solutions AE and AD were spiked with ²⁴¹Am. After adjusting its pH to 2, the solution containing AE was passed over a cation-exchange column where ²⁵³Fm and ²⁵³Es were sorbed. This column and the cation-exchange columns containing AD were stripped with 5 M HNO₃ and the solutions were evaporated to dryness. The residues were dissolved in 8 M HCl and iron (from the steel capillary of the gas jet) was extracted into Aliquat 336 in chloroform. After evaporation to dryness of the aqueous phases, the residues were dissolved in 10 μl of 0.1 M HNO₃ and transferred into isopropanol for molecular plating. The latter was performed with 1200 V onto thin Ti foils of 430 $\mu\text{g}/\text{cm}^2$ thickness. These were subject to α spectroscopy in 4- π -geometry for more than 60 days, three half-lives of ²⁵³Es.

Overall chemical yields determined via the ²⁴¹Am activity ($E_\alpha=5.5$ MeV) were 65% on the average. The background in the relevant energy window near 6.6 MeV was 1.5 α events per week.

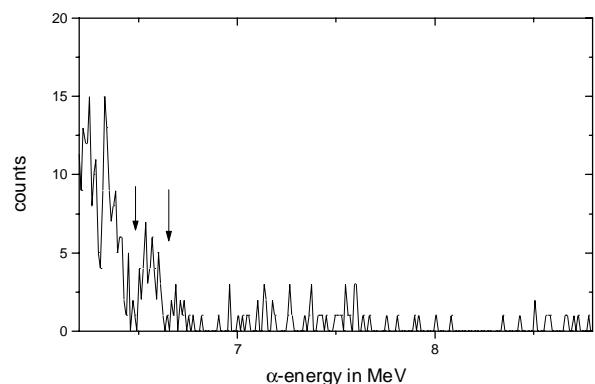


Fig 1: α -energy spectrum of sample AD in experiment 2 with the ²⁵³Es energy window indicated by arrows.

In the first experiment using 0.5 M HF/ 0.1 M HNO₃, the entire ²⁵³Es activity (80 events) was found in AE and no activity above background in AD indicating that the retention time of rutherfordium on the anion-exchange column must have been very short. From the activities, a $\underline{\mathbf{K_d = 0^{+3}-0 \text{ mL/g}}}$ results in **0,5 M HF / 0,1 M HNO₃**. This is lower than the K_d value of ~5 determined with ARCA [3]. This is no major problem as it is known that K_d values < 10 are difficult to determine with ARCA and tend to be systematically high.

In the second experiment using 0.01 M HF (without HNO₃), we observed 90 events attributable to ²⁵³Es in AD while no events above background were detected in AE. This leads within 1 standard deviation to a K_d value of $\underline{\mathbf{394^{+100}-100 \text{ mL/g}}}$ in good agreement with data taken off-line by ARCA [4]. The error limits on the K_d values were determined by a Monte-Carlo simulation assuming a half life of ²⁶¹Rf of 78^{+11}_{-6} s [5] and a background due to electronic noise of 1.5 events per week in the interesting energy region ($E_\alpha=6.6$ MeV).

These on-line experiments with ²⁶¹Rf corroborate the interesting finding [3,4] that the sensitivity of Rf fluoride complexes to the presence of the counter ion NO₃⁻ is much more pronounced than for the fluoride complexes of the lighter homologs Zr and Hf.

References

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