High-activity ^{172}Hf/^{172}Lu generator

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^{172}Lu is an isotope of the heaviest rare-earth element which decays to stable ^{172}Yb with an half-life of 6.70 days. As it is formed by the decay of its long-living ($T_{1/2}$ = 1.87 yr) ^{172}Hf parent, the development of an isotope generator remains an interesting subject of radiochemistry research.

Radiohafnium: Hafnium isotopes were produced in a massive (195 g) tantalum target which was installed as a proton-neutron converter in UC-target of the ISOLDE facility at CERN and had received a total dose $2.5 \cdot 10^{18}$ protons of 1 GeV energy [1].
The tantalum target was dissolved $HF_{(conc)}$ + HNO_3 solution and the radiolanthanides were isolated. About one tenth of the remaining tantalum solution was used to isolate ^{172}Hf. The fraction was passed through a series of anion-exchange columns (Dowex 1×8), starting with a big column of $V_0 \approx 150$ ml. While tantalum is absorbed, radiohafnium remains on the resin. Every eluate fraction was evaporated to dryness and subsequently transferred to the next, smaller column using 9 M HF + 1.2 M HNO_3 solutions. An average Ta / Hf separation factor of 50 was determined, thus resulting in an amount of < 100 ng of tantalum.

Isotopic purity: Apart of ^{172}Hf with 85.1 MBq, ^{175}Hf ($T_{1/2}$ = 70 d) activity amounted to 35.2 MBq. As chemical analogue, 2.2 MBq of ^{88}Zr ($T_{1/2}$ = 83.4 d) were detected. The tungsten isotopes ^{178}W ($T_{1/2}$ = 22 d) and ^{181}W ($T_{1/2}$ = 121 d) were not detected, because the have been separated in the curse of the Ta / Hf anion exchange purification.

Generator: This fraction was loaded onto a final extraction-chromatography generator column in 4 M HCl solution. The column of dimensions 0.4×8 cm ($V_0 \approx 0.5$ ml) was filled with supporting silicagel 45-60 μm impregnated with di-(2-ethylhexyl) phosphoric acid (HDEHP) (0.5 ml HDEHP per 1 gram of dried silikagel) [3]. An estimation of distribution coefficients for extraction system of HDEHP/HCl-aqueous solutions [2] showed that the separation of Lu from Hf may be achieved optimally with 6-7 M HCl.
The profile of the first elution is illustrated in Figure 1. All ^{172}Lu is eluted with 6.5 M HCl. For monitoring of the radionuclidic purity and absolute ^{172}Lu activity gamma spectrometry was used. Furthermore, a quality control concerning the total amount of trivalent cations was performed by means of the thin-layer chromatography via the labelled compound ^{172}Lu–DOTA-octreotide. The results indicated that the ^{172}Lu is produced in ultra-low concentration with high specific activity. No hafnium isotopes were found in the eluate.

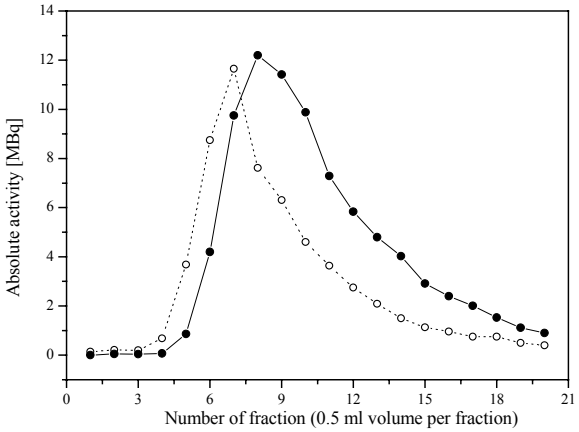

Figure 1. Closed circles represent the first elution after genrator loading; opened circles elution 6 months after

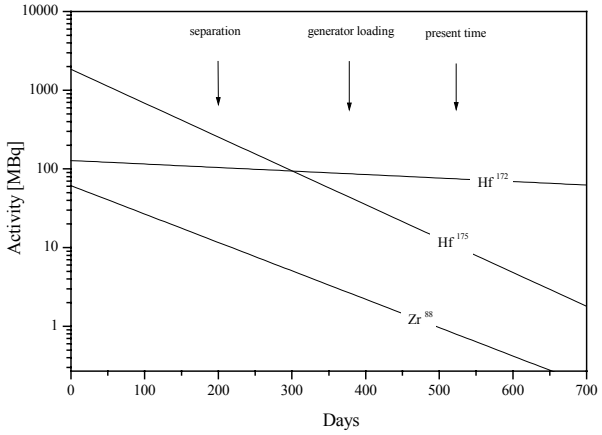

Figure 2. The decay of present isotops

For a high-activity generator the long-term stability is an important parameter. The presented system provides a radiation dose of 4.3 mSv/h, 480 μSv/h and 60 μSv/h at distances of 5, 15 and 50 cm, respectively. The elution 6 months after the generator loading is also given in Figure 1. It was determined that the performance of the system remains unchanged within that period.

Conclusion: The presented ^{172}Hf/^{172}Lu generator was found to be a convenient and effective system to provide ^{172}Lu. It appeared to be an optimum time for the preparation of the generator of about one year after the target irradiation due to the decay of ^{175}Hf, the radiotungsten isotopes, ^{88}Zr and ^{44}Ti Figure 2. Eluate fractions are thus almost radionuclidic pure. The high specific activity of the daughter nuclide alongside with the long-term stability of the system makes it suitable for several scientific studies.

[1] A. F. Novgorodov et al., COST chemistry, Action D18, Heidelberg (2002). [2] T.Braun and G.Ghersini (Eds) in "Extraction Chromatography", Vol. 2, Elsevier, New York (1975). [3] N.A. Lebedev, E. Herrmann, J. Echn, Radiochimia 26 (1984) 223