High-activity $^{172}\text{Hf} / ^{172}\text{Lu}$ generator

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$^{172}\text{Lu}$ is an isotope of the heaviest rare-earth element which decays to stable $^{172}\text{Yb}$ with a half-life of 6.70 days. As it is formed by the decay of its long-living ($T_{1/2} = 1.87$ yr) $^{172}\text{Hf}$ 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 $2.5 \times 10^{18}$ 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 $^{172}\text{Hf}$. The fraction was passed through a series of anion-exchange columns (Dowex 1$^\times$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 $^{172}\text{Hf}$ with 85.1 MBq, $^{175}\text{Hf}$ ($T_{1/2} = 70$ d) activity amounted to 35.2 MBq. As chemical analogue, 2.2 MBq of $^{88}\text{Zr}$ ($T_{1/2} = 83.4$ d) were detected. The tungsten isotopes $^{178}\text{W}$ ($T_{1/2} = 22$ d) and $^{181}\text{W}$ ($T_{1/2} = 121$ 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 $0.4 \times 8$ 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 $^{172}\text{Lu}$ is eluted with 6.5 M HCl. For monitoring of the radionuclidic purity and absolute $^{172}\text{Lu}$ 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 $^{172}\text{Lu}$–DOTA-octreotide. The results indicated that the $^{172}\text{Lu}$ is produced in ultra-low concentration with high specific activity. No hafnium isotopes were found in the eluate.

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 $^{172}\text{Hf} / ^{172}\text{Lu}$ generator was found to be a convenient and effective system to provide $^{172}\text{Lu}$. 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 $^{175}\text{Hf}$, the radiotungsten isotopes, $^{88}\text{Zr}$ and $^{44}\text{Ti}$ 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.