Development of production method and radiochemical separation of no-carrieradded ⁹⁰Nb from bulk amount of ^{nat}Zr.

V. Radchenko¹, S. Busse¹, H. Hauser², M. Eisenhut², F. Rösch¹

¹ Institute of Nuclear Chemistry, Johannes Gutenberg-University Mainz, Fritz-Strassmann-Weg 2, D-55128 Mainz, Germany; ² Radiopharmaceutical Chemistry, German Cancer Research Center, Im Neuenheimer Feld 280, 69120 Heidelberg, Germany

Introduction: The positron emitter ⁹⁰Nb is a promising radioisotope for applications in positron emission tomography (PET). One feature of ⁹⁰Nb is the half-life (14.6 hours), which enables this isotope for the visualization of processes with medium and slow kinetics, such as the pharmacokinetics of nanobodies, polymers and antibodies. Another advantage of ⁹⁰Nb is its decay parameters: relatively high positron branching of 51% and a rather low β^+ -energy of E_{mean}=662 keV (E_{max}= 1.5 MeV), which allows high quality and high resolution images even at low amounts of ⁹⁰Nb.

Experimental: ⁹⁰Nb was produced via the ⁹⁰Zr(p,n)⁹⁰Nb reaction at the cyclotron MC32NI of the German Cancer Research Center Heidelberg. For irradiation, three discs of natural zirconium (51,45% ⁹⁰Zr) foil of 10 mm diameter and a thickness of 0,25 mm were used (Figure 1). Irradiation was performed with 20 MeV protons (17.5 MeV at 1st foil), current 5 μ A for 1 hour. 24 hours after irradiation, the production yield was measured by gamma spectroscopy.

The separation procedure follows partially the procedure described by S. Busse *et al.* [1]. The zirconium metal target (240-360 mg) was transferred into a 50 ml vial and water (2 ml) was added. Under cooling in a ice bath, 0.63 ml of 48% HF was added in small portions. After complete dissolution, 6 ml 10M HCl and 3.4 ml saturated boric acid were added. The ⁹⁰Nb fraction was extracted with 5 ml of 0.02M of *N*-benzoyl-*N*-phenylhydroxylamine (BPHA) in CHCl₃ by vigorous stirring of the two phases in a 50 ml vial for 20 min. The aqueous phase additionally washed with 3 ml CHCl₃. The organic phases were combined and washed with 2 ml of a mixture of 9M HCl/0,001M HF and with 2 ml of pure 9M HCl. The re-extraction was carried out with 5 ml aqua regia.

For a final separation of ⁹⁰Nb from trace amounts of zirconium an anionic exchange method was employed. The aqueous phase after re-extraction was evaporated to dryness. The residue dissolved in 0.5 ml of a mixture of 0.25M HCl/0.1M oxalic acid and adsorbed on a small Aminex A27 15±2 μ m anionic exchange column (20 x 1.5 mm). Elution was performed under slight overpressure of 0.3 bar. After loading, the column was washed with 100 μ l 10M HCl. Residues of Zr were removed by washing with 200 μ l of a mixture of 9M HCl/0,001M HF. ⁹⁰Niobium was eluted with 200 μ l of a mixture of 6M HCl/0,01M oxalic acid.



Figure 1. Setup for irradiation: A.- Schematic view of the target holder, B.- The target holder, C.- Zirconium discs Ø 10 mm for irradiation.

Results: Production yield of ⁹⁰Nb under given irradiation parameters was 145 MBq/µAh. The distribution of activity in the foils was: 49% (1st foil), 40% (2nd foil) and 10% (3rd foil). The results show, that the third foil has only 10% of activity and 30% of the mass of the whole target. To increase the separation factor, only foils 1 and 2 should be used for work-up and separation. The isotopic purity of ⁹⁰Nb was more then 96%. Minor isotopic impurities were found: ⁸⁹Zr (0.29%), ^{92m}Nb (1.79%), ⁹⁵Nb (0.42%) and ⁹⁶Nb (0.74%). All impurities of zirconium isotopes were efficiently removed by the separation procedure. The extraction steps allow crude separation of ⁹⁰Nb from target material. In the organic phase more than 99% of ⁹⁰Nb was found. After the re-extraction procedure the 5 ml aqueous phase contained 90-95% of the 90 Nb activity. This led already to a high separation factor for Zr/Nb after both extractions of 10⁴. The final anionic exchange separation allowed complete separation from residues of the target material. The final 200 µl of 6M HCl/0,01M oxalic acid contained 80-85% of the activity of ⁹⁰Nb. The final separation factor for Zr/Nb was $>10^{7}$ and the final yield of ⁹⁰Nb was 76-81%.

References

 Busse S., Brockmann J., Rosch F. (2002) Radiochemical separation of no-carrier-added radioniobium from zirconium targets for application of ⁹⁰Nb-labelled compounds. *Radiochim. Acta*, 90:411–415.