Radiochemical Separation of No-Carrier-Added ^{177}Lu as Produced via the ^{176}Yb(n,g)^{177}Yb ® ^{177}Lu Process

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The β^- emitter ^{177}Lu ($T_{1/2}$ = 6.71 d, max. and aver. β^- energies of 421 and 133 keV) is a promising therapeutic radioisotope for the curative treatment of cancer using labelled proteins [1-4]. The decay is accompanied by the emission of low energy γ-radiation with E_γ = 208.3 keV (11.0%) and 113 keV (6.4%) suitable for simultaneous imaging. Moreover, ^{177}Lu attracted a special interest because of the very high cross section of 2100 barn of the ^{176}Lu(n,γ)^{177}Lu production process. Irradiation of 100 mg of ^{nat}Lu at reactors providing 10^{14} n cm^{-2}s^{-1} for 100 h yields specific activities of 1.15 GBq/µmol, which can be increased by a factor of 35 in the case of 95% isotopically enriched ^{176}Lu. Nevertheless, a minimum amount of stable ^{176}Lu cannot be avoided and might cause some problems concerning the labelling of tumour affine biomolecules. Thus, a no-carrier-added (nca) ^{177}Lu seems to be useful, providing the maximum specific activity of 720 GBq/µmol ($1.1\cdot10^5$ Ci/g).

For this purpose, the alternative production route ^{176}Yb(n,γ)^{177}Yb ($T_{1/2}$ = 1.9 h) —β^-→ ^{177}Lu was investigated, providing a nca state of ^{177}Lu. It was the aim of this work to develop an efficient separation of nca ^{177}Lu from macroscopic amounts of the ytterbium target material despite of the chemical similarity of these neighboured lanthanides. The separation of the nca ^{177}Lu from the macro-amounts of the ytterbium target based on the cementation process, i.e. the selective extraction of Yb by Na(Hg) amalgam from Cl⁻ / CH_3COO^- electrolytes [5-8] followed by a final cation exchange purification.

^{177}Lu was produced in a neutron capture reaction on natural or isotopically enriched ytterbium. The isotopic composition of isotopically enriched ^{176}Yb was 0.0034% ^{168}Yb, 0.114% ^{170}Yb, 0.634% ^{171}Yb, 1.157% ^{172}Yb, 1.014% ^{173}Yb, 2.355% ^{174}Yb and 94.72% ^{176}Yb. 200 mg (99.9999% chemical purity) Yb_2O_3 were irradiated for 6 h at the TRIGA II reactor Mainz at a neutron flux of $2\cdot10^{12}$ n cm^{-2}s^{-1}. 12.4 mg of enriched ^{176}Yb-Yb_2O_3 were irradiated for two days at the HMI neutron source BERII at $2\cdot10^{14}$ n cm^{-2} s^{-1}, resulting in 8.1 GBq ^{177}Lu at one day after EOB.
Sodium amalgam was prepared via electrolysis of a 20% solution of NaOH, as described elswhere [8]. 200 mg Yb_2O_3 were dissolved in 1.4 ml 4 M HCl. Next, 3 ml 4.5 M CH_3COONa and H_2O were added to a total volume of 6 ml of pH ≈ 3.4. 4 ml of Na(Hg) amalgam (0.4% Na) were added and this system is stirred for 90 sec. The amalgam is removed from the system. After 4 of these cycles, about 99% of the ytterbium were removed from the aqueous solution. The nca ^{177}Lu is isolated from this solution by precipitation as the hydroxide using 4 M NaOH. The hydroxide is isolated by centrifugation and dissolved in 2.5 ml 0.1 M HCl. After adding of 2.5 ml 4.5 M CH_3COONa, another 4 cementations are performed in a new vessel. After this procedure, the amount of $Yb^{(III)}$ is reduced to about 0.01-0.02 % of the initial mass, cf. Figure 1, while about 85±5% of the nca ^{177}Lu are remaining in the solution.

In conclusion, the radiochemical separation process developed provides radiochemically pure nca ^{177}Lu within a total volume of less than 0.5 mL with an overall separation yield of 75±5% within 4-5 h, with Yb contaminations of < 10^{-6} %, i.e. < 1 ng $Yb^{(III)}$ for a 100 mg ^{176}Yb target. It thus can be used to synthesise nca ^{177}Lu labelled radiotherapeuticals.

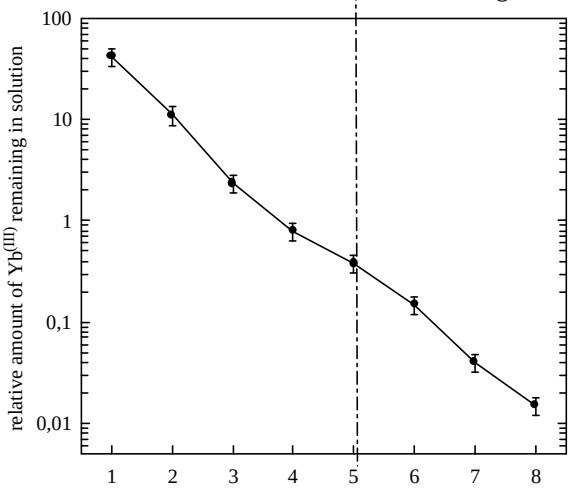

Figure 1: Successive separation of macro-amounts of $Yb^{(III)}$ in individual cementation cycles

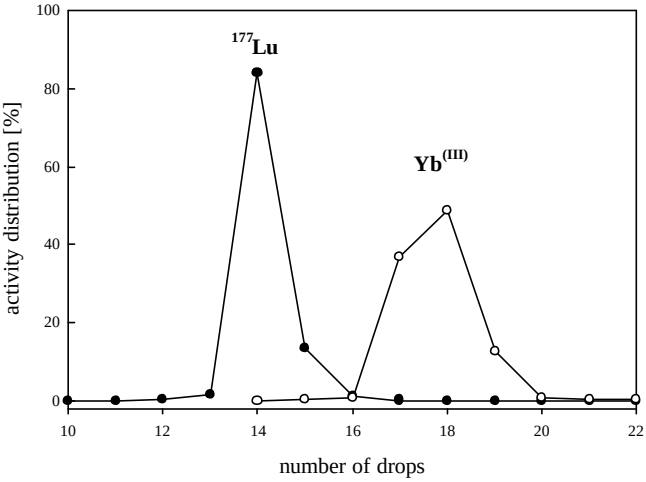

Figure 2: Ion exchange purification ^{177}Lu from $Yb^{(III)}$ using 0.07 M α-HIB, pH 4.7; column: Aminex A6, 2 mm x 80 mm

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