Isolation of accelerator produced ^{140}Nd from macro-amount of Ce and Pr by means of cation-exchanger chromatography

K.P. Zhernosekov[1], D.V. Filosofov[2], N.A. Lebedev[2], S.M. Qaim[3], F. Rösch[1]

[1]Institut für Kernchemie, Johannes Gutenberg-Universität, D-55128 Mainz, Germany; [2]Joint Institute of Nuclear Research, LNP, 141980 Dubna; [3]Institute für Nuklearchemie, Forschungszentrum Jülich GmbH, Germany

Introduction: ^{140}Nd (100 % EC, T½ = 3.37 d) generates the short-lived intermediate ^{140}Pr (49% β^+, E_{max} = 2.4 MeV, T½ = 3.39 m), which decays to stable ^{140}Ce. This system was supposed to be useful as generator or *in vivo* generator systems for PET [1]. A detailed study of ^{nat}Ce(^{3}He,xn)^{140}Nd and ^{141}Pr(p,2n)^{140}Nd nuclear reactions was published recently [2]. The overall yields of ^{140}Nd are 12 and 210 [MBq/µA·h], respectively. Whereas irradiation of praseodymium provide higher overall yields, chemical isolation of ^{140}Nd(III) seems to be more efficient if cerium is irradiated. In this work both routes were applied for the production and separation of ^{140}Nd. Radiochemical separations were performed by means of cation-exchange chromatography according to Nd(III)/Ce(III) and Nd(III)/Pr(III) separations.

Experimental: ^{140}Nd was produced irradiating natural cerium oxide with 36 MeV ^{3}He-particles and irradiating praseodymium oxide with protons of 30 MeV at the CV28 cyclotron of the Forschungszentrum Jülich. Irradiated CeO$_2$ (500 mg ≡ 2.9 mmol) was dissolved in HCl$_{conc}$ solutions by reduction of Ce(IV) to Ce(III) in the presence of I$^-$ ions. The target material was boiled in ~ 40 ml of HCl$_{conc}$ with addition of 0.5-1 g KI within 1–1.5 hours. After complete dissolution, the remaining bulk (~ 10 ml) was adjusted up to 110 ml with H$_2$O and filtrated on a standard glass filter. The solution was loaded on a chromatography column of 400×20 mm dimension (V$_{fr}$ ~ 127 ml), filled with Bio-Rad AG 50W-X8, 200-400 mesh in hydrogen form. The resin was washed with about 600 ml of 0.5 M NH$_4$Cl to transfer the cation-exchanger into the NH$_4^+$-form. Chromatographic separation was performed by isocratic elution (Fig. 1). ^{140}Nd(III) was selectively eluted with 0.30 M α-HIB solution. Ce(III) was washed down at the concentration 0.40 M. The eluate was fractionated by 30 ml. Irradiated Pr$_2$O$_3$ (200 mg ≡ 0.6 mmol) was dissolved in 5 ml of HCl$_{conc}$ by heating within 20 – 30 minutes. After addition of $^{142/141}$PrCl$_3$ and 5 mmol of NH$_4$Cl the mixture was evaporated under argon atmosphere. The dried residue was dissolved in 20 ml of H$_2$O to achieve a pH 1 - 2 and filtrated on a standard glass filter. The primary chromatography column had optimised dimension of 390×16.1 mm (V$_{fr}$ ~ 80 ml), filled with Bio-Rad AG 50W-X8, 200-400 mesh. To improve separation conditions, isotopes were loaded onto the cation-exchanger directly in NH$_4^+$ form. The resin was washed with 120 ml of 0.20 M α-HIB solution. ^{140}Nd(III) was selectively eluted with 0.29 M α-HIB solution. Pr(III) was washed down at the concentration 0.40 M (Fig. 2). The eluate was fractionated by 12 ml. In both cases final purification of ^{140}Nd(III) could be performed on a small Aminex A6 column (100×2 mm), using α-HIB eluent systems.
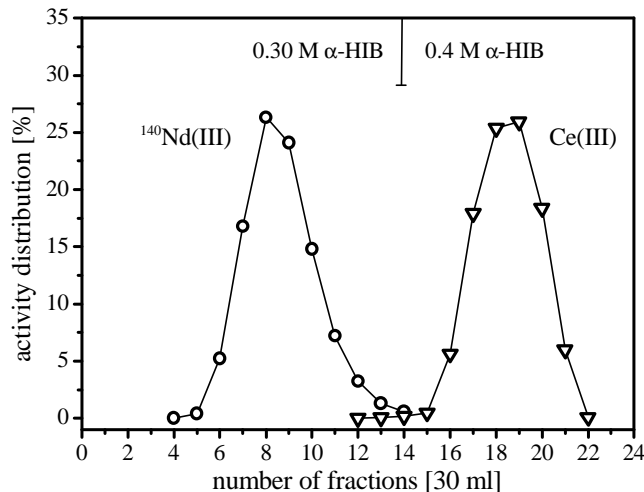

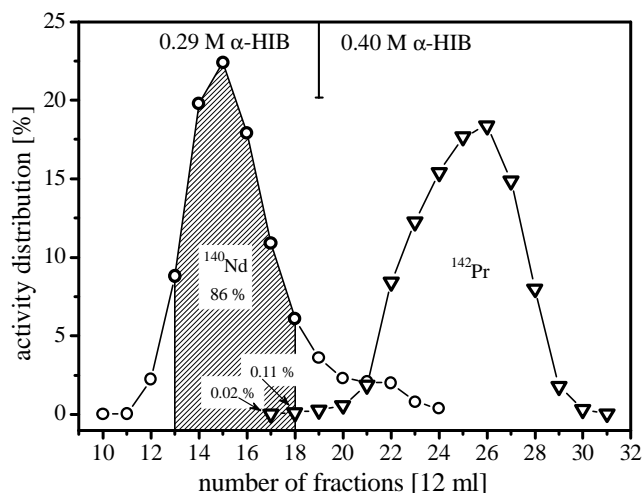Figure 1. ^{140}Nd(III)/Ce(III) separation. Profiles of an isocratic elution on the primary chromatography column.

Figure 2. ^{140}Nd(III)/Pr(III) separation. Profiles of elution on the primary chromatography column

Results: The isolation of ^{140}Nd(III) by means of cation-exchanger chromatography from the target materials was evidently more efficient if cerium oxide is irradiated (*decontamination factor* ≥ *10^8*). However, superior purification within two steps only could be performed for the ^{140}Nd(III)/Pr(III) system (*decontamination factor* ≥ *7·10^5*). In both cases the evaluated amounts of the target material remaining was below 1 nmol. With consideration of higher ^{140}Nd overall yield, the ^{141}Pr(p,2n)^{140}Nd production route seems to be absolutely superior.

References
[1] Hilgers K, Shubin YN, Qaim SM. Appl Rad. Isot. 2006
[2] Roesch F, Knapp FR. Radionuclide Generators. In: Vértes, A., Nagy, S., Klencsár, Z. Handbook of Nuclear Chemistry. Amsterdam, 2003; 4: 81 - 118.