Contribution ID: **978** Type: **Verbal**

Electrochemical oxidation of terbium(III) in aqueous media towards purification of medical Tb-161

Thursday, 19 May 2022 11:20 (20 minutes)

Radiolanthanides are very promising for use in nuclear medicine. Due to their similar chemical properties, one and the same type of coordination compounds can find various diagnostic and therapeutic applications. Terbium has four radioisotopes with different decay properties, energies and half-lives and these can be used for noninvasive theranostic purposes. Carrier-free terbium-161 is produced via neutron irradiation of highly enriched gadolinium-160 targets in the Belgian Reactor 2 (BR2) at SCK CEN. Terbium-161 emits low-energy beta particles accompanied by gamma photons and shows similar properties to lutetium-177. Furthermore, the co-emission of Auger electrons make terbium-161 more attractive towards to a combined β^-/Auger electron therapy.

Apart from the omnipresent trivalent oxidation state, several lanthanides can occur in the divalent or tetravalent oxidation states as well. A change in valence state alters the chemical properties and therefore facilitates intragroup lanthanide separations. The hydrated terbium(III) ion has a highly positive reduction potential (E^0 = + 3.1 V vs. SHE), but it can be oxidized to its tetravalent state via electrolysis and stabilized in highly concentrated carbonate solutions. In this study, Tb(III) is electrochemically oxidized to terbium(IV) in aqueous carbonate, nitrate and periodate media. Spectroscopic and elemental analyses were done to characterize Tb(IV) and to gain insight in the stability of these complexes in the aqueous electrolytes. A detailed parameter study on pH, terbium concentration, salt concentration and applied electrical potentials was performed. Nitrate and periodate media allow for oxidation at lower pH values than carbonate medium. The least positive applied potential value to oxidize Tb(III) was achieved in periodate medium at + 0.9 V vs. Ag/AgCl. However, nitrate and periodate media were poorly stabilizing Tb(IV), whereas the most efficient stabilization of Tb(IV) was observed in carbonate medium. Therefore, aqueous carbonate medium seem to be the most promising medium for developing a separation process based on the change in oxidation state of terbium. Oxidation of Tb(III) to Tb(IV) and the stability of Tb(IV) in carbonate media were confirmed and quantified by UV-Vis spectroscopy and XANES. Finally, Tb(IV) was separated from carbonate medium by solvent extraction and column chromatography.

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Session Classification: Separation & Speciation

Track Classification: Separation Methods, Speciation