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Multi-method investigation of europium(III) complexation with the decorporation agent HEDP over a wide pH range

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In case radionuclides (RN) enter the food chain and are incorporated by humans, they pose a possible health risk due to their radio- and chemotoxicity. In case of such incorporation, HEDP (1-Hydroxyethylidene-1,1-diphosphonic acid; etidronic acid) and DTPA (diethylenetriaminepentaacetic acid; pentetic acid) are common decorporation agents for uranium and transuranium RN, respectively.

Since HEDP also binds trivalent actinides, An(III), we investigated the complexation of HEDP with Eu(III) as a luminescent non-radioactive analog of An(III) at I = 0.1 M (NaCl) from pH 1 - 12 using a variety of spectroscopic and analytical methods. The pH-dependent ligand behavior was studied with infrared spectroscopy with attenuated total reflection (ATR-FT-IR) combined with density functional theory (DFT) and pKa values were determined by nuclear magnetic resonance spectroscopy (NMR). The Eu(III)-ligand system was, then, investigated by time-resolved laser-induced fluorescence spectroscopy (TRLFS), ATR-FT-IR, solution and solid-state NMR as well as mass spectrometry with inductive coupled plasma (ICP-MS) and with electron spray ionization (ESI-MS).

Depending on both the pH and the metal to ligand ratio, several Eu(III)-HEDP complex species were observed and characterized within this study. Over a wide pH range and especially at physiological values, the complexation of Eu(III) and HEDP leads to the precipitation of hardly soluble species.

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