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ORAL PRESENTATION - Complexation of Cm(III) with 2,6-bis(5-(2,2-dimethylpropyl)-1H-pyrazol-3-yl)pyridine (C5-BPP) studied by time-resolved laser fluorescence spectroscopy

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Reducing the long-term radiotoxicity and heat load of spent nuclear fuel by separating long-lived radionuclides and converting them into shorter lived or stable nuclides by fast neutron induced fission reactions is the aim of the so called partitioning and transmutation strategy (P&T).[1] Key step is the separation of the trivalent actinides Am(III) and Cm(III) from the trivalent lanthanides in the SANEX process. Due to the great chemical similarity of these metal ions, an efficient separation is very challenging. Alkylated 2,6-bis(1,2,4-triazin-3-yl)pyridines (BTPs) belong to the most promising extracting agents.[2] Although a multitude of BTP-type ligands has been synthesized, the molecular reason for the high selectivity of these soft N-donor ligands is not fully understood. To gain a better understanding, further investigations on the complexation of An(III) and Ln(III) with BTP-type ligands with systematic variations in the aromatic system are of particular interest.

In the present work, the complexation of Cm(III) with 2,6-bis(5-(2,2-dimethylpropyl)-1H-pyrazol-3-yl)pyridine (C5-BPP) in methanol is investigated by TRLFS. Time-resolved laser fluorescence spectroscopy (TRLFS) is a very sensitive spectroscopic method which makes it possible to follow the complex formation of fluorescing ions like Cm(III) in solution.[3] Spectroscopic parameters like position and shape of an emission band provide information on the number and the type of coordinating ligands. For the Cm(III) complexation with C5-BPP, three different species, the 1:1-, 1:2- and 1:3-complex, with emission bands at 603.7 nm, 607.7 nm, and 611.6 nm, respectively, are found. The species distributions for various ligand concentrations are determined and stability constants are derived ($\log K_{03} = 14.8$). As extraction from nitric acid solutions with the C5-BPP ligand is only possible in the presence of a lipophilic anion, the complexation of Cm(III) with C5-BPP and 2-bromohexanoic acid is investigated. It is found, that 2-bromohexanoate ions are coordinated to the metal ion at low C5-BPP concentrations and are successively replaced by the N-donor ligand in the inner coordination sphere.

[1] Actinide and Fission Product Partitioning and Transmutation, Status and Assessment Report, OECD Nuclear Energy Agency, 1999. [2] a) Z. Kolarik, U. Müllich, F. Gassner, Solvent Extr. Ion Exch. 1999, 17, 1155-1170. b) S. Trumm, A. Geist, P. J. Panak, Th. Fanghänel, Solv. Extr. Ion Exch. 2011, 29, 213-229. [3] R. Klenze, J. I. Kim, H. Wimmer, Radiochim. Acta 1991, 52/53, 97-103.

Author: BREMER, Antje (Institute for Nuclear Waste Disposal - Karlsruhe Institute of Technology, Germany)

Co-authors: Dr GEIST, Andreas (Institute for Nuclear Waste Disposal - Karlsruhe Institute of Technology); Prof. PANAK, Petra J. (University of Heidelberg - Institute of Physical Chemistry)

Presenter: BREMER, Antje (Institute for Nuclear Waste Disposal - Karlsruhe Institute of Technology, Germany)

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