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Complexation studies of Eu(III) with NTA at high ionic strengths

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Radionuclide speciation inside long-term radioactive waste repositories needs to be understood in order to ensure effective containment of the waste. Organic ligands originating from the degradation of organic components inside such a repository can possibly affect the mobility of radionuclides in solution. The present study focuses on nitrilotriacetic acid, NTA, as a model molecule and europium, Eu(III), as a nonradioactive analog with outstanding luminescence and magnetic properties.

The complexation of NTA with Eu(III) (in ratios of 1:2 and 1:1 Eu:NTA) as a function of pH was studied using nuclear magnetic resonance (NMR) spectroscopy in 1 M NaCl D_2O solution. The ^{1}H and ^{13}C NMR spectra of the NTA solutions with Eu(III) show clearly distinguishable signals for the free NTA and two Eu-NTA complexes, which is indicative of a 1:1 and a 1:2 Eu-NTA complex. The interaction of Eu(III) with NTA is relatively strong and favors the 1:2 Eu-NTA complex even in solution containing 1:1 Eu-NTA ratio, unless in very acidic solutions.

As a repository relevant cationic groundwater components, the influence of Ca(II), Al(III) on Eu(III) complexation is studied in detail.

A combination of NMR spectroscopy and time-resolved laser-induced fluorescence spectroscopy yields qualitative and quantitative information on the coordination environment from the ligand's and the metal ion's perspective, respectively. In subsequent studies focusing on ternary systems comprising repository relevant solid phases, radionuclides and organic ligands this will allow the identification of radionuclide speciation in solution and their sorption to solid phases.

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