Abstract

In case of an accidental release of radionuclides actinides can cause a serious health risk upon incorporation. With regard to the development of potential decontamination therapies, a detailed understanding of the mechanisms of relevant biochemical reactions is necessary. Human serum transferrin, an iron carrier protein in the blood, is only saturated to 30% with iron indicating a high capacity for the complexation of other metal ions.
The complexation of Cm(III) with human serum transferrin is investigated in a pH range from 3.5 to 11.0 using time-resolved laser fluorescence spectroscopy (TRLFS). At pH ≥ 7.4 Cm(III) is incorporated at the Fe(III) binding site of transferrin whereas at lower pH a partially bound Cm(III) transferrin species is formed. The fluorescence lifetime of the incorporated Cm(III) transferrin species correlates with two water molecules in the first coordination sphere resulting in a 4-fold coordination of Cm(III) via amino acid groups of the protein and coordination of three additional ligands (e.g. synergistic anions).
Furthermore, the complexation of Eu(III) with human serum transferrin was investigated at T < 20K using TRLFS. At high pH (pH 7.4/8.0) the spectra are characterized by a high fluorescence intensity and maximum splitting of the emission band indicating a highly unsymmetrical coordination environment of Eu(III) in the transferrin complex. The fluorescence lifetime indicates a sevenfold coordination of Eu(III) via amino acid groups of the protein and synergistic anions. This species corresponds to the incorporated Cm(III) transferrin species. The spectra at lower pH (pH 4.5/6.0) are less intense and show no visible splitting indicating the formation of an Eu(III) transferrin species with partial coordination of Eu(III) to the protein.
Further structural investigations were performed by EXAFS spectroscopy on Am(III) transferrin.[3] They show the formation of an Am(III) transferrin complex at pH 8.5 which is characterized by a very short average distance of R = 2.38 Å in the first coordination shell. This indicates strong multidentate coordination of Am(III) as expected in case of coordination of Am(III) at the transferrin binding cleft. The presence of several shells at higher distances points to a well-defined bonding environment in the Am(III) transferrin complex.