Soft X-ray spectromicroscopy of natural organics affecting actinide mobility – An overview

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Securing nuclear repository safety requires profound understanding of molecular processes determining the mobility of released radionuclides (i.e., actinides). In this context, systematic spectromicroscopy studies have been performed over the last decade [1]. Scanning Transmission X-ray Microscopy (STXM) is an appropriate tool to visualize natural organics and their interactions with metal cations or mineral phases, providing excellent spatial resolution down to the nm-level in combination with high chemical sensitivity. This chemical speciation information can be obtained from K- or L-edge NEXAFS (Near Edge X-ray Absorption Fine Structure). STXM/NEXAFS investigations are performed at three different endstations: (1) X-1A-STXM, NSLS, USA, (2) PolLux-STXM, SLS, Switzerland, (3) ALS-MES-STXM, LBNL, USA. Moreover, STXM benefits from the ability to characterize environmental samples in thin films of aqueous suspensions or in thin sections. The studies cover various aspects of natural organics and their interactions with metal ions and mineral phases: (a) basic research on model compounds (e.g., polyacrylic acid (PAA)), (b) metal ion interaction with conditioned humic acid (HA) and (c) organic-mineral associations in natural systems (e.g., claystone). Appropriate model compounds are selected, helping to assign spectral features in the more complex natural matter. These assignments are corroborated by quantum-chemical calculations [2]. A distinct metal ion complexation effect is visible in both the C1s-NEXAFS of PAA and HA metal ion complexes depending on sample pH [3]. Metal cations are enriched in a HA minority fraction as determined in Eu(III)-HA aggregates [4]. Furthermore, HA are found associated and fractionated on certain mineral phases, e.g., Al-oxides. Characteristic organic-mineral associations are also observed in claystone samples originating from a possible repository site.

[1] Plaschke, M. et al. (2011) in: Actinide nanoparticle research, (Kalmykov, S. N. and Denecke, M. A., eds.), Springer-Verlag, Berlin, Heidelberg, pp. 161-184.
[2] Bâldea, I. et al. (2007) J. Electron Spec. Relat. Phenom. 154, 109–118.
[3] Armbruster, M. K. et al. (2009) J. Electron Spec. Relat. Phenom. 169, 51–56.
[4] Naber, A. et al. (2006) J. Electron Spec. Relat. Phenom. 153, 71–74.