

Introduction to synchrotron-based radionuclide speciation investigations

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One of the key concerns in the safe disposal of high level nuclear waste is the possibility for radionuclides to leach out of a waste form, breach the repositories multi-barrier system and enter the surrounding environment. Predicting or ultimately controlling or prohibiting the transport of released radionuclides through the environment requires a detailed understanding of the physical and chemical factors and processes determinant in their transport. Speciation of radionuclides and their interaction with ground and pore water and with minerals comprising rocks of proposed host geological formations is requisite to performance assessment of nuclear repositories. Synchrotron-based techniques -including X-ray scattering and spectroscopic methods- are valuable speciation methods providing fundamental molecular scale information, thereby advancing our understanding of processes determinant in the behavior of radionuclides released into the environment, their transport properties (mobilization/immobilization), reactivity, bio-availability and, hence, potential risk. In this presentation both general experimental basics of synchrotron-based X-ray techniques and numerous application examples of radionuclide speciation are presented. The examples presented illustrate important aspects of research dealing with nuclear waste disposal safety, as well as basic and advanced aspects of X-ray absorption fine structure (XAFS) spectroscopy. In addition to examples conveying the general capability and information content of XAFS data in both the hard and soft X-ray regime, examples of the application of sophisticated spatially resolved techniques will be presented, as these take on a central role in molecular radionuclide speciation of reactions at the water/mineral interface and in heterogeneous natural systems. These examples include measurements at a grazing angle of incidence (GIXAFS) providing surface selectivity to study interface processes and micro-focus X-ray spectroscopic techniques (μ -XRF, μ -XAFS, and μ -XRD) providing lateral and depth resolution (in confocal mode) to help tackle the complex challenge of environmental radionuclide speciation.