XAS based speciation of technetium and long-lived fission products

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X-ray absorption spectroscopy (XAS) and related, primarily synchrotron-based speciation techniques have become indispensable to tackle open issues in fundamental radiochemistry and nuclear materials science alike. XAS provides element specific *in situ* physicochemical structure information by recording the linear mass absorption coefficient μ(E) of a specimen at X-ray photon energies in the vicinity of element specific electronic core levels of a selected atom type. The observed X-ray Absorption Fine Structure (XAFS – encompassing X-ray Absorption Near Edge Structure XANES and Extended X-ray Absorption Fine Structure EXAFS) is either obtained in a direct transmission measurement, or via monitoring a decay channel of the initial core-hole (e.g., X-ray fluorescence or photoelectron emission). XAS techniques are often unmatched if it comes to the precise determination of the local coordination structure and electronic (i.e., bonding) state of atomic species in complex multi-element or -phase materials. However, only very few of the worldwide more than 50 synchrotron radiation facilities offer specialized infrastructure and safety protocols allowing for experiments on radioactive samples with activities beyond exemption limits, as detailed in [1]. While certain aspects of radionuclide behavior in nuclear materials or geochemical settings may be approached by their substitution with either stable isotopes (for fission product elements) or chemical homologues (e.g., 4f- for 5f-elements) in simulated samples, this practice fails for generally instable radioelements such as Tc. Examples for XAS investigations of Tc compounds – both at the hard X-ray K- and tender X-ray L_3-edges – will be discussed, and augmented by an overview of recent XAS spectroscopy results obtained for fission products in spent nuclear fuel.

References

[1] Rothe, J. et al. (2019) *Geosciences* 9, 91.