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X-ray absorption fine structure (XAFS)-based radionuclide research at the KIT Light Source

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Abstract. In the past two decades, X-ray absorption spectroscopy (XAS) and related synchrotron-based radionuclide speciation techniques have become indispensable, supporting open issues in fundamental radiochemistry and nuclear waste disposal alike. Physicochemical structure information – encoded in the element-specific X-ray absorption fine structure (XAFS) – is obtained by the precise determination of the linear mass absorption coefficient $\mu(E)$ of a specimen in an X-ray energy window ΔE encompassing an electronic bonding level of a selected atom type. The characteristic modulations of $\mu(E)$ in the vicinity of an "absorption edge" (i.e., the steeply rising absorption probability of photons tuned to the binding energy of inner shell "core" electrons) are attributed to electronic transitions into unoccupied states due to the photoelectric effect. While absorption features close to the edge (arising from excitations into molecular orbitals or empty band structure levels close to the Fermi level) are generally denoted as an X-ray absorption near-edge structure (XANES), the scattering states at higher excitation energies are causing the well-known extended X-ray absorption fine structure (EXAFS). EXAFS oscillations of the post-edge $\mu(E)$ are caused by the periodicity of the constructive and destructive interference of outbound and backscattered inbound photoelectron waves (superimposing at the absorbing atom site) when continuously increasing the excitation energy. Quantitative analysis of EXAFS data based on electron scattering theory is a well-established short-range structural probe, providing precise values of interatomic distances (between the absorber atom and its neighbors) and additional information on the type, number, and ordering of neighbor atoms within a local cluster. Recent significant advancements in the quantum theoretical description of core-excited final states have been successful in overcoming persisting ambiguities in XANES data interpretation. Meanwhile, although there are more than 50 synchrotron radiation centers operational in the world, facilities with specialized beamline infrastructure and safety protocols to allow experiments on radioactive samples with activities beyond the exemption limits are still scarce. At the KIT Light Source (at the Karlsruhe Institute of Technology, Karlsruhe, Germany), the Institute for Nuclear Waste Disposal (Institut für Nukleare Entsorgung, INE) operates two beamline end-stations dedicated to the investigation of radionuclide materials by XAFS spectroscopy and related techniques, i.e., the INE-Beamline (operational since 2005) and the ACT station of the CAT-ACT (CATalysis and ACTinide) wiggler beamline (operational since 2016). Samples with total activities amounting to 1.0×10^6 times the isotope specific exemption limits and 200 mg of fissile isotopes ^{235}U and ^{239}Pu are feasible at both beamlines. Highly versatile detection systems and sample holders – generally avoiding extensive shielding or sample handling in a glove box – have been conceived since the initial commissioning of the beamlines and have enabled unique XAFS measurements such as the first ever investigation of aqueous technetium species at the Tc L_3 edge. Investigations of highly radioactive specimens (e.g., spent nuclear fuel or nuclear waste glass fragments) are feasible, regardless of the actual contact dose rate if the radiation field is limited to a few microsieverts per hour at the edge of the experimental table.

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