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High resolution X-ray spectroscopy of actinides at the KIT light source"for the session on "Beamline Innovations

Tuesday 27 August 2024 11:30 (20 minutes)

High energy resolution X-ray absorption and emission spectroscopy techniques have become indispensable methods in actinide research.1,2 In the last 15 years, we have substantially advanced the experimental capabilities for high resolution X-ray spectroscopy of actinides at the KIT Light Source. This continuous effort has recently been expanded to include both additional beamlines as well as soft X-ray spectroscopy capabilities. One important motivation is conducting studies to obtain a deep understanding of the retention mechanisms of long-lived actinides and fission products in geochemical processes relevant for the long-term safety of a deep geological nuclear waste repository. Here, the X-ray spectroscopy techniques allow for in-depth insights into the actinide-ligand binding properties, which are still very controversially discussed. Our recent efforts also include the development of spectroscopic tools to probe metal-chelating ligand bond covalency of radiopharmaceuticals for the targeted alpha-treatment of tumors. We will discuss recent developments at the ACT experimental station of the CAT-ACT beamline3,4 and at the SUL-X and X-SPEC5 beamlines at the KIT Light Source.

One experimental technique, particularly powerful in differentiating oxidation states of actinides (An), is the An M4,5-edge high-energy resolution X-ray absorption near-edge structure (HR-XANES) method. This presentation highlights the latest technological developments at the ACT station, enabling HR-XANES for samples with low actinide loading (down to 1 ppm), in combination with a cryogenic sample environment that reduces beam-induced sample alterations.4,6 In addition, an in situ cell for studying interaction mechanisms of actinides with clay minerals at fixed redox conditions, in combination with tender (3-4 keV) X-ray spectroscopy at the actinide An M4,5 edges, will be presented. These experimental capabilities pave the way for examining coupled redox/solid-liquid interface reactions.6,7

Examples of applications of An M4,5 edge core-to-core and valence-to-core resonant inelastic X-ray scattering (CC-RIXS and VB-RIXS) for probing the electronic structure and binding properties of the actinide elements will be illustrated.1 An important new instrumental development is the possibility to record An M4 edge CC-RIXS at near-backscattering geometry, using the newly commissioned NEXT X-ray emission spectrometer at ACT or the recently installed single crystal analyzer X-ray emission spectrometer (Rowland circle geometry) at the SUL-X beamline.

The design and first experiments using a versatile chamber (the "Actinide Chamber") with solid, liquid, and gas state cells for soft X-ray spectroscopy of actinides at the X-SPEC beamline at the KIT Light Source will also be illustrated. Furthermore, the ROXS ("Radionuclide materials Observed with soft X-ray Spectroscopy") experimental station at the X-SPEC beamline, currently in the design phase, will be presented, focusing on the application of magnetic microcalorimeters for high energy resolution and high efficiency X-ray spectroscopy at the KIT Light Source.

References:

[1] Tonya Vitova et al. Nature Commun. 8, 16053 (2017); T. Vitova et al. Chem. Sci. 13 (37), 11038 (2022); [2] Ivan Pidchenko et al. , Environ. Sci. Technol. 51 (4), 2217 (2017); [3] A. Zimina et al. Rev Sci. Instrum. 88 (11), 113113 (2017); [4] Bianca Schacherl et al. J. of Synchrotron Rad. 29 (1),80 (2022); [5] L. Weinhardt et al. , J. of Synchrotron Rad. 28, 609 (2021); [6] Bianca Schacherl et al. Anal. Chim. Acta 1202, 339636 (2022); [7] B. Schacherl et al. Environ. Sci. Technol. 57 (30), 11185 (2023).

I plan to submit also conference proceedings

No

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