

HERFD in actinide research

S. M. Butorin

Department of Physics and Astronomy, Uppsala University, Sweden,
sergei.butorin@physics.uu.se

Among various methods of gaining the knowledge about the properties of actinide materials, X-ray spectroscopy plays the most important role. Better understanding of the electronic structure and fundamental properties of the actinide systems contributes to the knowledge relevant for the technological performance of actinide materials under study. In case of actinides, only techniques, such as XAS and EXAFS, can provide accurate information about the chemical state, homogeneity of compounds, (non)stoichiometry, oxygen/metal (O/M) ratio, local symmetry and environment and charge distribution.

I have been involved in recent development of the advanced technique, such as high energy resolution fluorescence detection X-ray absorption spectroscopy (HERFD-XAS) applied both at the actinide L_3 ($2p_{3/2}$ -6d transition) and $M_{4,5}$ (3d-5f transition) edges (the hard and tender X-ray range, respectively). In particular, the application of this technique at the $M_{4,5}$ edges has led to a tremendous improvement in the experimental resolution (up to 8 times higher) due to reduced core-hole lifetime broadening and revealed additional structures in the XAS spectra, not measurable before.

In my talk, I will present a few examples of studies of actinide materials using this advanced technique at the L and M edges, in particular at ESRF, where the best quality data to date were measured. I will discuss how the development of this technique and its application to actinides help to resolve long standing and debated questions. The interpretation of the data is supported by calculations in framework of the Anderson impurity model using the modified TT-MULTIPLETS package and by DFT+ U calculations using the OCEAN program.

In the end, I will discuss an extension of a similar technique, based on a selective detection of X-ray emission, to the soft X-ray range and shallow $O_{4,5}$ (5d-5f transition) edges of actinides which increases the sensitivity of XAS to the chemical state of dilute species.