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Advanced X-ray spectroscopy at M and N edges of actinides

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Data obtained in the tender and soft x-ray ranges for the Th, U, Np and Pu systems are presented and discussed. The use of the so-called Hämäläinen scan at the actinide 3d edges by monitoring the 4f-to-3d x-ray emission transition drastically improves the energy resolution compared to conventional XANES and reveals multiple structures never seen before in contrast to a single line in conventional XANES spectra. This allows for an access in some cases to unique information which is difficult to obtain with other techniques. The data are compared with the results of calculations in the framework of the Anderson impurity model accounting for the Coulomb interaction (U) within the 5f shell and between 5f electrons and a core-hole. The combination of such experiment and theory enables the study of detailed properties of the electronic structure such as ligand field effects and spin interactions involving the 5f electrons as well as the degree of hybridization of the 5f states.

The measurements at the actinide 4f edges reveal all the fine structure in the distribution of unoccupied 6d states in actinide systems which cannot be observed in commonly used XANES spectra at the actinide L edges due to their large 2p(3d) core-hole lifetime broadening. This enhances the sensitivity of the spectroscopy to fine changes in the electronic state distribution due to e.g. doping/non-stoichiometric effects. The data are compared with the results of LDA+ U calculations which also provide the estimation of the U value by probing the delocalized electronic component in a number of actinide systems.

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