

High energy resolution X-ray spectroscopy and diffraction studies of plutonium oxide nanoparticles

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The release of radioactive plutonium (Pu) into the environment is of general concern due to the high radiotoxicity and long half-life of its main isotopes. Previous research has shown that plutonium migrates in the subsurface environment on the kilometer scale at some previously contaminated sites [1-4]. Additionally, previous research demonstrated the spontaneous formation of Pu oxide nanoparticles under certain environmental conditions [5]. However, fundamental properties of such Pu oxide nanoparticles, including their local, crystal and electronic structure, remain largely unexplored, hence it is difficult to understand their formation or to predict their transport in the environment.

Plutonium may exist in four oxidation states, III, IV, V, VI, in aqueous solution under environmental conditions, which can change relatively easily. While Pu(IV) is the dominant oxidation state in such PuO₂-like nanoparticles, their exact composition in terms of oxidation states and local structure remains an open question. Therefore, it is necessary to advance the fundamental understanding of the Pu oxide nanoparticles and to review the processes, through which the formation of Pu oxide nanoparticles takes place.

This contribution will give an overview on the results of Pu oxide nanoparticle research conducted at the Rossendorf Beamline at The European Synchrotron (ESRF) [6]. Pu oxide nanoparticles were prepared by rapid chemical precipitation using precursors in the different oxidation states (Pu(III), Pu(IV), Pu(V) and Pu(VI)). These precursors were obtained by chemical reduction or oxidation of Pu stock solution.

The recently upgraded ROBL beamline at the ESRF, dedicated to actinide science, provides now a unique opportunity to characterize actinide materials by several experimental techniques simultaneously. We will show how the detailed information about local and electronic structure and Pu oxidation state in different nanoparticles can be obtained using the variety of methods: Extended X-ray absorption fine structure (EXAFS [7]), X-ray absorption near edge structure (XANES), high-energy resolution fluorescence detection (HERFD) X-ray absorption spectroscopy [8-10], resonant inelastic X-ray scattering (RIXS [11]), and X-ray diffraction techniques.

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