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The actinide dioxides nanophase stability in the presence of environmental-related anions

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Recently actinide(IV) oxides (AnO_2) nanoparticles (NPs) have increased their importance for environmental safety issues and the development of new technological schemes. The progress in high-precision methods of solid phase analysis has increased our understanding of the structures of actinides-based particles at the nanoscale. Actinide dioxides NPs of different crystallinity can be formed in solutions under various conditions. This study aims to determine the phase stability of actinide dioxides (PuO_2, NpO_2, ThO_2) in the form of nanoparticles during aging in solutions. Experiments with a non-radioactive analog of actinide dioxide - CeO_2 were also carried out. Solubility and phase transportations of AnO_2 and CeO_2 in pure solutions (without complexing agents), phosphate, and carbonate-containing media are studied to evaluate the possible aging pathways.

To study the aging process, AnO_2 or CeO_2 nanoparticles were kept in aqueous media with different pH values in the presence of $NaClO_4$, Na_2HPO_4/NaH_2PO_4, or $NaHCO_3$ for up to 4 years. Additionally, the dioxide's aging process was studied under mild hydrothermal treatment (HT) conditions (< 150 °C). The structure features and phase composition of the samples were investigated by synchrotron X-ray diffraction (XRD) and absorption spectroscopy (XAS), electronic microscopy (SEM and HRTEM), and Raman spectroscopy. It was shown that the X-ray amorphous ThO_2 trends to crystallize to nanocrystalline ThO_2particles through long-term aging and HT treatment in $NaClO_4$ solution. Simultaneously, no significant changes in PuO_2 and CeO_2 structure were observed under the same aging conditions. Under long-term storage and HT treatment conditions in a phosphate buffer medium, PuO_2, CeO_2, and ThO_2 nanoparticles are crystallized into the phosphates phase. Their second-generation phase structure depends on the pH value and the composition of the initial phosphate buffer. The effect of carbonate anions on Np-containing particle formation in solutions was also revealed.

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