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X-ray absorption and emission spectroscopic study at O K-edges of light actinide oxides

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X-ray absorption and emission spectroscopic study at O K-edges of light actinide oxides

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Here we present soft X-ray emission and X-ray absorption spectroscopic data recorded at O K edges of UO₂, NpO₂ and PuO₂. Interpretation of the experimental data is supported by first-principle calculations in framework of LDA+U and GGA+U formalisms (see Fig. 1). A discussion regarding the origin of different structures in the X-ray emission and X-ray absorption spectra is included. The effect of varying intra-atomic Coulomb interaction U for the f-electrons is investigated. Our data indicate that O-K X-ray absorption and X-ray emission spectroscopies can successfully be used to study correlation effects in compounds of light actinides.

For single-crystal Pu(²³⁹isotope)O₂, the measured O 1s X-ray absorption spectra show significant difference in intensity for the first two peaks between different areas/spots on the single crystal surface. From theoretical results, the first peak can be attributed to O 2p-Pu 5f hybridization, while the second peak is due to hybridization with Pu d-states. The observed difference in the O 1s X-ray absorption spectra was studied by simulating a number of defect structures of PuO₂ as well as existence of Pu(V) sites. The results indicate the presence of higher oxidation states than Pu(IV) in some surface areas of the single crystal. The results also suggest that plutonium oxide with a Pu fraction in a higher oxidation state than Pu(IV) consists of inequivalent sites with Pu(IV)O₂ and Pu(V)O₂ rather than is being a system where the Pu oxidation state is constantly fluctuating between Pu(IV) and Pu(V).

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Fig.1 Comparison of experimentally recorded (in red) and LDA+U calculated (in blue) O K X-ray emission and O 1s X-ray absorption spectra of UO₂, NpO₂ and PuO₂.

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