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Oxidation pathways of UO_2 thin film under combined oxygen-humidity-radiation conditions probed by XAFS

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Understanding the uranium dioxide (UO_2) oxidation and phase change is of importance to minimize the environmental impact of fuel rod damage and leaking of uranium. Here, we studied the effect of water vapor, oxygen gas, and radiation on the oxidation of UO_2 (001) thin film. We in-situ observed the oxidation of UO_2 thin film in radioactive and anoxic/oxic environments with different humidity by utilizing grazing incidence X-ray absorption fine structure (XAFS). Fitting of XAFS spectra suggested that the oxidation led to the formation of multiple alteration phases. A phase deconvolution method was adopted to extract phase type and ratio from the spectra based on comparing coordination numbers between this study and previous publications. The phase deconvolution result showed that UO_2 thin film rapidly oxidized in both anoxic and oxic environment when humidity and X-ray radiation were presented. The UO_2 thin film in both anoxic and oxic environment shared similar phase transition pathways (from UO_2 to U_4O_9 and then U_3O_7). The thin film in anoxic environment was eventually dominated by U_3O_7, while thin film in oxic environment was dominated by alpha-U_3O_8. The oxidation in anoxic environment was plausibly driven by radiolysis of water molecules (OH^- and H_2O_2). In oxic environment, oxygen gas may not only directly oxidize UO_2, while also increasing the amount of H_2O_2 due to presence of more precure, $HO_2\bullet$, by reaction between $H\bullet$ and O_2. This study showed that radiation play an important role in both anoxic and oxic oxidation of UO_2.