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# Gd-doped UO2 surface oxidation by H2O(g) in the presence of H2(g) at different temperatures

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To improve the efficiency of nuclear reactors by extending the fuel cycle, burnable neutron absorbers, such as Gd2O3, have been extensively added in the PWR and BWR fuels to control the reactivity due to their large neutron absorption cross-section. Doped fuels must be safely stored after their life cycle in a deep geological repository, therefore their behaviour in case of canister failure must be examined.

To assess the safety of a deep geological repository, redox conditions of the spent nuclear fuel are of a great main concern because of the more soluble U(VI) compared to U(IV), which would result of significant release of radionuclides to the environment. Such conditions depend on the formation of both oxidizing and reducing species. Water vapour has a significant impact on the oxidation of uranium dioxide and might be present in the near-field due to residual water coming from the cooling pools or groundwater evaporation. Conversely, reducing species such as molecular hydrogen could be present because of the anoxic corrosion of the metallic canister. However, there is no information regarding the interaction among Gd-doped UO2, water vapour and hydrogen. For this reason, in this work, the effect of hydrogen stream saturated with water vapour on the surface oxidation of Gd-doped UO2 at different temperatures was studied.

In situ experiments were conducted in a high pressure chamber (HPC) connected to an X-ray Photoelectron Spectroscopy (XPS) analysis chamber. Hydrogen stream was saturated with water vapour at room temperature and introduced into the HPC, where the sample was located. Then, the temperature of the chamber was increased to the desired value and maintained for a specific time. Prior to perform any experiment, a reducing treatment with only H2 at high temperature was conducted to completely reduce the sample surface. The XPS spectra obtained after each experiment were used to determine the uranium oxidation state in the surface by the de-convolution of the U4f7/2 band into U(IV), U(V) and U(VI) contributions. Preliminary results show that the surface of Gd-doped UO2 sample was less oxidized than previous studies conducted under similar conditions on non-doped samples, where UO2 was found to be oxidized in the hydrogen/water vapor experiments. The addition of gadolinium oxide to uranium oxide could strongly reduce the water oxidizing effect on the UO2 matrix in the presence of H2(g).

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