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Influence of 316 stainless steel and Zircaloy-4 on UO_2 redox behavior

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The redox condition at the near-field of spent nuclear fuel (SNF) strongly governs its ability to undergo dissolution and is an important factor that needs to be considered for the risk assessment of nuclear waste disposal in a deep geological repository (DGR). Previously published work has shown that direct contact interfaces with canister and cladding can accelerate corrosion [1]. It is therefore crucial to evaluate a more complex scenario of technical barrier failure where groundwater interacts with the waste canister insert, cladding material, and the UO_2 matrix in SNF. The influences of waste canister material and cladding towards the redox behavior of UO_2 mimicking SNF were investigated through hydrothermal corrosion experiments in direct contact with 316 stainless steel (316 SS) and Zircaloy-4 coupons, mimicking the waste canister and cladding materials. Experiments for 1-6 weeks were conducted in simulated Mont Terri water at an elevated temperature of 250 °C with direct interfaces between all three materials.

Secondary phases formed were analyzed with SEM-EDS and Raman spectroscopy. Hydrothermal experiments with Zircaloy-4 in contact with UO_2 reveal the formation of small (~1 μm), uranium precipitates over the Zircaloy-4 body. Experiments with 316 SS in contact with UO_2 instead show large quantities of magnetite (Fe_3O_4) and chromite ($FeCr_2O_4$) secondary phases that are concentrated at the contact area between the two materials. In the final three-component experiment where UO_2 is in direct contact with Zircaloy-4 and 316 SS, there is a delay of at least 1 week for secondary uranium phases to be detected on the Zircaloy-4 coupon while magnetite and chromite continue to form from 316SS oxidation. This might suggest that, while corrosion resistant, the 316 SS is able to be oxidized enough to form magnetite and chromite such that the released iron is able to dominate the prevailing redox conditions during barrier failure. Further analysis with Raman spectroscopy is underway to identify the uranium secondary phases that have formed across the probed experimental matrix.

[1] Nguyen, et al. (2026), *J. Nucl. Mater.*, 156516.