Goldschmidt 2023 Abstract
https://doi.org/10.7185/gold2023.15415

Hydrothermal Alteration of Zircaloy and Interactions with Engineered Barrier Materials

ALYSSA J MCKANNA[1], AMBER ZANDANEL[2], MARLENA J ROCK[2], FLORIE CAPORUSCIO[2] AND KIRSTEN SAUER[3]

[1]Los Alamos National Lab
[2]Los Alamos National Laboratory
[3]NA

Presenting Author: ajmckanna@lanl.gov

In the case of canister breach within a nuclear repository setting, it is important to understand the interactions between fuel canister components, groundwater, and other engineered barrier materials to assess the long-term safety and function of the repository. Here we assess the reactivity of two Zircaloys (ZRC-2 and ZRC-4), which are potential nuclear fuel cladding materials, in contact with fluids of variable chemistry and Wyoming bentonite at elevated temperature and pressure conditions. The six hydrothermal experiments presented were pressurized to 150 bar and heated isothermally to 250 °C for two weeks. Reaction fluids were sampled 1-2 times per week and analyzed to investigate the aqueous geochemical evolution in relationship to mineralogical alterations. Solid run products were characterized using scanning electron microscopy and X-ray diffraction.

A blue or blue-purple iridescent sheen developed on all reacted Zircaloy indicating oxidation of the metal alloy which is consistent with the drop in pH observed in 3 of the 4 experiments in which Zircaloy was reacted with either double deionized water (DDI) or synthetic ground water (GW). A heterogeneous, discontinuous layer of alteration phases is observed on the surface of Zircaloy reacted with DDI or GW. Observed phases are similar between the two Zircaloy types and dominantly included ZrO_2 and Al_2O_3 or AlO(OH). Trace phases identified include Cr-oxide as well as calcium sulfate and chloride salts that likely precipitated directly from the GW solution. The impact of solution-Zircaloy interactions is further evidenced by increased concentrations of Si, Fe, Mg, and Al over the course of the experiments which are impurities that were likely leached from the Zircaloy. Notably, the experiment with ZRC-4 and synthetic GW produced a sharp drop in pH, which, in combination with the brine solution, stripped gold from the walls of the gold reaction cell and precipitated it onto the Zircaloy. Characterization of fluid and solid reaction products for the experiments that reacted Zircaloy with GW and bentonite are currently underway.