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Corrosion Study of SIMFUEL in Aerated Carbonate Solution Containing Calcium and Silicate

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Abstract

As an analog for behavior of spent nuclear fuel, the corrosion behavior of simulated nuclear fuel (SIMFUEL) in a high-level radioactive waste repository disposal setting was investigated using electrochemical and solution chemistry analyses. The SIMFUEL was exposed to aerated solutions of NaCl+$NaHCO_3$ with and without calcium and silicate. Two SIMFUEL compositions were studied, representing spent nuclear fuel (SNF) with 3 or 6 (weight) percent burnup in terms of fission products equivalents of surrogate elements. For all tested cases, the corrosion potential of the SIMFUEL increased with time and reached steady state values in the range of 0.06 to 0.12 V_{SCE} within 72 hours. The polarization resistance increased with increased immersion time, indicating possible blocking effects due to accumulation of corrosion products on the SIMFUEL electrode. The potential-pH diagram suggests formation of schoepite, and this phase may cause the increase in the polarization resistance with time. The addition of calcium and silicate produced no measureable change in values of the polarization resistance measured at the corrosion potential. The impedance was similar for solutions with or without calcium and silicate, indicating a relatively minor effect of calcium and silicate on the oxidation and dissolution rate of SIMFUEL. The dissolution rate, estimated by applying both the Stern-Geary Equation and Faraday's Law, ranged from 1 to 3 mg/m^2-day, which is similar to the range of dissolution rates for SIMFUEL and SNF reported in the literature for comparable conditions. SIMFUEL with simulated high burnup (6 percent) resulted in a minimally higher dissolution rate than the samples with simulated lower burnup (3 percent). Analysis of the solution chemistry shows that uranium is the dominant element dissolved in the post-test solutions, and the dissolution rates calculated from uranium concentration are consistent with the dissolution rates obtained from impedance measurements. Simulated-fission product elements (i.e., barium, molybdenum, strontium, and zirconium) dissolved from the SIMFUEL electrode at a relatively high rate. Sorption test results showed that uranium concentrations in the post-test solutions with a stainless steel disk decreased about 20 percent after 21 days of immersion of the stainless steel compared to the concentrations without immersed stainless steel, suggesting sorption of uranium onto the stainless steel oxide. Electrochemical impedance was found to be an effective technique for measuring uranium dissolution rate in real time. SIMFUEL characterization results indicate that SIMFUEL is a valid analog material to evaluate the SNF matrix dissolution rate after long-term containment, when gamma and beta radiation have decreased significantly in a repository environment.

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