Goldschmidt 2022 Abstract
https://doi.org/10.46427/gold2022.12155

A Review of High Temperature Engineered Barrier Systems Experiments at Los Alamos National Laboratory

FLORIE CAPORUSCIO, KIRSTEN SAUER, MARLENA ROCK AND AMBER ZANDANEL

Los Alamos National Laboratory

Presenting Author: floriec@lanl.gov

The US Department of Energy (DOE) manages nuclear waste created by civilian nuclear power. Since 2009 the DOE Used Fuel Disposition campaign has had the following mission statement: "long-term, science-based R&D of technologies with the potential to produce beneficial changes to the manner in which the nuclear fuel cycle and nuclear waste is managed.â€ Generic studies were initiated to cover all rock types (crystalline/argillite/salt) and make use of Engineered Barrier Systems (EBS), consisting of a layer of bentonite clay (+/-) cement to retard radionuclide transport. In 2018, the program became the Spent Fuel and Waste Disposition (SWFD) campaign, which emphasized dual purpose canisters (DPC**)** and high temperature studies related to these larger waste packages.

Since 2012 our research group at Los Alamos has conducted a systematic long term research program using rocking autoclaves to investigate evolution of the mineralogy and geochemistry of EBS bentonite clay at predicted repository temperatures and pressures (300-250-200 °C, 150 bar). This included interaction with repository wall rocks, simulated groundwater, canister outer skins and cement liners. Wyoming bentonite has been used in all experiments as the EBS buffer material. We chose Grimsel granodiorite for a crystalline wall rock, and Opalinus Clay as the argillite wall rock, both for the availability of material and long term research by the Swiss repository agency (NAGRA). Canister materials investigated included stainless and carbon steels, along with copper overcoats. Recently, cements such as ordinary Portland cement and low pH cured cements have been added to the reactant mix.

A summary of the experimental results will be discussed, including characterization of solid reactant products, aqueous geochemistry, and interpretations of mineral stability and fluid speciation. Observations of note include 1) lack of illite generation in the majority of experiments due to bulk system chemistry, 2) transformation of precursor clinoptilolite to analcime at the highest temperatures, 3) creation of abundant analcime and CASH minerals in the presence of cement reactants, and 4) growth of Fe smectite / chlorite at the steel bentonite interface. The results will be used to inform modeling efforts on the long-term function of deep geological repositories for nuclear waste.