Development of Novel Corrosion-Responsive Buffer Materials for Long-Term Immobilization of High-Level Nuclear Waste

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ABSTRACT:

The current design of repository infrastructure to safely contain, isolate, and dispose of high-level nuclear waste, such as heat-generating spent nuclear fuel (SNF), is limited in part by the materials available to construct this specialized infrastructure. Buffer materials, such as bentonite, surround the SNF metal canister to reduce interactions with the surrounding geological repository and limit the transport of any accidental radionuclide release. This crucial role is therefore an important consideration in the design of repository infrastructure. Bentonite clays have been considered good buffer materials; however, these limit infrastructure design due to their failure at temperatures above 100 °C. To mitigate this risk, costly practices have been adopted, such as surface storage of SNF packages, which requires 200-540 years to cool nuclear waste to acceptable levels. Thus, in addition to other bentonite limitations, there is a critical need to develop novel buffer materials that can enable robust repository designs.

With the goal to improve the safe containment, isolation, and disposal of SNF waste, this project aims to develop a novel cementitious buffer material (CBM) that can prevent corrosion of SNF packages and also immobilize fugitive radionuclides over long timescales (>10^6 years) in generic disposal concepts in salt, crystalline rock, or clay/shale repositories. To achieve this, the primary aim of this project is to identify and develop novel magnesium aluminophosphate (Mg-Al-P) CBMs, complete with assessments of their repository material stability as well as their transport and immobilization capacity of radionuclides. Additionally, given the accelerated corrosion of HLNW canisters, the secondary aim of the project is to employ advanced monitoring systems to understand the corrosive failure between the canister and CBM. The developed CBM and degradation science knowledge will contribute to the long-term performance understanding of the waste container-package through the creation of reliable digital twin models. Ultimately, this project will make significant strides to advance the robust design of the U.S. waste disposal infrastructure with greater capabilities to isolate and contain nuclear waste.

The project aims will be accomplished through four main research objectives (OBs), namely: (***OB.1***) characterization of phase assemblage time-based (0-12 months) and thermal evolution (100-1000 °C) in Mg-Al-P CBMs via x-ray diffraction, thermogravimetric analysis, and pore solution chemical analysis; (***OB.2***) assessment of the physio-chemical reactive transport of radionuclides (i.e., ^{137}Cs, ^{90}Sr, ^{243}Am, ^{237}Np, ^{99}Tc, ^{129}I) and of high-risk repository geochemical conditions (i.e., brine and groundwater flow) within Mg-Al-P pore networks; (***OB.3***) in-situ monitoring of the corrosive degradation at the canister steel-CBM interface through bespoke ultrasound (UT) and electrochemical impedance spectroscopy (EIS) non-destructive measurements; and, (***OB.4***) near-field reactive transport modeling (i.e., digital twins) for long timescale performance predictions of disposed HLNW packages in mined geological repositories with novel Mg-Al-P CBMs. Upon successful completion, this project will result in a better understanding of corrosive failure in HLNW canisters and CBM radionuclide-immobilization where the interaction between steel package, new CBM, and repository host-rock is considered.