Computational and Experimental Investigation of Thermal-Mechanical-Chemical Mechanisms of High-burnup Spent Nuclear Fuel (SNF) Processes at Elevated Temperatures and Degradation Behavior in Geologic Repositories

PI: Eunja Kim, UNLV

Collaborators: Frederic Poineau, UNLV
Philippe F. Weck, SNL
David C. Sassani, SNL
Kenneth R. Czerwinski, UNLV

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ABSTRACT:

The overarching goal of the combined computational and experimental R&D activities proposed in this project is to enhance understanding of the mechanisms and thermal-mechanical-chemical (TMC) parameters controlling the instant release fraction (IRF) and matrix dissolution of high-burnup (HB; burnup > 45 GWd/MTU) spent nuclear fuels (SNFs) and the subsequent formation, stability, and phase transformations of HB SNF alteration products under long-term storage and geological disposal conditions (*e.g.,* high-temperature storage, α-radiolysis). Uranium dioxide may undergo oxidative corrosion/alteration, and the IRF may be increased for HB SNF, both of which may affect environmental systems associated with SNF long-term storage and disposal. The oxidative matrix dissolution may form various complex uranyl-based phases, including a rich variety of oxides, silicates, carbonates and other secondary minerals in varied geological environments (e.g., studtite, metastudtite, amorphous uranyl peroxide, uranium trioxide, triuranium octoxide, schoepite, dehydrated schoepite, metaschoepite, becquerelite, soddyite, rutherfordine,...). These uranyl phases generally have higher mobility UO_2^{+2} species than less soluble U^{4+} phases. However, limited information on the thermodynamic properties and formation kinetics of these uranyl-bearing phases is available to predict explicitly paragenesis under the conditions relevant to long-term storage or disposal.

The proposed project draws on complementary expertise and research backgrounds from University and National Labs partners:

(i) to apply a combined *ab initio* modeling (UNLV and SNL) and experimental (UNLV) strategy investigating the high-temperature TMC mechanisms of alteration of HB SNF under α-radiolysis conditions, including He generation and pressure effects;

(ii) to investigate the mechanistics of phase transformations of UNF degradation products under various conditions expected in storage and disposal, including oxidative systems;

(iii) to determine high-accuracy TMC parameters such as heat capacity, bulk modulus, and thermal expansion that are easily influenced by microstructures and porosity of complex uranyl-based phases formed in storage or geological disposal environments (e.g. $UO_3(H_2O)_2$, $Ca[(UO_2)_6O_4(OH)_8]8H_2O$, $(UO_2)_2(SiO_4)_32H_2O$,…).

The results of this project will be used to enhance the mechanistic detail of process models to reduce uncertainty in, and improve the technical bases of, safety cases and performance assessment (PA) analyses used by the U.S. DOE for the long-term storage and geological disposal of HB UNFs. This proposal addresses the Grand Challenge for the U.S. DOE NE, Used Fuel Disposition Campaign (UFDC) to provide a sound technical basis for the safety and security of long-term storage and disposal of used nuclear fuel from the nuclear energy enterprise.