
Simulated Used Nuclear Fuel Dissolution as a Function of Fuel Chemistry and Near Field Conditions

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Program: Used Nuclear Fuel
Disposition (FC-4)

Collaborators:

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ABSTRACT:

This proposal aims to develop a fundamental and transformative understanding of the various effects of simulated used nuclear fuel (UNF) microstructure on its dissolution in geologic repository conditions, through an integrated research program delivered by a partnership of US and UK scientists and engineers. This understanding will underpin the maturation of models for UNF evolution and interaction under different potential repository conditions, enabling reliable prediction of degradation and adjustment of repository conditions to achieve desired long-term performance, and providing increased confidence in predicting behavior for up to one million years.

The US currently (April 2016) has 80,150 metric tons of UNF in wet and dry storage, with a prediction of a total of approximately 140,000 metric tons by 2050 when currently operating reactors reach their designated life. While the US has halted research on the Yucca Mountain geologic repository for UNF and high level waste, research continues in other countries and through the DOE-NE to identify and resolve knowledge gaps relating to the long-term degradation of UNF after emplacement. Fundamental mechanistic understanding is of paramount importance for US and international waste management programs which seek to satisfy citizens and regulators regarding the reliability of long-term degradation predictions for UNF originating from a variety of fuel designs, burnups, reactor operations, and storage conditions. The goal of this work is to explore the effects on UO₂ dissolution of 1) fuel microstructure and single- and multi-species doping, 2) oxidizing radiolysis products presence, 3) cladding presence, and 4) canister overpack residue, as a function of relevant parameters including temperature, pH, and oxidizing and reducing environment. The overarching aims of this work will be two-fold: (i) understanding the *microstructure* effects on dissolution, including grain size, strain, UO₂ stoichiometry and impurity species; (ii) understanding the influence of *near field materials* on the dissolution of UO₂ and related analogues.

Major outcomes of this effort include: training of graduate students in engineering and understanding of ceramics related to nuclear fuel; highly relevant publications in peer-reviewed journals; and formal technical exchange between US and UK researchers. Nuclear waste management and UNF is a long-term problem involving a worldwide technical community, and these projects solidify international cooperation towards joint technical goals. Consequently, this program is a joint collaborative enterprise between leading researchers from the US and UK who, collectively, bring mutually complementary and compatible skills, capabilities, and interests required to achieve a paradigm shift in technical understanding of the role of microstructure, fuel chemistry, and local environment on UO₂ dissolution. The team is comprised of researchers at a US university (WSU), a UK university (UoS), and a US national lab (PNNL). Prof. McCloy (WSU) has extensive experience with high temperature ceramic materials, nuclear waste forms, and managing complex technical projects. Dr. Hanson (PNNL) is an expert in dissolution of UNF and has done extensive research relating to US nuclear waste repository siting. Dr. Corkhill (UoS) has deep knowledge of geochemical considerations of UNF durability and Prof. Hyatt (UoS) has extensive experience on ceramic nuclear waste management. The US portion of the research will be funded by the Department of Energy–Nuclear Energy NEUP program, and the UK portion will be funded by Engineering and Physical Sciences Research Council, Research Councils UK Energy Programme.