Integrating Used Fuel Degradation Models into Generic Performance Assessment

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Within the Used Fuel Disposition Campaign of the United States Department of Energy Office of Nuclear Energy (DOE-NE), activities to understand used fuel (UF) degradation and radionuclide mobilization (UFD&RM) have produced/implemented a set of models encompassing radiolytic processes, UF matrix degradation, instant release fractions (IRF) of key radionuclides, and molecular first-principles models for UO_2 and its potential corrosion products. The goals of this collaborative effort among three different national laboratories (Argonne National Laboratory, Pacific Northwest National Laboratory, and Sandia National Laboratories) are to enhance the understanding of UF degradation processes and the technical bases for safety analyses in a range of generic disposal environments. In addition to these modeling efforts, integrated experimental studies are being conducted at both ANL and PNNL to evaluate and validate (and ultimately expand) process models for UF radiolytic phenomena and degradation in various repository conditions. Integration and coupling of these process models into a generic performance assessment model (GPAM) is one focus of SNL efforts within the generic analyses of the Engineered Barrier System (EBS) for various repository environments. As discussed below, the present work has produced a set of models for implementation into the GPAM as an initial step towards an enhanced coupled model of source-term processes[1].

Currently, generic disposal system models (GDSM) for various repository settings include constraints on radionuclide release from UF based on sampled distributions for the general ranges of fractional degradation rates taken from the literature. In addition the GDSM constrain radionuclide mobilization away from the UF using published solubility-limited radionuclide concentrations for those radioelements expected to be reprecipitated under local conditions within the EBS. The potential IRF of key radionuclides have not been substantially incorporated into the GDSM at this point. The current UFD&RM work provides process-model-based fractional degradation rates for UF that account more directly for the effects of the major chemical variables in the source-term. This model may also be directly coupled with the radiolysis model for consideration of other radiolytic conditions directly within GPAM. Implementation of these models into GPAM facilitates enhanced coupling to chemical variables, thermal and pressure variations, as well as more explicit treatment of phase evolution. Moreover, two sets of constraints for the fast/instant release fraction of radioelements (Cs, I, Tc, Sr, C and Cl) based on literature values are provided as distributions for sampling within the GPAM. Furthermore, first-principles models for the bulk chemical properties and surface chemistry of UO_2, as well as for the structures of studtite and metastudtite, provide a rigorous methodology for evaluating/validating continuum scale models of UF degradation, and facilitate the enhanced mechanistic analysis of processes governing fuel degradation.

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