Coupled processes in performance assessment source-term models for geologic repository systems

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To evaluate the expected range of post-closure behavior of the proposed geologic repository for nuclear waste at Yucca Mountain, NV, performance assessment models integrate a wide range of temporally and spatially varying coupled processes into the the evolution of the source-term model. Examples of such processes are evolving hydrologic conditions (e.g., relative humidity, water film formation), corrosion of metallic barriers such as waste packages and drip shields, evolving solution compositions from reaction of materials inside packages (e.g., high-level radioactive waste glass and spent nuclear fuel), waste-form degradation, and secondary phase saturation/precipitation, and radionuclide transport. The continual evaluation of these process interactions delineates the controlling processes for release of radionuclides from the waste forms. For example, dissolution-rate-controlled versus solubility-limited release of radionuclides depends on radionuclide flux away from the waste-forms, which depends on the physical and chemical environment in the drift outside the waste package.

Such processes are incorporated into the performance assessment using simplified representations (abstractions) of detailed process models. Abstractions are implemented to capture the possible range of expected behavior, including principle uncertainties, and are evaluated stochastically. Commonly, coupling is achieved by direct dependence of abstraction model output on major independent variables evaluated in other abstractions. For example, the degradation-rate abstraction for spent nuclear fuel is a function of the solution pH that is obtained from the in-package chemistry abstraction. Demonstration of confidence in the subsystem abstraction models is rooted primarily in confidence developed for underlying process models. Providing confidence in the process models involves robust development from a large range of test data, consideration of primary sources of uncertainty (including conceptual model uncertainty), observations from natural analogs, and independent evaluation/testing of the process model after it is developed. For example, testing for spent-fuel degradation ranges from short-term, flow-through studies of the dissolution rate to long-term (~decade) dripping on spent fuel to constrain radionuclide releases under low water conditions.

Calorimetric study of stability of phases containing exchangeable anions: Sodalite, cancrinite, hydrotalcite

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Because of the importance of radioactive iodine and technetium, which form readily soluble anions (I^-, TcO_4^-) in the environment, it is desirable to find crystalline phases which trap these elements and retard their migration. Furthermore, liquid waste, as in the Hanford tanks, contains high concentrations of anions such as OH^-, NO_3^-, Cl^-, CO_3^{2-}. A number of zeolite - related phases in the sodalite and cancrinite family contain varying amounts of these anions, charge balanced by sodium. Hydrotalcites are layered double hydroxides typically of aluminum and divalent cations (Mg, Fe, Co, Ni, Zn), whose positively charged layers are compensated by interlayer anions. To determine the thermodynamic stability of such anion - containing solids, we have fine tuned the methodology of high temperature oxide melt solution calorimetry. New calorimetric data are shown for hydrotalcites, with variations in the nature of the divalent cation, M, in the M/Al ratio, and in the nature of the anion. New thermodynamic data for nitrate and carbonate containing cancrinite and for sodalites containing OH^- are summarized.