SOURCE TERM THRUST

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Source Term Thrust Area

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The goal of the Source Term Thrust is to enhance the understanding of the performance of nuclear waste forms (mainly spent nuclear fuel (SNF) and nuclear waste glass) and to quantify the release of radionuclides in the evolving near-field environment expected at the proposed nuclear waste repository at Yucca Mountain, Nevada. The behavior of the source term, mainly SNF and vitrified waste, limits radionuclide releases, both initially and over the long term. Interactions of the source term with the near-field environment, such as corroded waste packages, place additional constraints on the long-term behavior, including retention and mobility of important radionuclides.

This program is directed at developing a basic understanding of the fundamental mechanisms of radionuclide release and a quantification of the release as repository conditions evolve over time. Radionuclide release will be critically sensitive to variations in temperature, radiation field, redox conditions, pH, pCO_2, surface area-to-solution volume, and presence of near-field materials. Among the important processes that can control radionuclide release are: (1) kinetics of waste form corrosion, (2) formation of secondary, alteration phases, and (3) reduction and sorption onto the surfaces of near-field materials.

Predictions of the long-term behavior of nuclear waste forms cannot be based entirely on models of laboratory results that are then extrapolated to long periods. Hence, this program will integrate multiple lines of evidence (e.g., results from natural analogue studies) to clarify the scientific basis for waste form degradation mechanisms in relation to source term models. The program will evaluate the uncertainties introduced by parametric uncertainty, variations in environmental conditions, and the use of different conceptual models.

The present source term and near-field models in the Yucca Mountain performance assessment are conservative, but the uncertainties are large. To the extent that the mechanisms of release for specific radionuclides can be understood or the uncertainties decreased, the subsequent analysis of the far-field barriers becomes less important. In the present performance assessment, important processes that will certainly occur are not explicitly included, such as the formation of secondary, U^{6+} alteration phases. Very little credit is taken for the potential chemical interactions between released radionuclides and the alteration products of the SNF and metal waste packages, or their internal components. The conservatism of the present models may be reduced by an improved understanding of the fundamental geochemical/hydrologic processes that will control the corrosion of SNF under oxidizing conditions and the potential interactions that may occur among the corroded nuclear fuel, the high-level-waste (HLW)-borosilicate glass, and the waste package components. This focused and integrated research program will provide the scientific basis for the development of the next generation of more realistic models for source term and near-field processes. Although there has been considerable research on the corrosion of SNF and nuclear waste glasses, there has been much less work on developing models that are applicable to an oxidizing environment, such as that at the proposed Yucca Mountain repository.

There are two compelling reasons for the importance of understanding the source term and near-field behavior. First, essentally all radioactivity is initially in the waste form, mainly SNF or nuclear waste glass. An enhanced understanding and realistic estimates of the extent to which radionuclides will be retained in the waste form or near-field environment reduce demands on the performance of subsequent, far-field barriers. Realistic estimates of radionuclide release can also reduce uncertainties in the Total System Performance Assessment. Second, over long periods, as engineered barriers degrade, it is the waste forms that eventually provide the release of radioactivity to the environment. Thus, it is essential to predict with confidence the physical and chemical evolution of the waste form over hundreds of thousands of years.

Finally, the need to understand the corrosion and alteration of SNF and nuclear waste glasses is a research subject unique to the needs of the Office of Civilian Radioactive Waste Management (OCRWM), Office of Science and Technology and International Program. Other agencies and DOE offices, such as the National Science Foundation (NSF) or Basic Energy Sciences (BES), generally do not fund research on the properties or corrosion of SNF or nuclear waste glasses. Hence, OCRWM, by supporting this program, is creating a community of scientists and engineers who are actively working on and knowledgeable in this field. An important component of this program is the connection to international efforts, particularly to the European Community (EC) programs. We are in the process of connecting our research program to NF-Pro, a four-year integrated project (2004–2007) supported by the EC, as part of their Sixth Framework Program. NF-Pro brings together 40 nuclear research and waste management organizations with the aim of integrating European research on the near field. We have also initiated contact with the EU MICADO (Model uncertainty for the mechanism of dissolution of spent fuel in a nuclear waste repository) program. Through these collaborations, we will leverage present U.S. funding for increased understanding and enhance the international knowledge and reputation of the U.S. program.

The present program, summarized in the following pages, consists of 15 separate programs that involve five national laboratories and five universities (one in Great Britain). In many instances, university investigators have programs closely tied to national laboratories in order to utilize unique facilities for handling highly radioactive materials. Three of the graduate students in this program are supported by OCRWM Fellowships and have completed practica at national laboratories this past summer.

The research programs address four critical areas:

SNF Dissolution Mechanisms and Rates

The initial release of radionuclides is governed by the specific mechanisms and rates of dissolution, which in turn, vary as a function of pH, dissolved oxygen concentration, temperature, solution composition, fuel chemistry (i.e., burn-up), and mode of contact with water. Systematic studies are being conducted of radionuclide release from SNF as a function of these parameters, with special attention being paid to the effects of radiolysis on matrix dissolution rates and the formation of secondary phases. The program also investigates the effects of water that may condense and accumulate on failed spent fuel pins. By the process of deliquescence of hygroscopic fission product phases, alteration products may form on the surfaces of the SNF.

Descriptions of this individual program area are provided in the two-page summaries on pp. 9–16.

Formation and Properties of U^{6+} Secondary Phases

Under an oxidizing environment, the corrosion of SNF leads to the formation of a complicated array of U^{6+} secondary phases that may retard release of radionuclides either by co-precipitation/incorporation, sorption, or by forming a physical barrier to the continued contact with water or release of radionuclides. We have established a multi-pronged approach that includes carefully controlled experiments to determine the means and extent of radionuclide incorporation (e.g., ^{237}Np, ^{239}Pu, and ^{129}I) and sorption into and onto the structures of U^{6+} secondary phases. The experimental work is supported by the application of advanced techniques (e.g., x-ray absorption spectroscopy at the Advanced Photon Source at Argonne National Laboratory, laser-ablation inductively coupled

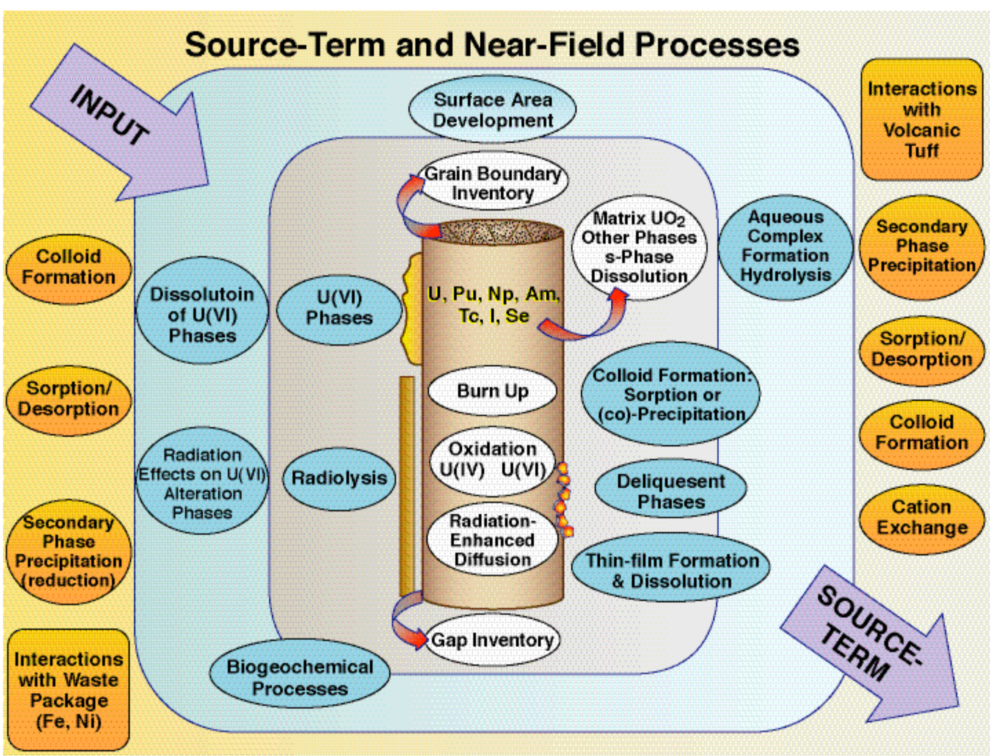

Figure 1. Schematic illustration of the potentially important processes in the Source Term Thrust research program

plasma mass spectroscopy, and *in situ* spectroelectrochemical techniques) in order to determine the extent of incorporation and sorption of radionuclides in/on U^{6+} secondary phases. The stability of these phases is being determined by high-temperature oxide-melt solution calorimetry and reversible solubility measurements, as well as systematic irradiation experiments. Computational simulations involving quantum mechanical calculations are being used to investigate the energetics of the incorporation and sorption processes. In addition, actinide complex stability constants at elevated temperatures are being measured using potentiometry, solvent extraction, spectrophotometric, and nuclear magnetic resonance measurements. The experimental and modeling studies are supported and confirmed by studies of natural occurrences of SNF, namely at the Oklo natural reactors.

Descriptions of this individual program area are provided in the two-page summaries on pp. 17–36.

Waste Form–Waste Package Interactions in the Near Field

The SNF will corrode and release radionuclides in an environment dominated by the presence of uranium, iron, and solutions whose compositions are the result of interactions with the waste forms, waste packages, and the surrounding volcanic tuff. These interactions buffer the environment in which the SNF corrodes and also offer additional opportunities for reduced mobility of radionuclides. We have research programs to investigate the types and behaviors of waste package corrosion products, and their potential for reduction and sorption of key radionuclides (e.g., uptake of ^{99}Tc onto iron oxyhydroxides). We also will investigate the interactions of UO_2 with solutions in contact with volcanic tuff using the pressurized unsaturated flow (PUF) system and *in situ* analytical techniques to investigate and characterize the secondary phases that form. Again, computational simulations of atomic-scale interactions will be used to interpret and support the experimental results.

Descriptions of this individual program area are provided in the two-page summaries on pp. 37–44.

Integration of In-Package Chemical and Physical Processes

The greatest challenge for this program is to integrate the scientific results into a larger scale model of the source term and near-field interactions. To accomplish this, we have immediately begun creating numerical models that capture the important physical and chemical processes that lead to radionuclide release from SNF. The source term model will be coupled to a drift/near-field-scale model that captures the relevant thermal-hydrologic-chemical regimes as a function of time. This integrated model will be used to establish the scientific links among our science programs and provide a basis for understanding the behavior of SNF at different time frames: (1) prior to breach of the waste package; (2) at the time of early breaches of the waste package at elevated temperature; and (3) after breach at long periods, greater than 100,000 years, but at essentially ambient conditions.

Descriptions of this individual program area are provided in the two-page summaries on pp. 45–51.

Acknowledgments

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