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PLUTONIUM STABILIZATION AND STORAGE RESEARCH IN THE DNFSB 94-1 CORE TECHNOLOGY PROGRAM

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INTRODUCTION. Recommendation 94-1 of the Defense Nuclear Facility Safety Board (DNFSB) addresses legacy actinide materials left in the U.S. nuclear defense program "pipeline" when the production mission ended in 1989. The Department of Energy (DOE) Implementation Plan responding to this recommendation instituted a Core Technology program "to augment the knowledge base about general chemical and physical processing and storage behavior and to assure safe interim nuclear material storage, until disposition policies are formulated". The Core Technology program focuses on plutonium, in concert with a complex-wide applied R/D program administered by Los Alamos National Laboratory. This paper will summarize the Core Technology program's first two years, describe the research program for FY98, and project the overall direction of the program in the future.

DESCRIPTION OF THE WORK. The DNFSB 94-1 program focuses on the four DOE sites which hold the bulk of DOE's legacy defense actinide materials: the Rocky Flats Environmental Technology Site (RFETS), the Hanford Plutonium Finishing Plant (PFP), the Savannah River Site (SRS), and the Los Alamos Plutonium Facility (LANL). Each of these sites executes a plan to stabilize its materials and package it for safe 50-year interim storage. Based on input from these four sites and the applied 94-1 R/D program, Core

DISCLAIMER

Technology projects are selected by a systematic process to support DOE's overall stabilization and storage mission. The Core Technology workscope increasingly emphasizes storage (versus stabilization) issues to reflect programmatic needs as residues are stabilized at each site. All Core Technology projects contain each of the following elements:

- Supports at least one core competency key to long-term DNFSB 94-1 success
- Develops the science underlying one or more important DNFSB 94-1 process or issue
- Assures the rapid availability of highly skilled technical response when unforeseen issues arise during execution of the DNFSB 94-1 mission

As discussed below, the seven Core Technology activities being supported in FY98 fit into the three general competency categories of Characterization/Analysis, Aqueous Processes, and Pyrochemistry.

Characterization/Analysis

Pu Chemical States. The purpose of this activity is to identify deleterious changes in the long-term physicochemical state of plutonium in significant classes of residues and stabilized materials. Plutonium X-ray absorption spectroscopy is being developed as the principal tool of this task. Core competency in plutonium characterization is supported.

Pu-Residue Surface Interactions. This project focuses on integrating an array of molecular spectroscopic techniques to assess the interaction of plutonium with important residue classes included combustibles and incinerator ash, as well as stabilized residues. Evaluation of the nitration of organics as a result of plutonium operations also is included in the work scope. Core competency in plutonium characterization is supported.

Oxide-Moisture Interactions. This task uses surface analysis techniques to elucidate the nature of the interaction of water with actinide oxides. This task supports core competency in plutonium materials characterization and addresses the key scientific issue of gas generation in residue stabilization and storage.

Aqueous Processes

Solutions Spectroscopy. This activity enhances plutonium core competency in aqueous plutonium processing and characterization/analysis. The task also develops the science underlying processes currently used in plutonium residue stabilization and storage. Optical spectroscopy is the mainstay of this work.

Separations. The purpose of this activity is enhance core competency in aqueous actinide separations and to minimize secondary waste generation in current residue stabilization. The technical focus is the development of highly selective chelators in the form of functionalized soluble polymers and monolithic foams.

Pyrochemistry

Thermodynamics. This activity develops the science underlying residue stabilization and storage and enhances core competency in actinide thermodynamics and pyrochemistry. The current technical focus is on the behavior of ternary alkali metal-plutonium oxides which may form during stabilization and storage of impure residues.

Salt Distillation Science. The purpose of this project is to develop the science underlying salt distillation technology for stabilizing 94-1 materials and to enhance core competency in plutonium pyrochemistry. X-ray diffraction, electron microscopy, and x-ray photoelectron spectroscopy is used to characterize oxidation reactions and residues resulting from the salt distillation process.

Results. A few of the notable accomplishments of FY97 are as follows. High temperature gas mass spectrometry showed that PuOCl and AmOCl, known to be present in the salt distillation process, have vapor pressures too low to complicate the separation process. Hundreds of electronic absorption spectra of Pu under conditions realistic to aqueous processing have been measured and mathematically analyzed, leading to an unprecedented level of understanding of the Pu species actually present during chemical processing. An array of Pu characterization tools was validated and integrated for application to untreated residues as well as stabilized materials. A technical evaluation summarized the current technical basis and unknowns of corrosion potential during 50-year storage of stabilized residues in stainless steel cans

References.

1. DNFSB Recommendation 94-1. Issued by DNFSB on December 8, 1997.

2. DNFSB Recommendation 94-1 Implementation Plan. Issued by DOE on February 28, 1995.

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