WASTE FORM WORKSHOP REPORT

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CONTENTS

Section Page

ACKNOWLEDGMENTS iii

1 WASTE FORM WORKSHOP SUMMARY 1

2 IN-PACKAGE ENVIRONMENTAL CONDITIONS AT GENERIC SITES (J. PRIKRYL) 1

3 INVENTORY AND GENERAL FEATURES OF COMMERCIAL AND U.S. DEPARTMENT OF ENERGY-OWNED SPENT NUCLEAR FUEL (L. TIPTON) 1

4 DEGRADATION BEHAVIOR OF SPENT NUCLEAR FUEL IN OXIDIZING AND REDUCING ENVIRONMENTS (H. JUNG) 2

5 SIMULATED SPENT FUEL (SIMFUEL) CORROSION TEST (H. JUNG) 2

6 CLADDING PERFORMANCE (T. AHN) 2

7 HIGH-LEVEL WASTE GLASS AND SPENT MIXED OXIDE FUEL INVENTORY AND GENERAL FEATURES (L. TIPTON) 3

8 HIGH LEVEL WASTE GLASS AND SPENT MIXED OXIDE FUEL STRUCTURE AND DEGRADATION BEHAVIOR (T. AHN) 3

9 FUTURE FUEL CYCLE OPTIONS AND IMPACT ON WASTE FORM (T. SIPPEL) 4

10 CERAMIC WASTE FORM OVERVIEW (T. AHN) 4

11 METALLIC WASTE FORM OVERVIEW (H. JUNG) 4

12 THOUGHTS ON SOAR RELATED TO WASTE FORM (C. MARKLEY) 4

13 INTERNATIONAL PROGRAM ENGAGEMENT IN WASTE FORM (T. AHN) 4

APPENDIX—INTEGRATED SPENT FUEL REGULATORY ACTIVITIES WASTE FORM WORKSHOP AGENDA

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QUALITY OF DATA, ANALYSES, AND CODE DEVELOPMENT

DATA: None.

ANALYSES AND CODES: None.

1 WASTE FORM WORKSHOP SUMMARY

The waste form workshop between the U.S. Nuclear Regulatory Commission (NRC) and the Center for Nuclear Waste Regulatory Analyses (CNWRA®) was held on July 26, 2011, at NRC via video connection with CNWRA. The workshop was conducted to support the NRC Integrated Spent Nuclear Fuel Regulatory Activities (ISFR) program, specifically, technical issues on various types of waste forms including spent nuclear fuel (SNF), high-level waste (HLW) glass, and potential new waste forms (e.g., ceramic and metallic waste forms). The workshop also provided an opportunity to disseminate information to NRC and CNWRA staffs to promote integration with other ISFR activities. The purpose of this report is to briefly summarize the topics addressed during the workshop. The meeting ended with a discussion of potential future technical activities relevant to waste forms.

Attendees

NRC: T. Ahn, J. Davis, J. Gwo, Y. Kim, C. Markley, W. Reed, J. Rubenstone, and T. Sippel

CNWRA: X. He, H. Jung, Y. Pan, E. Pearcy, D. Pickett, J. Prikryl, and L. Tipton

The agenda for this workshop is in the appendix.

2 IN-PACKAGE ENVIRONMENTAL CONDITIONS AT GENERIC SITES (J. PRIKRYL)

This presentation provided an overview of disposal concepts, waste package characteristics, groundwater chemistries, and potential in-package geochemical conditions in various potential geologic repository systems. Geochemical conditions relevant to waste form dissolution and subsequent transport of released radionuclides are influenced by various factors that affect the chemistry of water in contact with waste forms in breached waste packages. These factors include the rate of groundwater movement in the near field, interactions of groundwater with components of the engineered barrier system (e.g., buffer/backfill), and interactions of groundwater or buffer-conditioned porewater with the products of canister corrosion. Geochemical parameters (e.g., pH, constituent concentrations, oxidation-reduction potential) and transport conditions relevant to developing models for waste dissolution were described for proposed and potential geologic repository systems hosted in crystalline rocks, argillaceous (clay) formations, and salt rocks.

3 INVENTORY AND GENERAL FEATURES OF COMMERCIAL AND U.S. DEPARTMENT OF ENERGY-OWNED SPENT NUCLEAR FUEL (L. TIPTON)

General features of commercial spent nuclear fuel (CSNF) and U.S. Department of Energy (DOE) SNF were described, including the variety of sources and configurations in the DOE SNF inventory. Known and projected total inventories of DOE SNF and CSNF were presented. For DOE SNF, the present inventory is well known and the projected inventory increase is small. Projected scenarios from the literature indicate that the CSNF inventory will increase substantially over the next several decades. Generally, there is a trend for increasing burnup and enrichment for CSNF. For the higher burnup fuels, effects like the formation of the rim structure should be considered due to potential effects on fuel performance. For example, the rim structure forms due to locally high burnup in the outer radial region of the fuel pellet. This effect leads to changes in the fuel grain structure and increased porosity in the outer rim of the fuel and could contribute to increased release of fission gases and radionuclides. Additionally, several radionuclide inventories for representative CSNF (average and high-burnup) assemblies were modeled using ORIGEN-ARP. The SOAR model was exercised to compare the relative effects of average and high-burnup CSNF on long-term geologic disposal performance. Very minor increases in total annual dose were observed for the high-burnup CSNF case compared to the lower burnup case.

4 DEGRADATION BEHAVIOR OF SPENT NUCLEAR FUEL IN OXIDIZING AND REDUCING ENVIRONMENTS (H. JUNG)

In a geologic repository for SNF, various environmental and material factors can affect SNF degradation behavior, including redox conditions, water chemistry, temperature, radiolysis, initial SNF condition, sorption of radionuclides onto iron oxide, and hydrogen gas generation. For long-term prediction of SNF degradation behavior and its dissolution rate, representative degradation models applicable to both initial oxidizing and later longer term reducing environments were reviewed, including an electrochemistry-based mixed potential model and a chemically dissolved matrix alteration model. Uncertainties or lack of knowledge for some aspects of both models were also identified, such as effective exposed surface area of SNF due to long-term evolution of surface, precipitation, and grain boundary openings; radiolysis efficiency; hydrogen gas; and high burnup effects.

5 SIMULATED SPENT FUEL (SIMFUEL) CORROSION TEST (H. JUNG)

Major findings of CNWRA corrosion tests of SIMFUEL as an analog of SNF were presented. Tests were conducted in carbonate-based solutions with and without addition of calcium and silicon using electrochemical methods. The primary purpose of the study was to confirm a range of dissolution rates of SNF in a repository environment, specifically under oxidizing conditions. The measured dissolution rates in this study were consistent with rates obtained under similar conditions in the literature. The results of posttest solution chemistry analysis revealed that uranium was a dominant species dissolved in the posttest solutions and the converted dissolution rates based on uranium concentration were consistent with the results of dissolution rates obtained from the impedance measurements. Fission product analogs, such as barium, molybdenum, strontium, and zirconium, were also dissolved at a relatively high rate. Sorption test results showed that uranium concentration in the posttest solution with a stainless steel disk decreased about 20 percent. Future work under an expanded range of conditions was identified.

6 CLADDING PERFORMANCE (T. AHN)

Cladding performance was reviewed and evaluated. Data, existing models, and some calculations for degradation kinetics were presented for important attributes in the geological disposal system. The attributes include

- Initial failure rate
- C-14 source term
- General corrosion and oxidation
- Hydride reorientation
- Delayed-hydride cracking

- Hydrogen absorption
- Impact mechanical failure
- Thermal and a thermal creep failure
- Effects of oxidation and hydration of SNF matrix
- Other corrosion failure, such as pitting

The risk significance of these attributes was illustrated by exercising the SOAR model and comparing the results with experimental data on radionuclide release through partially failed cladding. The likelihood of criticality was also discussed. Hydrogen-related degradation was identified to be important in radionuclide release and criticality control. Others except hydrogen-related degradation are relatively insignificant within uncertainties.

7 HIGH-LEVEL WASTE GLASS AND SPENT MIXED OXIDE FUEL INVENTORY AND GENERAL FEATURES (L. TIPTON)

General features of HLW glass and spent mixed oxide (MOX) fuel were described. The discussion included a general description of HLW glass production, U.S. production sources, and variability in glass composition. Spent MOX production was also discussed, including typical plutonium source streams (weapons and commercial reprocessing) and compositions. Known (expected) inventories and sources of HLW glass and spent MOX were presented. Much of the HLW glass and nearly all of the expected spent MOX waste forms have not yet been produced. Therefore, there may be some variability in radionuclide loading of the HLW. Potential effects of reprocessing on generation of additional HLW glass inventory and spent MOX were discussed. For higher burnup spent MOX, fuel gas pressure may increase more rapidly with increasing burnup than for comparable CSNF. Spent MOX also exhibits the formation of the rim structure at higher burnups. Potential inhomogeneity in mixing of spent MOX PuO_2 and UO_2 can lead to clusters of PuO_2, which can experience locally high burnup and result in regions of high porosity. Additionally, radionuclide inventories were modeled for a representative West Valley Demonstration Project HLW glass waste form, a commercial spent MOX waste form derived from reprocessing, and a weapons spent MOX derived from weapons plutonium.

8 HIGH LEVEL WASTE GLASS AND SPENT MIXED OXIDE FUEL STRUCTURE AND DEGRADATION BEHAVIOR (T. AHN)

Degradation of HLW glass and spent MOX was reviewed and evaluated. Data, existing models, and some quantitative degradation kinetics were presented for important attributes in the geological disposal system. The attributes include (i) types of HLW glass, (ii) borosilicate glass, (iii) mass and radionuclide inventory, (iv) dissolution mechanisms based on silica dissolution, (v) comparison of dissolution rate with other waste forms, (vi) dissolution kinetics controlled by affinity of dissolved silica species, (vii) colloid formation, (viii) vapor hydration, (ix) lanthanide borosilicate (LaBs) glass, (x) radiation-induced swelling/compaction and cracking, and (xi) sMOX dissolution mechanism. Finally, SOAR model exercises were conducted for HLW glass and sMOX. Within the uncertainties, the results are consistent with international assessments in generic disposal systems.

9 FUTURE FUEL CYCLE OPTIONS AND IMPACT ON WASTE FORM (T. SIPPEL)

The results of modeling several fuel cycles (for benchmarking purposes only) with different codes were presented at the U.S. Nuclear Waste Technical Review (NWTRB) Board Meeting, held on June 6–7, 2011. The codes presented at the meeting included VISION by Idaho National Laboratory, CAFCA by Massachusetts Institute of Technology, ORION by National Nuclear Laboratory of UK, NUWASTE by NWTRB, and an Excel spreadsheet by AREVA. Because all the tested codes gave similar results, we conclude that a number of codes can model different fuel cycle options. The discussion of this presentation focused on the parameters that affected performance and how different fuel cycle options impact those parameters. Reprocessing and factors that affect the heat generation rate were discussed in detail. The effects of reprocessing would include new waste forms and more radiotoxic inventories, but less waste requiring deep geologic disposal. NRC and CNWRA staff attended the NWTRB benchmarking meeting as observers.

10 CERAMIC WASTE FORM OVERVIEW (T. AHN)

Degradation of ceramic waste forms was reviewed and evaluated as an alternative waste form from various fuel cycle options. Data, existing models, and some degradation kinetics were presented on (i) types of ceramic waste forms, (ii) dissolution rate in comparison with other waste forms, (iii) pH and temperature effects on dissolution rate, and (iv) radiation-induced swelling and fracture. Finally, a SOAR model frame was presented, as bounded by HLW glass dissolution.

11 METALLIC WASTE FORM OVERVIEW (H. JUNG)

An overview of metallic waste forms was presented including development, background, and current status of SS-Zr, Zr-Cu-(Fe), and Fe-Mo-(Tc or Re) alloys. The main objective or advantage of metallic waste forms is to immobilize radionuclides within the metal alloys, uniformly distribute them, and increase waste loading compared to HLW glass. Based on limited knowledge and experimental data, metallic waste forms are, in general, highly corrosion resistant.

12 THOUGHTS ON SOAR RELATED TO WASTE FORM (C. MARKLEY)

To effectively integrate waste form study with performance assessment in SOAR, it was recommended that waste form study consider important scenarios in a repository and identify potential regulatory issues as well. SOAR should be used to achieve better understanding of the waste form domain in a quantitative manner.

13 INTERNATIONAL PROGRAM ENGAGEMENT IN WASTE FORM (T. AHN)

NRC/CNWRA engagement in international programs was reviewed. Two areas were highlighted: (i) dissolution of SNF and HLW glass with organizations of the European Commission, France, and Japan and (ii) cladding degradation with organizations of the European Commission and Japan. Current engagements are mainly for information exchange. Future development includes collaboration in selected areas. Staff discussed and identified future data needs.

APPENDIX
INTEGRATED SPENT FUEL REGULATORY ACTIVITIES
WASTE FORM WORKSHOP AGENDA

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Tuesday, July 26, 2011, 9:15 a.m.–4:30 a.m. NRC (Rockville, Maryland)
CNWRA (San Antonio, Texas)

Time	Topics	Presenter
9:15–9:30 a.m.	Introduction of the workshop	Jack Davis, et al.
9:30–10:00 a.m.	In-package environmental conditions at generic sites	Jim Prikryl
10:00–10:30 a.m.	Inventory and general features of commercial and DOE-owned SNF	Lynn Tipton, Tim Sippel
10:45–11:15 a.m.	Degradation behavior of SNF in oxidizing and reducing environments	Andy Jung, Tae Ahn
11:15–11:45 a.m.	Simulated spent fuel (SIMFUEL) corrosion test	Andy Jung
1:00–1:30 p.m.	Cladding performance	Tae Ahn, Andy Jung
1:30–2:00 p.m.	HLW glass and spent MOX inventory and general features	Lynn Tipton, Tim Sippel
2:00–2:30 p.m.	HLW glass and spent MOX structures and degradation behavior	Tae Ahn
2:45–3:15 p.m.	Future fuel cycle options and impact on waste form	Tim Sippel, Lynn Tipton
3:15–3:30 p.m.	Ceramic waste form overview	Tae Ahn
3:30–3:45 p.m.	Metallic waste form overview	Andy Jung
3:45–4:00 p.m.	Thoughts on SOAR related to waste form	All, PA
4:00–4:15 p.m.	International program engagement in waste form	Tae Ahn, Andy Jung
4:15–4:30 p.m.	Potential tasks considering NRC priorities	All