

MONITORING THE CONSISTENCY OF MULTIPHASE WASTE FORMS

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For presentation at

American Nuclear Society

2001 Winter Meeting

Reno, NV

11/11-15/01

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This work was supported by the U.S. Department of Energy under contract W-31-109-ENG-38.

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INTRODUCTION

Methods are being developed for demonstrating that nonstandard high-level radioactive waste (HLW) forms meet the intent of the product consistency requirement in the Waste Acceptance System Requirements document (WASRD). That requirement was established for borosilicate HLW glasses “to ensure a consistent glass product by controlling the vitrification process... Consistency is necessary to reflect consideration for the waste package designs.” [1] The test method specified in the WASRD for HLW glasses is the 7-day product consistency test (PCT). To meet the WASRD requirement, the response of an HLW glass in the PCT must be less than that of the environmental assessment (EA) glass. The EA glass is used as a benchmark “so that conservative but realistic assessments of the engineered barrier system performance can be made.” [1]

The PCT and the WASRD requirement were developed to bound the behaviors of the wide range of borosilicate HLW glasses that will be produced at DOE facilities for the purpose of repository design. However, the need to demonstrate that the physical, chemical, and radiological properties of HLW forms have been constrained within acceptable (i.e., as-qualified) ranges will probably apply to all HLW waste forms. The PCT may not be the appropriate method for nonstandard HLW waste forms being developed for DOE waste streams that are not amenable to direct vitrification. For example, the multiphase ceramic waste form developed for excess weapons plutonium or for glass/ceramic waste forms that contain substantial amounts of included crystalline phases may require other methods for demonstrating consistency.

DESCRIPTION OF ACTUAL WORK

Two multiphase waste forms have been developed to immobilize conditioned spent sodium-bonded nuclear fuel from the Experimental Breeder Reactor-II (EBR-II): a ceramic waste form (CWF) for waste chloride salts and a metallic waste form (MWF) for cladding hulls recovered from the electrorefiner. Insights gained from tests conducted to

determine the corrosion mechanisms of these waste forms are being used to evaluate potential methods for meeting the WASRD product consistency requirements, namely, that the waste forms are produced consistently and that their impact on the performance of the disposal system can be calculated.

RESULTS

The CWF is composed of approximately 70% sodalite, 25% borosilicate glass binder, and contains a total of about 5% halite and various rare earth and actinide oxides and silicate inclusion phases. The 7-day PCT is appropriate for monitoring the consistency of CWF products for several reasons: (1) The corrosion mechanisms of sodalite and glass binder are the same as that of HLW glasses. As for HLW glasses, the chemical durability of the CWF is the key property to monitor and can be measured using its dissolution rate. It varies with the compositions and abundance of the sodalite and glass binder phases. (2) The release of radionuclides is bounded by the degradation rates of the sodalite and glass binder matrices. The chemical durability of HLW glasses as modeled in performance assessment calculations can be shown to provide an upper bound to the release rates of radionuclides from the CWF over the range of potential environmental conditions. (3) The 7-day PCT with CWF is repeatable, reproducible, and sensitive to the composition of the CWF. The measured precision for PCTs with CWF is similar to that with borosilicate glasses. The PCT can be used to verify that the CWF was correctly batched and processed and that the CWF has acceptable chemical durability relative to the EA glass.

The MWF is comprised primarily of two alloys: stainless steel (from the cladding hulls) and an Fe₂Zr intermetallic phase (Zr from treated blanket fuel). The corrosion behaviors of these phases differ greatly from that of HLW glass and the PCT is not appropriate for monitoring its consistency. Besides the practical difficulty in producing finely divided MWF to achieve the surface-to-volume ratio called for in the test, its chemical durability precludes dissolving enough material to be reliably measured in a 7-day test at 90°C. Much more aggressive conditions are required to produce a measurable extent of corrosion. Also, matrix components cannot be used to bound the release of

radionuclides from the MWF, unlike HLW glasses and the CWF. Alloy components are released from the MWF by a two-step oxidation-dissolution mechanism. Thus, the release rates of individual radionuclides must be measured directly.

The abundance of the Fe₂Zr intermetallic phase may be a better measure of MWF consistency because this phase sequesters the majority of radionuclides. Separate actinide-bearing inclusion phases are formed if there is an insufficient amount of Fe₂Zr intermetallic phase. Although those phases may be as durable as the Fe₂Zr intermetallic phase, the resulting microstructure may be significantly different than that of the MWF to be described in the waste form qualification report. Additional Zr will be added to the MWF to ensure that a sufficient amount of Fe₂Zr intermetallic phase forms. Several techniques to monitor the amount of Fe₂Zr intermetallic phase and the Zr content are being evaluated.

CONCLUSIONS

An understanding of the corrosion mechanism of multiphase waste form can be used to facilitate development of a method to monitor its consistency and to meet the requirements of production control and durability for waste form acceptance. This approach is currently being used for qualification of two nonstandard waste forms to be produced with sodium-bonded spent nuclear fuel, and it can be applied to other multiphase waste forms.

REFERENCES

1. *Waste Acceptance System Requirements Document (WASRD), Rev. 04D (draft)*, U.S. Department of Energy, Office of Civilian Radioactive Waste Management report E00000000-0081101708-00001, February 2000.