Initial Results of Metal Waste Form Development Activities at ANL-West

by

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DISCLAIMER

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INTRODUCTION

Argonne National Laboratory is developing a metal alloy to contain metallic waste constituents residual from the electrometallurgical treatment of spent nuclear fuel [1,2]. This alloy will contain stainless steel (from stainless steel-clad fuel elements), ≈15 wt.% zirconium (from alloy fuel), fission products noble to the process (e.g., Ru, Pd, Tc, etc.), and minor amounts of actinides. The alloy will serve as a final waste form for these components and will be disposed of in a geologic repository. The alloy ingot is produced in an induction furnace situated in a hot cell using Ar cover gas.

This paper discusses results from the melting campaigns that have been initiated at ANL-West to generate the metal waste form using actual process materials. In addition, metal waste form samples have been doped with Tc and selected actinides and are described in the context of how elements of interest partition between various phases in the alloy and how this distribution of elements in the alloy may affect the leaching behavior of the components in an aqueous environment.

EXPERIMENTAL RESULTS

Two full-scale ingots have been cast in an induction furnace located in a hot cell called the Fuel Conditioning Facility (FCF). The first cast ingot consisted of unirradiated cladding hulls and Zr to make a Type 316 stainless steel (SS) - 15 wt.% Zr alloy ingot. A depleted uranium-10 wt.% Zr alloy was electrolytically dissolved from the cladding hulls in the electrorefiner before the cladding hulls were run through a distillation furnace (the cathode processor) to drive off adhering LiCl-KCl eutectic salt. Approximately 4 kg of the cleaned hulls were then loaded into an yttria crucible along with around 0.7 kg of Zr. The charged crucible was then placed in a drilled-out graphite susceptor that was coated with zirconia mold wash, and heated to 1600°C in an induction furnace and held at temperature for approximately 0.5 hours. A controlled cooldown sequence was used to control cracking of the yttria crucible, which has low thermal shock resistance. The resulting ingot was homogeneous, had limited porosity, and exhibited negligible evidence of a significant slag layer.

The second ingot cast in the FCF casting furnace contained Type 316 stainless steel cladding from fuel elements irradiated to around 8.0 at.% burnup in the Experimental Breeder Reactor II located at ANL-West. Around 1.9 kg of the irradiated cladding hulls were loaded into an yttria crucible and heated to 1600°C and held at temperature for 0.5 hours. Consolidation of the hulls into an ingot was observed.

Additional melting experiments have been completed on a small-scale using actual irradiated cladding hulls and a muffle furnace located in a hot cell. The goal of these investigations was to determine the optimal furnace operating temperatures and hold times required to most efficiently melt irradiated cladding hulls. 5 to 10 cladding hulls at a time were heated to 1600°C in small yttria crucibles and held at temperature for 0.5 and 2.0 hours. One test was conducted at 1700°C. The results of this work showed that the irradiated hulls that come out of the electrorefiner and distillation furnace located in FCF can be difficult to melt. Unirradiated hulls that have been through the same two pieces of equipment melt more easily. As a result, it appears that it is not the distillation process that keeps the hulls from melting, but instead it seems to be something that is different about the irradiated hulls over the unirradiated hulls.

Once the issue of melting irradiated hulls is resolved, the overall performance of the metal waste form alloy in a geologic repository will be one of the more important issues to understand. To this end, 10 and 20 gram buttons of metal waste form alloy have been generated using resistance and induction furnaces. These buttons have been doped with the long-lived fission product technetium (Tc), which has a half-life of 2.15×10^5 years (this fission product, as an oxide, is observed to leach out of glass waste forms in aqueous environments), and samples have also been generated that contain the actinides U, Pu, and Np. Corrosion tests have been performed on selected samples. Analysis of Tc-doped samples using scanning electron microscopy with energy dispersive spectroscopy showed that the Tc was uniformly distributed between the metal waste form alloy phases in the as-cast buttons, and the Tc exhibited a slight preference for an Fe_2Zr phase over an Fe solid solution phase. Corrosion testing of Tc-containing samples showed very little leaching of the Tc in static immersion tests using de-ionized water. For the actinide-containing samples, U, Pu, and Np were observed to uniformly distribute into the Fe_2Zr phase of the alloy, and when corrosion tested in a high temperature steam environment, a metal waste form sample containing 10 wt.% Pu displayed negligible signs of corrosion[3].

REFERENCES

1. S. M. McDeavitt, D. P. Abraham, and D. D. Keiser, Jr., "Alloy Waste Forms for Metal Fission Products and Actinides Isolated by Spent Nuclear Fuel Treatment," in: *Proceedings of the Second International Symposium on Extraction and Processing for the Treatment and Minimization of Wastes*, Scottsdale, AZ, Oct. 27-30, 1996, TMS, Warrendale, PA, (1996), p. 177. (4)

2. S. M. McDeavitt, D. P. Abraham, D. D. Keiser, Jr., and J. Y. Park, "Stainless Steel-Zirconium Alloy Waste Forms," in *Proceedings of SPECTRUM '96: International Topical Meeting on Nuclear and Hazardous Waste Management*, Seattle, WA, August 18-23, 1996, American Nuclear Society, LaGrange Park, Illinois, (1996), p. 2477. (4)

3. D. Wronkiewicz, Argonne National Laboratory, Chicago, IL, unpublished research.