Corrosion Properties of Metal Waste Alloys for Long-lived Radionuclides Immobillization

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1. Introduction

Technologies used in solidification processing by melting spent nuclear fuel assembly materials and hulls for metal waste reduction that occurs during the pyro-processing have been studied, mainly in the United States [1-3]. A cladding hull in a spent fuel assembly to be applied to a domestic PWR is composed of a metal alloy such as Zircaloy-4, ZIRLO accounts for the highest volume and weight ratio in the generation of process waste. Recently, at KAERI, we have been conducting research on the recycling of a cladding hull as a solidification host matrix in order to solidify and handle highly radioactive anode sludge residue including noble metal (NM) that occurs from electro-refining from pyro-processing. If we produce an MWF (Metallic Waste Form) by adding and melting the alloying elements and anode sludge in the cladding hull, the effect on the volume reduction and weight loss can be maximized because the cladding hull can be 100% recycled to the solidification host matrix of MWF. According to the existing results of MWF alloys manufactured in the US, although a stable production of MWF is possible with SS-15Zr alloy, it is disadvantageous in terms of the domestic cladding hull recycling efficiency, because the Zr content is low. In addition, the mechanical stability and corrosion resistance of Zr-8SS is relatively low [3]. As a result of the new composition with Zr-Cr-Si-NM in KAERI, a stable MWF was formed without cracks. Moreover, it was possible to confirm the possibility that the cladding hull and anode sludge generated from the pyro-processing can be fabricated MWF alloy with an excellent corrosion resistance.

2. Experimental Results

The ZIRLO-SS304-INCONEL718 composition of the 9 kinds of solidification host matrix compositions has the eutectic point composition. All specimens were fabricated by high frequency induction heating in a silica crucible in an Ar atmosphere after forming a vacuum of 5×10^{-5} torr. To ensure the integrity of the specimen, it was held at 1600 to 1800℃ for 5 minutes. After forming the melt completely, it was kept at 1350℃ for about 15 minutes, furnace cooling was then applied to reduce the solidification shrinkage cracks and porosity. All prepared specimens were cut and polished, and XRD, SEM, and EDS analyses were performed for a microstructure and phase analysis.

For an evaluation of the corrosion properties, potentiodynamic (PD) and potentiostatic (PS) tests were carried out in the acidic brine aqueous solution condition (0.0001 mol•kg^{-1} H_2SO_4 + 0.01 mol•kg^{-1} NaCl in demineralized water, pH). The main phases composing the Zr-Cr-Si-NM alloy consist of α-Zr, $ZrCr_2$, and Zr2Si. The eutectic structure and $ZrCr_2$ phase increased with increasing Cr content. Pd was concentrated on α-Zr, Ru on α-Zr and $ZrCr_2$, Re on Zr_2Si phase. Electrochemical accelerated corrosion tests such as PD and PS were carried out on the Zr-Cr-Si and Zr-Cr-Si-NM alloys prepared in this study. The Zr-Cr-Si-NM alloys exhibited a high corrosion resistance and low corrosion rate owing to their low I_{corr} values with high E_{corr} values compared to conventional materials. As a result of the analysis of the composition of the leachate after the PS test, there was no NM element leaching below 500 mV, or leaching of the elements excluding Re, which is a surrogate Tc at 800 mV. In particular, the Zr_2Si

phase, which is encapsulating phase of Re, was very stable, and showed no shape change or leaching of Re after the corrosion test, suggesting that the Tc immobilization ability is excellent.

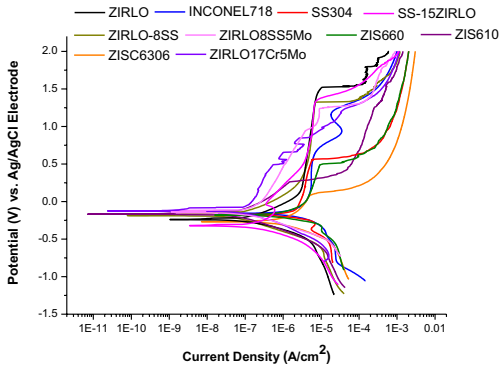

Fig. 1. Potentiodynamic test results of all specimens.

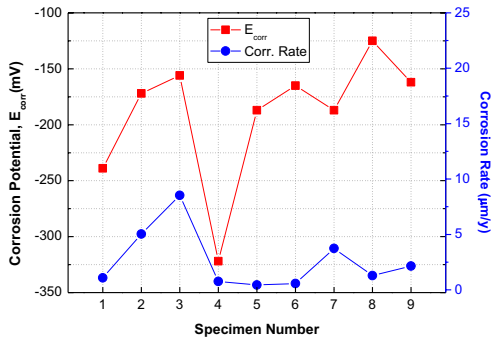

Fig. 2. Corrosion potential and Corrosion rate values of all specimens.

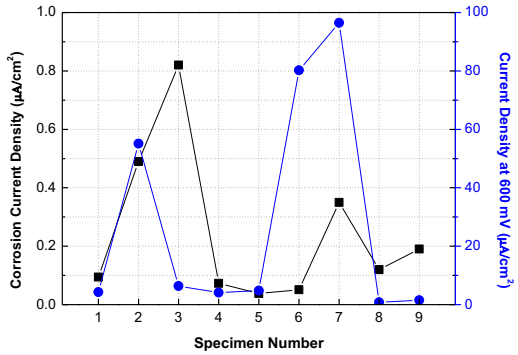

Fig. 3. Corrosion current density and current density at 600 mV of all specimens.

3. Conclusion

The main phases composing the Zr-Cr-Si-NM alloy consist of α-Zr, $ZrCr_2$, and Zr2Si. The eutectic structure and $ZrCr_2$ phase increased with increasing Cr content. Pd was concentrated on α-Zr, Ru on α-Zr and $ZrCr_2$, Re on Zr_2Si phase. Electrochemical accelerated corrosion tests such as PD and PS were carried out on the Zr-Cr-Si and Zr-Cr-Si-NM alloys prepared in this study. The Zr-Cr-Si-NM alloys exhibited a high corrosion resistance and low corrosion rate owing to their low I_{corr} values with high E_{corr} values compared to conventional materials.

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