Be-free Brazing Process Proposed for CANDU Fuel Bundle Manufacture

Jung-Gu Lee [a*], Min-Ku Lee [a], Chang-Kyu Rhee [a], Choon-Ho Park [b]
[a]*Nuclear Materials Development Division, Korea Atomic Energy Research Institute, Daejeon, 305-353, Korea*
[b]*KEPCO NF, Daejeon, 305-353, Korea*
[*]*Corresponding author: jglee88@kaeri.re.kr*

1. Introduction

In manufacturing CANDU fuel bundles, various appendages (spacers and bearing pads) are brazed onto the surface of the cladding tubes made of Zircaloy-4. For the brazing filler metal (BFM), physically vapor-deposited (PVD) metallic beryllium (Be) has currently been used. Unfortunately, the utilization of PVD Be as a BFM requires very complicated physical protection and safety systems, as its metallic vapor is highly toxic to the human body and is extremely harmful to the environment [1]. This hazard of airborne Be has been a primary trigger for our recent studies to find a new BFM as a substitute for the PVD Be [2-4]. In this paper, an alternative Be-free brazing process, where Zr-based multi-component amorphous sputter coatings are employed as a novel BFM, is proposed for CANDU fuel bundle manufacturing.

2. Methods and Results

2.1 Selection of Filler Compositions

Based on a number of reported Zr-based alloys, practical considerations have been addressed to determine suitable constituent elements of a BFM for CANDU fuel bundle manufacturing. First, the Be should be excluded from the filler composition, and the elements of the filler need to have a low neutron absorption cross section ($\sigma_a < 10$ barns), a low possibility of hydrogen uptake, and an adequate ability to reduce the melting temperatures of the filler alloys. According to these criteria, the alloying elements of Ti, Cu, Fe, and Al were selected properly, and the four Zr-rich ternary or quaternary compositions were determined by exploring their low eutectic temperatures in the given alloy systems (Table I).

Table I: Compositions of the Filler Alloys (at.%)

Filler	Zr	Ti	Cu	Fe	Al	Liquidus (°C)
Fe-1	74	-	13	13	-	880
Fe-2	58	16	10	16	-	850
Al-1	60	-	30	-	10	930
Al-2	57	3	25	5	10	880

2.2 Sputtering of Multi-component Zr-based Alloys

By sputtering the multi-component crystalline targets prepared by vacuum arc melting as well as a powder metallurgical process, the homogeneous and amorphous-structured layers were uniformly coated onto the Zircaloy-4 substrate, as shown in Fig. 1. By preserving the near equivalent target composition, the coating showed an excellent compositional uniformity in the spatial distribution, as well as almost the same melting characteristics as those of the parent target.

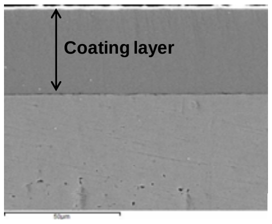

Fig. 1. Zr-based amorphous sputter coating layer on the Zircaloy-4 substrate.

2.3 Structure of the Brazed Joints

By applying amorphous sputter coatings, highly homogeneous joints were obtained with the formation of predominantly grown α-Zr grains owing to a complete isothermal solidification (Fig. 2). At the early stage of brazing, the Zircaloy-4 base metal dissolved considerably into the molten filler. Such a compositional change initiated an isothermal solidification of the Zr-rich molten filler. When the α-Zr phases nucleated and grew into a joint, excessive filler elements were expelled from the growing α-Zr phases and segregated at an intercellular region owing to their limited solubility in the α-Zr. As a result, fine lamellar α-Zr and γ phases were formed from transformed β-Zr phases at an intercellular region by the eutectoid reaction of $\beta\text{-Zr} \rightarrow \alpha\text{-Zr} + \gamma$ at the subsequent cooling stage.

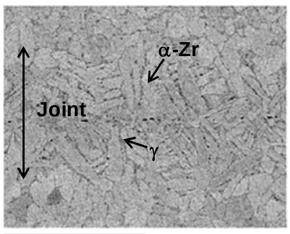

Fig. 2. Joint structure brazed using a Fe-2 sputter coating.

2.4 Properties of the Brazed Joints

According to the tensile tests for the brazed Zircaloy-4 samples, fully developed stress-strain curves were obtained regardless of the testing temperatures up to 400 °C, indicating that a fracturing did not occur in the joint areas, but in the base metal with a considerable plastic deformation (Fig. 3). It is obvious that the bonding strengths of the joints were higher than those of the bulk Zircaloy-4. This result signified an excellent durability of the present joints at a high operating temperature of a PHWR (250 to 350 °C).

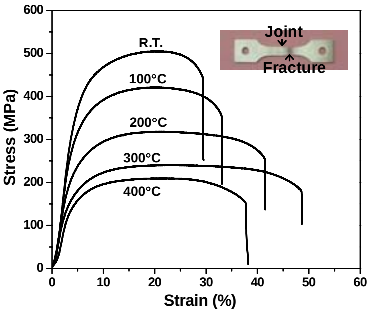

Fig. 3. Stress-strain curves obtained by tensile tests for the samples brazed using an Al-2 sputter coating.

In contrast with the tensile properties, the corrosion resistance of the joints was sensitively affected by the compositions of the applied filler metals. Some elements in the filler metals caused the selective leaching in the joint area, but the others showed little effect on the corrosion resistance of the brazed joints. Autoclave test results according to ASTM G2 specification indicate that the corrosion rate of the joined Zircaloy-4 sample brazed by one of the optimized Zr-based amorphous filler metals was almost same as that of the bulk Zircaloy-4, and even lower than that of the sample brazed by the $Zr_{63}Be_{37}$ filler metal (Fig. 4). This result demonstrates the excellent corrosion resistance of the present joints in a PHWR operating environment.

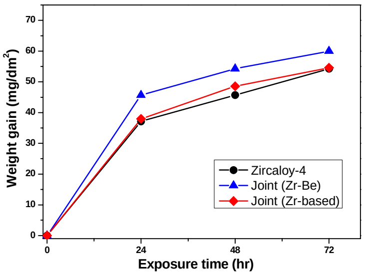

Fig. 4. Weight gains of the brazed samples together with the result of the bulk Zircaloy-4.

2.5 Prototype Manufacture of CANDU Fuel Rods

Based on the fundamental studies above, we tried to braze the appendages onto the cladding tubes using the Zr-based amorphous sputter coatings as a filler for CANDU fuel bundle manufacturing. The brazing procedure including punching, coining, tack-welding and brazing heat-treatment was set up based on the standard fuel fabrication process used at KEPCO NF in Korea. The results were quite promising, such that the appendages were successfully brazed without any defects in the entire joint area (Fig. 5). In some brazing conditions, however, a considerable amount of pores were generated, and further study is necessary to find the optimum brazing conditions when using the Zr-based amorphous sputter coatings as a brazing filler.

Fig. 5. Prototype fuel cladding tubes.

3. Conclusions

Amorphous sputter coatings of Be-free multi-component Zr-based alloys were applied as a novel brazing filler metal for brazing Zircaloy-4. By applying homogeneous and amorphous-structured layers coated by sputtering the crystalline targets, highly reliable joints were obtained with the formation of predominantly grown α-Zr grains owing to a complete isothermal solidification, exhibiting high tensile strength as well as excellent corrosion resistance. By applying the Zr-based amorphous sputter coatings, the appendages were successfully joined to the fuel cladding tubes using the standard CANDU fuel brazing procedure. All of these results show that the Be-free and Zr-based multi-component amorphous sputter coatings can offer great potential for the brazing and manufacturing of a CANDU fuel bundle system.

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