Annealing Effect on Corrosion Behavior of the Beta-Quenched HANA Alloy

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1. Introduction

The advanced fuel cladding materials named as HANA cladding have been developed at KAERI for application of high burn-up and that cladding showed an improved performance in both in-pile and out-of-pile conditions [1]. However, the cladding performance could be changed by the annealing conditions during the tube manufacturing process. Especially, the corrosion resistance is considerably sensitive to their microstructure which is determined by a manufacturing process in the high Nb-containing zirconium alloys [2, 3]. They reported that the corrosion properties of the Nb-containing Zr alloys were considerably affected by the microstructure conditions such as the Nb-concentration in the matrix and the second phase types. Therefore, the corrosion behavior of HANA cladding having the high Nb could be considerably affected by the annealing time and temperatures. The purpose of this study is focused on the annealing effect of the beta-quenched HANA alloy to obtain the optimum annealing conditions.

2. Methods and Results

The manufacture of the HANA alloy (HANA-4; Zr-1.5Nb-0.4Sn-0.2Fe-0.1Cr) involved a vacuum sequence in which the alloy was melted four times to promote the homogeneity of the alloying element. The melted ingot was forged at β-phase region and then quenched in a water from the β solution region at 1050℃.

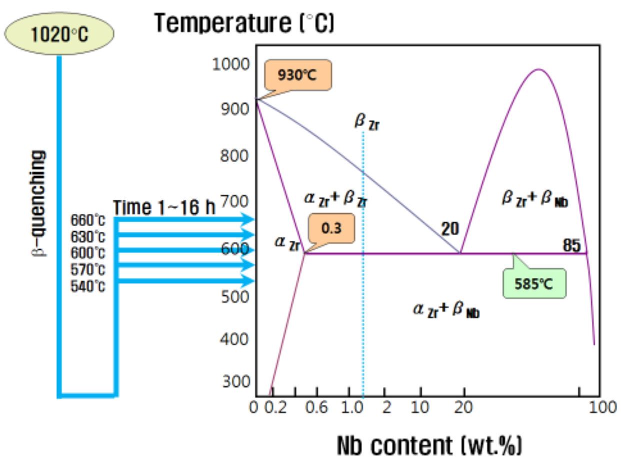

Fig. 1 Schematic illustration of annealing conditions of HANA-4 (Zr-1.5Nb-0.4Sn-0.2Fe-0.1Cr) alloy after β-quenching

The quenched ingot was cut in to dimensions of 16 mm x 28 mm x 12 mm and then aged in a vacuum environment at various temperatures and times as shown in Fig. 1.

The corrosion test was performed in a static autoclave with a 360℃ water environment under saturated pressure of 18.9 MPa. The size of corrosion test specimens was of 15 mm x 25 mm x 1 mm after mechanically ground with 1200 grit SiC paper for the aged samples. The ground specimens were then pickled in a mixed solution of H_2O (40 vol. %), HNO_3 (30 vol. %), HCl (25 vol. %) and HF (5 vol. %). The corrosion resistance was evaluated by measuring the weight gain of the corroded samples after suspending the corrosion test at a periodic interval.

The microstructure observation for the each aged sample was performed by using transmission electron microscope (TEM) equipped with energy dispersive spectra (EDS) capabilities. The specimens for the TEM observation were prepared by being mechanically thinned to about 70 μm and then being subjected to a twin-jet polishing method with a mixed solution of C_2H_5OH (90 vol. %) and $HClO_4$ (10 vol. %).

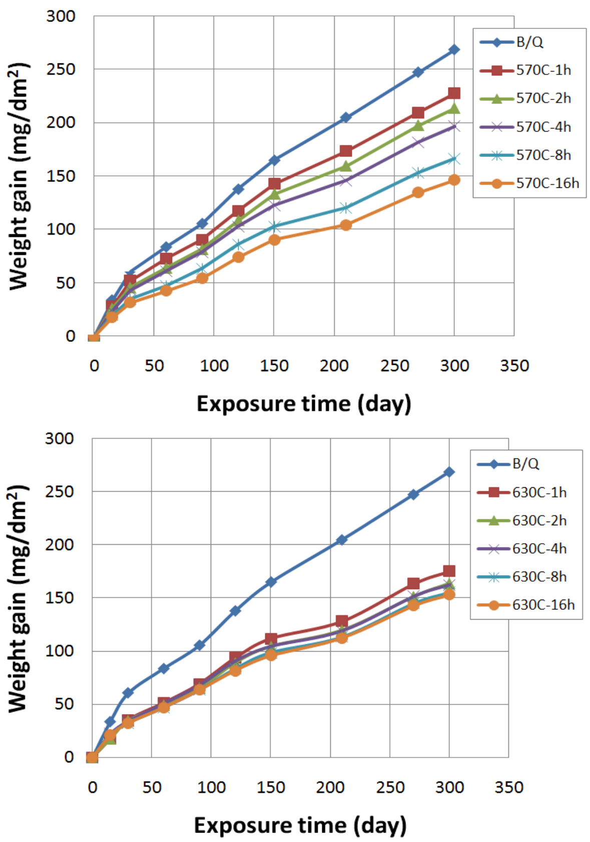

Fig. 2 Corrosion behavior of HANA-4 alloy annealed at 570℃ (upper) and 630℃ (lower) after β-quenching

Fig. 2 shows the corrosion weight gain of HANA-4 alloy as a function of exposure time up to 300 days. The weight gain of the annealed HANA-4 alloy after beta-quenching was increased with increasing the exposure time. The weight gain of all annealed specimens in both 570℃ and 630℃ was lower than that of water quenched specimens. So, it is known that the corrosion resistance of the HANA-4 alloy is increased by applying the annealing after the water quenching. After the annealing at 570℃, the corrosion weight gain was gradually decreased with increasing the annealing time. However, the corrosion weight gain was considerably decreased in all the annealed specimens after the annealing at 600℃, and the weight gain was not changed with the difference aging times in this temperature.

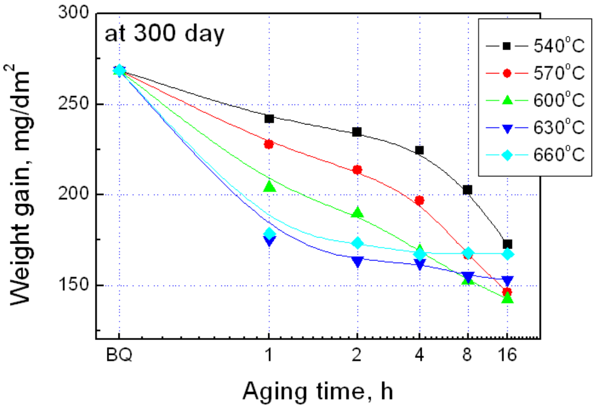

Fig. 3 Comparison of corrosion weight gain with the annealing time and temperatures of HANA-4 alloy

Fig. 3 shows the comparison of corrosion weight gain after 300 day test. The value of the weight gain for the annealed specimens was selected from all the annealing condition both time and temperature. The corrosion resistance of all annealed specimens was increased up to about 50~60% when compared to the water quenched specimens. Since, the lowest weight gain was shown when the specimens were annealed at 570 ℃ and 600 ℃, the optimum temperature for the annealing of HANA-4 alloy was ranged from 570 to 600 ℃.

The TEM microstructure analysis for the annealed specimens is shown in Fig. 4. The second phases were precipitated in the annealed specimens in both 570 and 640℃. But the size and type of the second phase were shown differently with the annealing temperature and time. The size of the second phases in the matrix was smaller in the 570℃ annealed specimen than in the 640℃ annealed specimen. From the analysis of the microstructural characteristics, $Zr(NbFeCr)_2$ phase was formed in the matrix annealed at 570℃, whereas, two types of β-Zr and $Zr(NbFeCr)_2$ phases were formed in the matrix annealed at 630℃. So, it is known that the second phase characteristics such as type, size, and distribution could be changed by the annealing time and temperature. Therefore, the second phase characteristics have considerably effected on the corrosion resistance, since the corrosion weight gain of the annealed specimens ranged from 540℃ to 660℃ in temperature and from 1h to 16 h in time was considerably changed. On the other hand, it could be considered that the corrosion resistance with the annealing of HANA-4 alloy was affected by the decrease of the supersaturated element concentrations of Nb, Fe, and Cr in the α matrix.

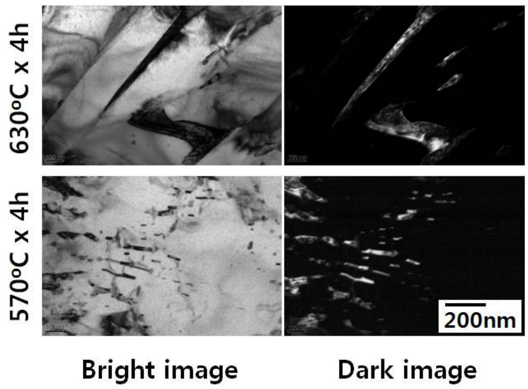

Fig. 4 TEM micrographs of the HANA-4 alloy annealed at 570℃ and 630℃ for 4h

3. Conclusions

The corrosion resistance of the HANA-4 alloy was increased by applying the aging process after the beta-quenching. And the optimum temperature for the annealing of that alloy was ranged from 570 to 600 ℃.

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