Effect of Manufacturing Process on Corrosion Characteristics of HANA-alloy

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1. Introduction

The corrosion resistance of fuel claddings has been considered to be one of the key properties to control the performance and the safety of nuclear fuel. Zr-based alloy such as Zircaloy-4 has been used as fuel cladding materials for the last few decades. Since, the corrosion of fuel claddings is the most critical issues in high burn-up operating condition in PWRs, the development of the advanced Zr-based fuel cladding with an improved corrosion resistance was demanded.

The HANA-alloy designed in KAERI was one of the newly developed materials having an improved corrosion resistance. It was reported that the corrosion properties of Zr-based alloys were very sensitive to their microstructure such as the texture, dislocations and precipitate characteristics [1-3]. The microstructural characteristics of Zr-based alloy were determined by the performed manufacturing process. Therefore, to obtain the good corrosion resistance, the Zr-based alloy as fuel cladding was applied to the optimized manufacturing process. The purpose of this investigation is to get the optimized manufacturing process of HANA-alloy.

2. Experimental procedure

The experimental HANA-alloy was manufactured by the sequence of the vacuum arc re-melting of 4 times to promote the homogeneity, β-quenching at 1050℃, and hot and cold rolling. To study the manufacturing effect such as annealing temperature and hot and cold rolling effect, the β-quenched HANA-alloy was hot-rolled after pre-heating at 580 or 610℃ for 30 min and than applied the cold rolling steps of different times. The cold-rolled samples were intermediate-annealed at 580 or 610℃ for 3 hours and than some samples were final-annealed at 580 or 650℃ for 3 hours. Therefore, the sheet type HANA-alloy having different 8 conditions was manufactured shown as fig. 1.

The microstructure with annealing temperature and rolling sequence was observed using optical microscope with polarized light. The precipitate characteristics were analyzed using transmission electron microscope equipped with energy dispersive spectra. Specimens for TEM observation were prepared by twin-jet polishing with a solution of C_2H_5OH (90 vol.%) and $HClO_3$ (10 vol. %) after mechanical thinning to about 70μm.

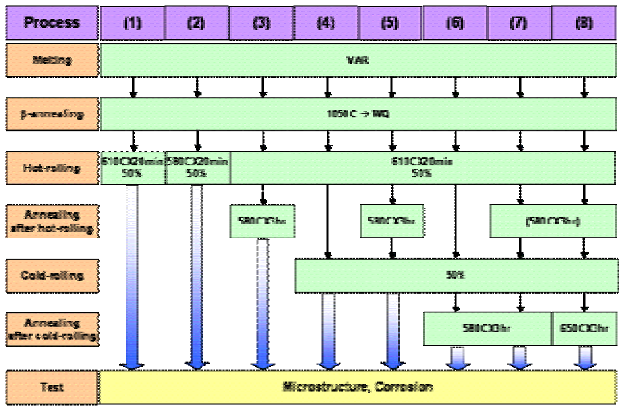

Fig. 1 Manufacturing process of HANA-alloy

The corrosion test was performed in a static autoclave of 360℃ pure water under saturated pressure of 18.9 MPa. Corrosion testing specimens of 15mm x 25mm x 1mm in size were cut from the prepared sheets and mechanically ground up to 1200 grit SiC paper. Also, the ground specimens for the corrosion test were pickled in a 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 of the corroded samples after suspending the corrosion test at a periodic term.

3. Results and discussion

The microstructure observation was performed on the characteristics of the homogenous grain and precipitates, since it is well known that corrosion properties of Zr alloys is highly depended on the microstructural characteristics.

Fig. 2 shows the optical microstructures of HANA-alloy with different manufacturing process. Their microstructural characteristics were largely changed by manufacturing process shown as fig. 1. The martensite structure (prior β phase) was formed at water quenched samples from β region of 1050℃. The massive large grains were observed in the case of process (1), (2) and (3), and the elongated grains were observed in the case of process (4) and (5). The small recrystallized grains were observed in case of process (6), (7) and (8). From the these results of microstructure observation, homogenous microstructure such as small recrystallized grains was formed by the process of annealing after cold rolling.

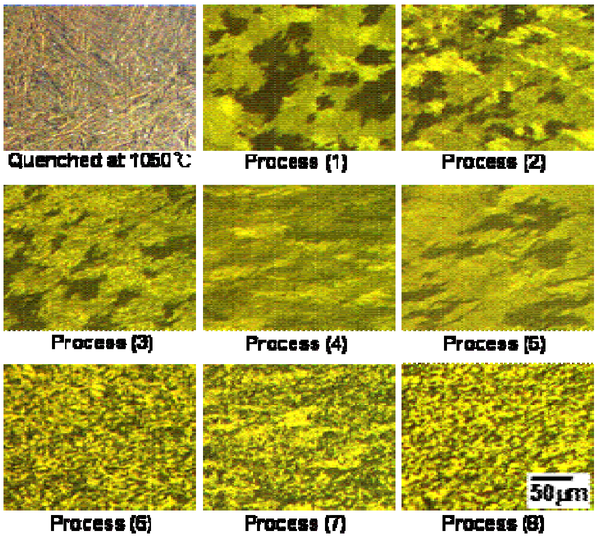

Fig. 2 Optical microstructures of HANA-alloy with different manufacturing process

From the result of precipitate analysis using TEM, the alloying elements of HANA alloy were homogeneously supersaturated in the martensitic matrix, because the precipitates were not observed in the martensite structure formed by water quenching. The precipitate in the matrix was observed after hot rolling process of 590 or 610℃. The precipitate type such as β-Zr and β-Nb phase was differently showed by manufacturing process. When samples were annealed at 580℃ during the process, the β-Nb phase was formed.

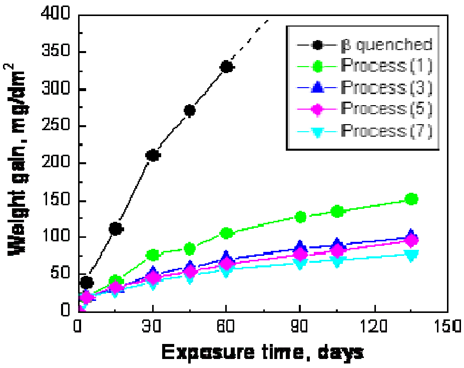

Fig. 3 Corrosion behaviors of HANA-alloy with the different manufacturing process

Fig. 3 shows the corrosion behavior of HANA-alloy manufactured by different process in an autoclave of 360℃ pure water up to 135 days. The corrosion resistance of HANA-alloy was largely affected by the manufacturing process conditions. The corrosion resistance of that alloy was affected by the pre-heating temperature in hot-rolling and the intermediate-annealing temperature during cold rolling. When the specimens annealed at low temperature of 580℃, the weight gain was decreased. Also, the weight gain was decreased by the cold working after hot rolling and annealing procedure.

From the microstructure and corrosion studies, the corrosion rate was correlated with the microstructural characteristics. The lower corrosion rate of samples was showed when the samples matrix was consisted of the small recrystallized grains, which were formed by the cold rolling after hot rolling and annealing. The precipitation of β-Nb phase in the matrix was also increasing the corrosion resistance. It seems that the corrosion resistance of HANA-alloy would be related to the microstructural characteristics which were determined by manufacturing process conditions.

4. Conclusion

The specimens of HANA-alloy that was made by various manufacturing process conditions were investigated in order to get the optimized manufacturing process. The microstructure of HANA-alloy was largely changed by the manufacturing process conditions and the corrosion resistance of that was affected by the manufacturing process conditions. From the study of microstructure characteristics, the good corrosion resistance of HANA-alloy was obtained when the specimens were manufactured by the cold working after hot rolling and annealing which was performed at low temperature of 580℃.

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