Behavior of a Recrystallization in Zirconium-based Alloys

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1. Introduction

As high burn-up fuel cladding materials HANA(High performance Alloy for Nuclear Application) has been developed by KAERI(Korea Atomic Energy Research Institute) recently. A series of out-of-pile and in-pile tests demonstrated that the HANA cladding tubes are superior to commercial Zr-base alloys in the mechanical properties as well as the corrosion properties [1,2]. Along with the research for a fuel cladding application using HANA alloys, their application to a spacer grid for a fuel assembly is also being investigated.

For robust mechanical properties of a spacer grid, an appropriate microstructure is needed. Especially, a recrystallized microstructure is desirable for proper mechanical properties, a corrosion resistance, a creep resistance, and an irradiation growth [3]. In addition, an intermediate recrystallization is utilized to restore a ductility after a cold deformation process during a manufacturing. Therefore, it is very important to establish the recrystallization behavior of the developed alloys.

In this paper, the recrystallization phenomena of Zr-based alloys, i.e. HANA strips, will be presented, and the obtained kinetic parameters for the recrystallization to enable us to control the microstructures of the alloys.

2. Methods and Results

The chemical composition of the strips for this study was Zr-1.47Nb-0.38Sn-0.20Fe-0.11Cr (HANA-4) and Zr-1.11Nb-0.08Cu (HANA-6) with some impurities such as Si, H, N less than 100 ppm. The two strips were manufactured in accordance with applicable requirements. For homogenizing of the alloying elements, several meltings and betatizing at 1050°C for 15 min were introduced. The prepared ingots were hot-rolled and cold-rolled to have 0.66 thicknesses. The heat-treatments subsequent to every cold-rolling were conducted below a monotectoid temperature of about 600°C.

Final cold-rolled strips were heat-treated at various temperatures and times. The samples were heat-treated at 350, 400, 450, 475, 500, 525, 550, 600, 650, 700°C for 1 h for the isothermal heat-treatment, and 500, 550, 600°C for 20, 150, 400, 1000 min for the isochronal heat-treatment, respectively. For the heat-treatment, oxidation of the samples was prevented by using a quartz tubing with a vacuum. The heat-treated samples were mounted and then manually polished up to 2000 grit SiC paper. The polished sections were etched by a scrubbing with a $10HF–45HNO_3–45H_2O$ (vol%) solution. Optical micrographs were observed under a polarization filter. The hardness of the samples was measured by a Knoop indentation at a 1 kg load. For the determination of the hardness, an average value was calculated excepting the maximum and minimum values among the 12 times measurements for each sample.

2.1 Microstructures

Figure 1 shows the optical micrographs for the HANA-4 and HANA-6 strips with respect to the different heat-treatment temperatures. After 1 h of a heat-treatment a recrystallized microstructure begins to appear at 600°C for the HANA-4 strip, and the 550°C for the HANA-6 strip. As shown in Fig.1, it is considered that the higher concentration of the solute atoms in the Zr-based alloys induces a higher transformation temperature for the recrystallization.

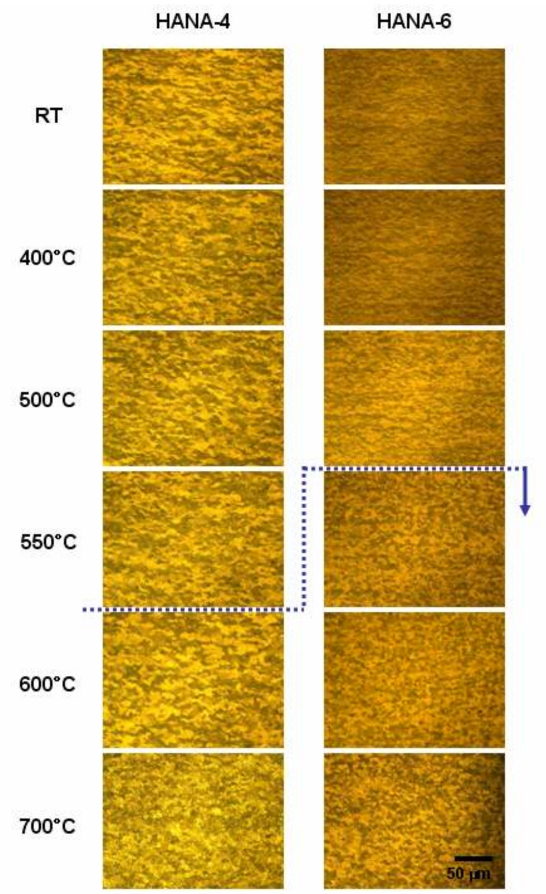

Fig. 1. Optical microstructures for HANA-4 and HANA-6 strips heat-treated various temperatures for 1 h.

2.2 Hardness

Figure 2 shows the hardness of the HANA-4 and HANA-6 strips for various annealing temperatures. In accordance with the micrographs (Fig. 1), the softening of the HANA-4 and HANA-6 strips was completed at 550°C and 600°C, respectively. The hardness of the HANA-4 strip is higher than that of HANA-6 regardless of the recrystallization, which is due to the high solute concentration of the added atoms.

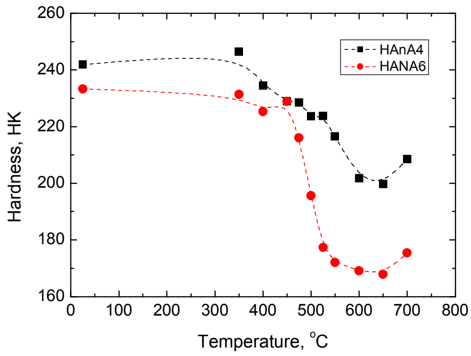

Fig. 2. Knoop Hardness number of HANA-4 and HANA-6 strips heat-treated various temperatures for 1 h.

2.3 Recrystallization

The volume fraction of the recrystallized grain with respect to the annealing time can be described well by using the Johnson–Mehl–Avrami–Kolmogorov (JMAK) equation [4]. The JMAK equation is expressed as

$$f = 1 - \exp(-kt^n) \qquad (1)$$

where f is a recrystallized volume fraction, k a temperature dependent constant, and n an exponent depending on the geometry and temperature, respectively. Since the hardness is inversely correlated with the volume fraction of the recrystallized grains, i.e.

$$H = (1-f)\cdot(H_{\max} - H_{rex}) + H_{rex} \qquad (2)$$

where $H_{\max}$ is the maximum available hardness for a sample (prior to recrystallization), and H_{rex} the hardness for a recrystallized sample. The values of k and n can be determined experimentally by (i) converting the hardness into a recrystallized volume fraction using Eq.(2) and (ii) conducting a regression of Eq.(1) with the introduction of a logarithm to have the form of

$$\log(-\log(1-f)) = \log k + n \log t \qquad (3)$$

Table 1 shows the obtained k and n values for the HANA-4 and HANA-6 strips. The k and n values are varied depending on the temperature; however, a specific relationship between the kinetic values and temperature can not be found. According to a classical treatment, the n value should be an integer, esp. 3 for a site-saturated process and 4 for a constant rate of a nucleation process. However, the n values obtained from this study are below 1. This signifies that a random nucleation didn't occur during the recrystallization process.

Table I: k and n values for recrystallization

ID	T(°C)	k	n
HANA-4	500	0.00081	0.77
	550	0.017	0.46
	600	0.032	0.51
HANA-6	500	0.012	0.52
	550	0.046	0.53
	600	0.046	0.58

For a certain fraction of recrystallization, f_X, the time required for that, t_X, can be deduced from Eq. (1):

$$t_X = \left[\frac{-\ln(1-f_X)}{k}\right]^{1/n} . \qquad (4)$$

Once the activation energy is obtained, the degree of recrystallization can be calculated and anticipated using Eq.(4).

3. Conclusions

The recrystallization behaviors of Zr-based HANA strips were investigated through isothermal and isochronal heat-treatments. The HANA-4 strip recrystallized at a higher temperature (>600°C) than the HANA-6 strip did (>550°C). Moreover, the obtained kinetic parameters in the JMAK equation, i.e. k and n, would make the prediction for the recrystallization of the strips possible.

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REFERENCES

[1] Y.H. Jeong, S.-Y. Park, M.-H. Lee, B.-K. Choi, J.-H. Baek, J.-Y. Park, J.-H. Kim, and H.-G. Kim, Out-of-pile and In-pile Performance of Advanced Zirconium Alloys (HANA) for High Burn-up Fuel, J. Nucl. Sci. Tech., 43[9], 977-983, (2006).
[2] K.W. Song, Y.H. Jeong, K.S. Kim, J.G. Bang, T.H. Chun, H.K. Kim, K.N. Song, High burnup fuel technology in Korea, Nucl. Eng. Tech., 40(1), 21-36, (2008).
[3] G.S. Lee, Nuclear Reactor Materials, Hyoilbooks, p.158 (2006). ISBN-89-8489-190-8
[4] F.J. Humphreys, M. Hatherly, Recrystallization and related annealing phenomena (2nd ed.), Elsevier Ltd. (2004).