Application of Image Quality Measured by EBSD to Avrami Model to Quantify the Degree of Recrystallization in Zirconium-Niobium Cladding

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1. Introduction

In general, metal and non-ferrous metal parts used in industry are subjected to cold working and then to final heat treatment for proper performance [1]. Likewise, after zirconium cladding tube is cold-pilgered, used in nuclear power plants, the final heat treatment is performed to obtain excellent performance in reactor. In this process, the cold-worked microstructure in the cladding is replaced with recrystallized structure depending on the temperature and time of the final heat treatment [2,3,4]. This is called recrystallization and is used as specification featuring the annealed product for predicting the performance of a cladding [2]. However, in the steel industry as well as the non-ferrous metal industry, it is not yet possible to quantify the recrystallization degree or accurately predict or use the predictive model.

First, the effort to accurately predict the degree of recrystallization of zirconium using the Avrami model equation has often been studied by previous studies [5, 6, 7,8]. However, the predicted values using the degree of recrystallization calculated using mechanical strength are somewhat inaccurate because of the interference characteristics of recovery accompanying process before recrystallization [5-10]. Therefore, when Avrami model equation is used, accurate measurement of the recrystallization degree itself should be basically preceded.

With the development of analytical methods, EBSD can be used to quantify microstructure that has been regarded as qualitative results [11-16]. In recent years, various calculations can be performed using numerical results measured from microstructures. In particular, methods that can replace the method of measuring the degree of recrystallization (which may vary depending on the measurer) using the area fraction of the microstructure have been studied. Commonly used methods using data measured with EBSD are GOS (grain orientation spread), KAM (kernel average misorientation), and GAM (grain average misorientation) which use the difference in grain orientation of the measurement site [17,18]. However, such a method in zirconium with strong texture tends to result in erroneous consequence [18]. However, this problem can be overcome by using the image quality (IQ) which is the degree of sharpness of the Kikuchi line at the measurement site [7,9]. The clear Kikuchi pattern means that the crystal structure of the region has a high crystallinity, that is, that new crystalline grains with high crystallinity are newly formed in the cold worked region in microstructure. T.S. Jung's study [9] showed that reliable recrystallization can be measured and calculated.

Therefore, in this study, it is described that the recrystallization degree obtained from the calculation formula developed through T.S. Jung's study [9] can be used to obtain the predicted value of recrystallization better than the result obtained from the mechanical strength.

2. Experimental procedures

The specimen used for measuring and calculating the degree of recrystallization was a zirconium - niobium cladding tube, which was final cold-pilgered (or deformed), and heat treatment was performed for 8 hours at 440 to 580°C. The uniaxial tensile test was performed to calculate the recrystallization using the mechanical properties. The recrystallization degree value calculation was performed using the following formula [6,7,11].

$$\text{Recrystallization degree} = \frac{\sigma_{y,Max} - \sigma_{y,P}}{\sigma_{y,Max} - \sigma_{y,Min}} \times 100\ \%,$$

Where $\sigma_{y,Max}$ is the yield strength of the as-cold pilgered specimen, $\sigma_{y,Min}$ is the yield strength of the fully recrystallized specimen, and $\sigma_{y,P}$ is the yield strength of the partially recrystallized specimen at the temperature of interest.

In addition, the vertical surface in the longitudinal direction was polished by mechanical and electrolytic method for EBSD scans which were conducted with FEI™ Quanta 3D-FEG (field emission gun) SEM (scanning electron microscopy) with TSL OIM™ analysis 7 software to measure the IQ value. The accelerating voltage of beam was 20 kV and the probe current was 4 nA as well as the camera exposure time was 19.62mn. The measured area was 100 x 100 μm^2 and step a size was set to 0.25 μm. Distribution histograms of IQ value of each specimen annealed at various temperatures were obtained. In addition, TEM images were taken in the radial direction to observe the directly recrystallized grains. To make the TEM specimen, the cladding tube was chemically etched to reduce the thickness to 30 μm and electropolished using twin-Jet polishing.

Finally, the recrystallization value obtained using the IQ values were applied to the Avrami model equation as input to confirm that the results with improved accuracy can be obtained.

3. Results and discussion

Figure 1 shows the microstructure images observed by TEM in the radial direction. It can be seen that new grains appear from 480°C, which indicates that the recrystallization temperature (T_{Rex}) of the zirconium-niobium cladding tube is around 480°C.

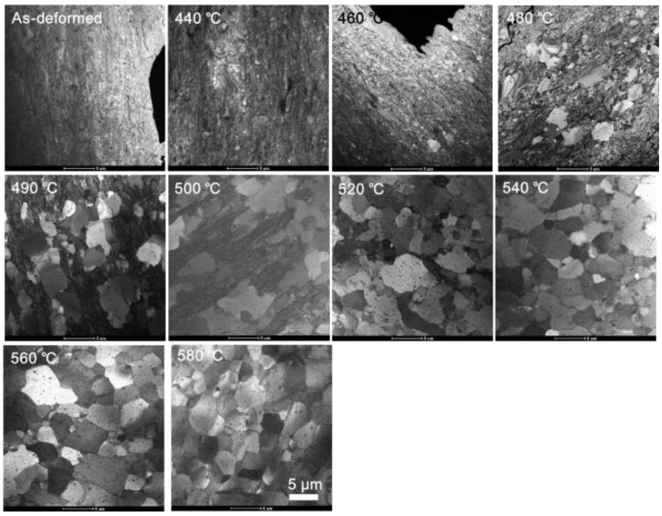

Fig. 1. High-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) images of a zirconium–niobium cladding specimen in the as-deformed state and specimens annealed for 8 h at various temperatures from 440°C to 580°C (fully recrystallized) [9].

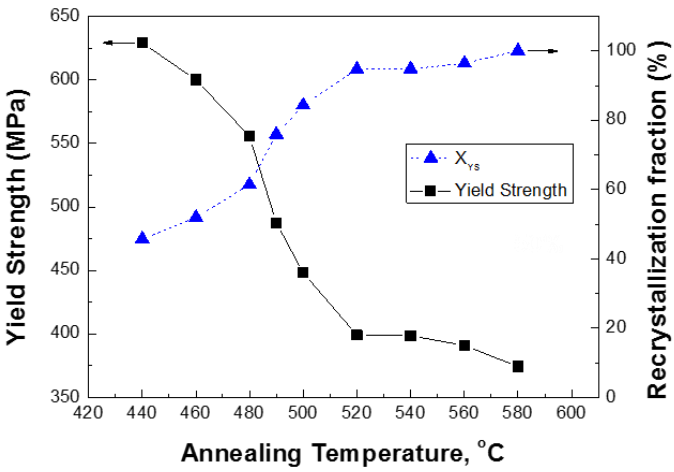

Fig. 2. The uniaxial tensile test results (yield strength and degree of recrystallization calculated using yield-strength drop) of cold-pilgered zirconium–niobium cladding as a function of annealing temperature at each time for 8 h [9].

The yield strength and the degree of recrystallization value obtained by the previous formula are shown in Fig. 2. Unusually, the yield strength decrease of 40% or more (The degree of recrystallization value is 40% or more) was observed at 440°C, unlike the case shown in Fig. 1. As mentioned in the introduction, it can be confirmed once again that the yield strength due to recrystallization cannot be accurately extracted from the mechanical properties, since recovery affects the yield strength reduction.

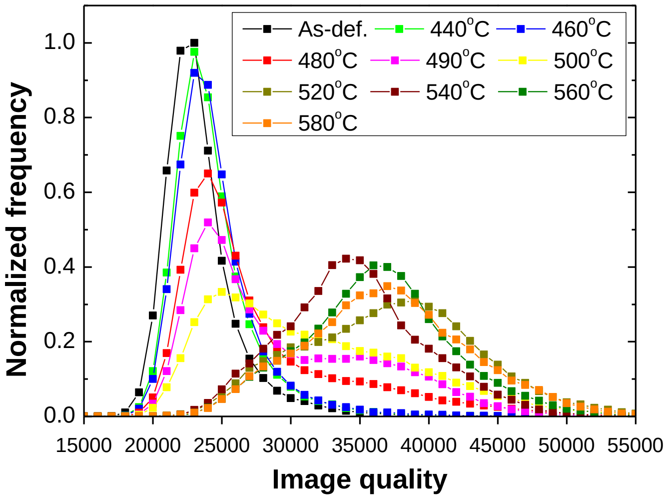

Fig. 3. Distribution of image quality values obtained through EBSD observation of the surface perpendicular to ND-TD of an as-deformed sample and samples annealed for 8 h at various temperatures from 440°C to 580°C (fully recrystallized) [9].

Figure 3 is a histogram showing the IQ values obtained using EBSD as the normalized frequency of all the specimen annealed ranging from 440 to 580oC incorporating as-deformed cladding called 'As-def.'. As the IQ distribution for each specimen is examined, it can be seen that the maximum frequency increases with increasing temperature from As-deformed. As the new grain appears, the sharpness of the Kikuchi line increases and eventually the IQ increases. In addition, it changes from unimodal shape to bimodal shape, and then the maximum frequency is unified again in the form of unimodal, which shows the change in the size distribution of newly formed grains. In other words, when the grain size is a normal distribution, the IQ distribution also forms a normal distribution. When the IQ distribution has a bimodal shape, the grain size distribution also has a bimodal shape. The reason why the grain size distribution is changed according to the heat treatment temperature seems to be that the recrystallization behavior is divided into heterogeneous (bimodal) and homogeneous (unimodal) form depending on the heat treatment temperature. This fact can be confirmed through the results of the previous studies [7,18]. Additionally, Fig. 3 shows the recrystallization temperature at which recrystallized grains grow rapidly. This is due to the fact that there is almost no change in the IQ distribution until 460°C, but the frequency starts to decrease rapidly from 480°C to the high IQ side. This is because the new grains have appeared.

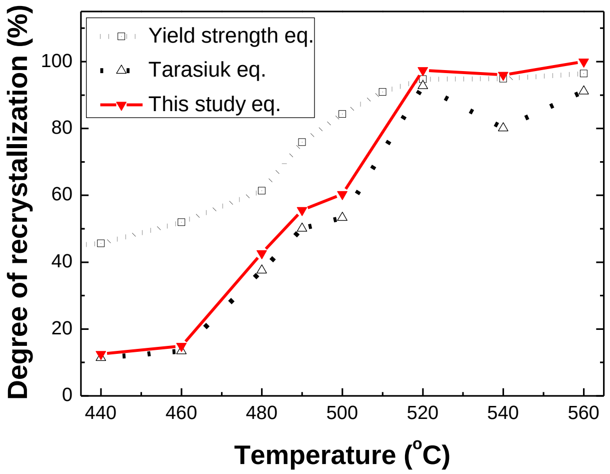

Fig. 4. Calculated degree of recrystallization using equations of yield strength, the method of Tarasiuk et al. [7], and the method proposed in the present study; samples were annealed at various temperatures for 8 h [9].

When the IQ frequency results are substituted into the equation developed in the previous study [9], the recrystallization value as shown in Fig. 4 can be obtained. The result is more accurate than the calculated value using the mechanical strength measurement value and the formula calculated by the existing researcher [7]. In the case of mechanical strength measurement, as mentioned above, since the calculated recrystallization degree is more than 40% at 440$_o$C, it is inferior to the TEM micrographs as shown in Fig. 1, resulting in an inaccurate result. In addition, when Tarasiuk's formula [7] is used, the degree of recrystallization decreases rapidly at 540°C. This seems to be due to the fact that the material studied by Tarasiuk is applicable only to materials with a cubic system structure.

T.S. Jung's equation [9] reflects the properties of the microstructures of the hexagonal system in the calculation of the IQ value of the zirconium alloy. As a result, a method for extracting the IQ value from the newly formed grains in the microstructure is presented. As a result, it was possible to derive the degree of recrystallization which corresponds well to the TEM micrographs in Fig. 1.

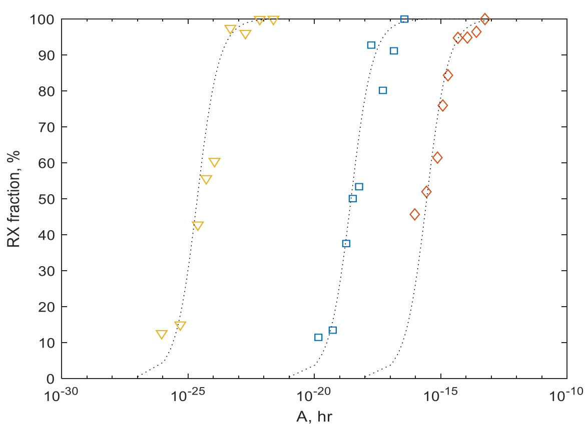

Fig. 5. The results obtained by adding various degree of recrystallization value to Avrami model equation.

Finally, the recrystallization degree obtained by the T.S. Jung's equation was substituted into the Avrami model equation, and the predicted value of the recrystallization degree was obtained as shown in Fig 5.

In the case of using the mechanical strength among the numerical values assigned to the Avrami model formula in the existing papers, it is necessary to obtain the inaccurate recrystallization value since the recrystallization incorporating the recovery is calculated with the value. The calculation result using Tarasiuk's equation shows a large error already at 540oC. The results of T.S. Jung show that the experimental and computational model results are similar not only in the vicinity of high recrystallization degree but also in the region where the recrystallization curve rapidly changes. However, the difference between 440°C and 500°C is 8% and 20%, respectively.

4. Conclusions

In order to predict the change of recrystallization due to the final annealing of zirconium - niobium alloy cladding, we used the recrystallization value calculated by using the IQ value measured by EBSD as a method to improve the accuracy of the result using Avrami model equation.

By using the IQ value, the degree of recrystallization was calculated and the recovery factor caused by the mechanical strength measurement could be excluded, as a result, the accuracy could be improved. In addition, the results obtained directly from the microstructure were quantified, and the results were objectified.

IQ values are measured according to the microstructure of the zirconium-niobium cladding tube, and IQ values diffracted from only the recrystallized microstructure, can be obtained through T.S. Jung's equation, as a result, the degree of recrystallization with high accuracy were calculated.

The degree of recrystallization by T.S Jung's equation was applied to the existing Avrami model equation and the recrystallization predicted value was obtained with higher accuracy than the conventional method.

Acknowledgement

This work was financially supported by the Nuclear R&D Program (No. 20151510101790) of the Korea Institute of Energy Technology Evaluation and Planning (KETEP) and (L17S083000) of Korea Hydro & Nuclear Power (KHNP).

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