Effect of Thermomechanical Parameters on Grain Growth and Recrystallization during Grain Boundary Engineering of Austenitic Stainless Steel

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Abstract. Grain boundary engineering has been attracting attention as an effective method to prevent intergranular corrosion of austenitic stainless steel and Ni based alloys. It has been considered that grain growth and recrystallization play important role in evolution of grain boundary character distribution during thermomechanical process. However, systematic researches to examine the effect of thermomechanical process parameters on grain growth and recrystallization have not been performed. In this study, grain boundary character distribution of 304 austenitic stainless steel after thermomechanical process with various parameters was analysed by electron backscatter diffraction (EBSD). Grain boundary character distribution was mainly affected by the reduction ratio of cold rolling. Abnormal grain growth was observed in the specimen with small reduction ratio (3%). Length ratio of coincident site lattice (CSL) boundaries was drastically increased to 86% in the 3% cold rolled and annealed specimens from 67% in the base material. On the other hand, normal grain growth was observed in thermomechanical processed specimens with slightly higher reduction of cold rolling (5%). In these specimens length ratio of CSL boundaries did not exceed 80%, which is required to disconnect the random boundary networks and to improve intergranular corrosion resistance effectively. Detailed analysis of grain boundary character distribution has shown that disconnection of random boundary networks was achieved by formation of annealing twins during abnormal grain growth.

1. Introduction

Austenitic stainless steels have been widely used in many industrial applications because they have good mechanical properties and high resistance to general corrosion. However, weld decay, which is severe intergranular corrosion in weld heat affected zone, often occurs at grain boundary during the welding process. Intergranular corrosion is caused by the formation of chromium carbides at grain boundaries.

Grain boundary engineering [1, 2] has been attracting attention as an effective method to prevent grain boundary degradation such as intergranular corrosion. Grain boundaries can be categorized into
coincidence site lattice (CSL) boundaries and the other random boundaries. It has been reported that the CSL boundaries have higher intergranular corrosion resistance than random boundaries because precipitation of chromium carbide was prevented in the CSL boundaries [3, 4]. Utilizing these properties, resistance to grain boundary degradation can be improved by an increase in the frequency of CSL boundaries and disconnection of random boundary networks. Recent researches has reported an increase in the frequency of CSL boundaries and an improvement of intergranular corrosion resistance of 304 austenitic stainless steel by application of grain boundary engineering [5-9]. Suppression of grain boundary degradations such as grain boundary cracking, segregation, and intergranular stress corrosion cracking has been reported in the previous study [10].

The thermomechanical process is often used to optimize the grain boundary character distribution (GBCD). Considering the practical use of the thermomechanical process for GBE, the optimization mechanism of GBCD needs to be revealed to improve the energy efficiency and reliability as a microstructural control process. However, the mechanism of the evolution of grain boundary character distribution during the thermomechanical process is not fully understood. In this study, systematic examination of thermomechanical process parameters and measurement of intergranular corrosion resistance were conducted to reveal the effect of thermomechanical parameters on the evolution of GBCD and intergranular corrosion resistance.

2. Experiments

2.1. Specimen preparation

The material used in this study was 304 austenitic stainless steel with the chemical composition of 18.36 Cr, 8.15 Ni, 0.58 Si, 1.27 Mn, 0.040 C, 0.032 P, 0.001 S (wt%). The solution heat treatment was conducted in electric furnace at air atmosphere in 1323 K for 0.5 h and subsequently quenched in cold water. The solution heat treated material will be denoted as the base material in this study. The thermomechanical process for grain boundary engineering can be divided into strain annealing and strain recrystallization based on their parameters, such as reduction ratio of cold rolling and annealing time [11, 12]. In this study, strain annealing process was conducted because the drastic increase of the frequency of CSL boundaries of 304 and 316 austenitic steel was reported in the previous study [5, 6]. Specimens were deformed by cold rolling, resulting in the reduction of the thickness of 3, 5 and 10%. The cold rolled specimens were cut into small chips, 5W × 4L × 2T mm3 in size. The annealing process was performed in an electric furnace with an annealing temperature of 1220, 1280, 1340 K and specimens were subsequently quenched in cold water.

Thermomechanical processed specimens were mechanically polished by the water-abrasive paper and subsequently polished with 1 µm Al2O3 particles. Electropolishing was conducted in 10% perchloric acid ethanol at 273 K at a potential of 25 V to obtain the suitable surface conditions for EBSD observation.

2.2. EBSD observation

The GBCD analysis was conducted with a field emission scanning electron microscopy HITACHI S-4300SE equipped with TSL OIM system. EBSD observation with a step size 5 µm was conducted in the 2 mm x 2 mm observed area, which was perpendicular to the rolling direction. The cleanup procedure in the OIM analysis software was performed using the neighbor confidence index (CI) correlation method with a minimum CI value of 0.1. Grain definition with a minimum grain size of 5 analysis pixels and a minimum misorientation angle of 15 degree was set for the grain dilation clean up method and other grain-based analysis such as grain orientation spread analysis. Brandon’s criteria [13] was adopted to define the maximum tolerance angle of CSL boundaries. The relationship between grain boundary character and intergranular corrosion resistance have been examined in recent studies. [5, 14, 15] Σ3 CSL boundary has been widely recognized as excellent corrosion resistant boundary while other Σ CSL boundaries do not always show the high corrosion resistance. On the other hand, other Σ CSL boundaries sometimes prevent propagation of intergranular
corrosion, because the previous study showed that non-corroded or non-sensitized $\Sigma 9, 13b, 17a, 29a$ boundaries disconnected corroded random boundaries [5, 15]. In this study, the grain boundaries in EBSD data were roughly categorized into CSL boundaries with low-$\Sigma$ value ($\Sigma \leq 29$) and the other random boundaries, because this study focused on the relationship between thermomechanical process parameters and the general tendency of the grain boundary character distribution. The frequency of CSL boundaries was measured as a length ratio of the CSL boundaries to a total length of all low angle and high angle boundaries.

3. Results and discussions

3.1. Effect of thermomechanical parameters on the grain boundary character distributions

The GBCD maps of the thermomechanical processed specimens with 3, 5, 10% of cold rolling and annealing in 1220, 1280, 1340 K are shown in figure 1. Random boundary and CSL boundary were depicted as black and green lines in the GBCD maps. The GBCDs of the thermomechanical processed specimen was mainly affected by the reduction ratio in cold rolling. A significant increase of CSL boundaries during grain growth and annealing twinning was observed in the 3% cold rolled and annealed specimens. The CSL boundaries that replaced and disconnected random boundaries were observed. The random boundary networks were effectively disconnected in the specimen.

|       | 3%       | 5%       | 10%      |
|-------|----------|----------|----------|
| 1340 K|          |          |          |
| 1280 K|          |          |          |
| 1220 K|          |          |          |

Figure 1 Grain boundary character distribution map of the thermomechanical processed materials.
The 5% cold rolled and annealed specimen also showed an increase of CSL boundaries, but most of CSL boundaries were distributed inside of random boundary grains and significant disconnection of random boundary networks was not observed. The GBCDs of the specimens with 10% cold rolling did not change significantly from that of the base material. Although the grain size of the specimens with 10% cold rolling was slightly increased in higher annealing temperature condition, a significant effect of the annealing temperature on the GBCD was not observed in figure 1. Annealing time for GBCD change in high annealing temperature conditions was shorter than that in low annealing temperature conditions. These results showed that the processing time for the thermomechanical process can be shortened by increasing annealing temperature in this system.

Figure 2 shows the frequency of CSL boundaries of thermomechanical processed specimens with different annealing times. The frequency of the CSL boundaries in the base material was 51%. The frequency of the CSL boundaries of the 3% cold rolled specimen increased until 16 h of annealing and then stabilized above 80%. The frequency of CSL boundaries of the 5% cold rolled specimen increased within 2 h and the highest frequency was 78%, which was lower than that in the 3% cold rolled specimen. The 10% cold rolled specimen did not show the increase in the frequency of CSL boundaries. It has been reported that the frequency of CSL boundaries higher than 80% is effective to disconnect three-dimensional random boundary networks [16] and to improve intergranular corrosion resistance [6]. Therefore, the improvement of intergranular corrosion resistance was expected in the thermomechanical processed specimens with 3% cold rolling.

In order to discuss the reason why the GBCDs of the thermomechanical processed specimen were mainly affected by reduction ratio in cold rolling, grain orientation spread (GOS) maps of the
specimens are shown in figure 3. GOS represents the average deviation between the orientation of each analysis pixel in a grain and the average orientation of the grain. Grains with higher GOS value, those are depicted as darker red in figure 3, are considered to have larger strain. The grains with lighter red were considered as growing grains by strain induced grain growth. Large growing grains with lower GOS value were observed in the 3% cold rolled specimen, as shown in right side of the observed area in figure 3 (a). Previous research has shown the frequency of CSL boundaries increased through the annealing twinning during abnormal grain growth [17]. It is considered from this result that microstructural change for optimization of the GBCD could be successfully obtained in the 3% cold rolled and annealed specimen. In the 5% cold rolled and annealed specimen, size of the growing grains was relatively smaller than that in the 3% cold rolled specimen. It is estimated that impingement of growing grains should occur more frequently in this condition. Therefore, abnormal grain growth could not occur efficiently in this condition because impinged grains stopped growing. This might be the reason why the frequency of CSL boundaries could not be increased adequately in the 5% cold rolled and annealed specimen in figure 3. In the 10% cold rolled specimen, homogeneous distribution of small growing grains with low strain was observed. This result shows recrystallization occurred in this condition. Optimized GBCD could not be obtained in this condition because recrystallized grains were mainly surrounded by random boundaries as shown in figure 1.

4. Conclusion
The GBCDs of thermomechanical processed specimens were mainly affected by the reduction ratio of cold rolling. The 3% cold rolled and annealed specimen showed the abnormal grain growth and drastic increase of CSL boundaries, while the 5% cold rolled and annealed specimen showed normal grain growth. It is suggested that increase in length ratio of CSL boundaries was achieved by the abnormal grain growth and frequently formation of annealing twins.

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