

Investigation of Microstructural Changes of Type-II Boundary by Thermal Aging

Seung Chang Yoo, Kyoung Joon Choi and Ji Hyun Kim*

School of Mechanical and Nuclear Engineering, Ulsan National Institute of Science and Technology (UNIST), 100 Banyeon-ri, Eonyang-eup, Ulju-gun, Ulsan 689-798, Republic of Korea

*Corresponding author: kimjh@unist.ac.kr

1. Introduction

One important issue deciding the life of nuclear power plants is the susceptibility of structural materials to stress corrosion cracking (SCC). SCC is caused by two key factors: 1) stress from high pressure and radiation, and 2) corrosion from the corrosive environment of nuclear power plants. Dissimilar metal weld (DMW) joints are known to be susceptible to SCC because of 1) residual stress induced by the difference of the thermal conductivity and thermal expansion coefficients between the base metal and weld metal, 2) the residual stress caused by the welding process and oxidation, and 3) the complicated microstructure at the weld fusion boundary because of repeated welding heat and compositional gradients [1].

Alloy 152 is frequently used as a filler metal for joining austenitic Ni-based Alloy 690 and low-alloy steel (LAS) A533 Gr. B. This DMW joint is commonly used in nuclear power plants in pressure vessel nozzles or steam generator nozzles. Before DMW joints were performed with Alloy 152 filler metal, Alloy 182 was used as a filler material. However, Alloy 182 is found to be particularly susceptible to SCC after long-term operation in power plants. For this reason, the filler metal was replaced with Alloy 152. To date, there is no experience of SCC in the weld side of DMW joints where Alloy 152 is used as a filler metal in operation power plants. However, it is believed that the current operational experience is not sufficient to conclude that high Cr, Ni alloys are immune to SCC.

Many studies emphasize the importance of the SCC of the weld fusion boundary because of the complex microstructure near the fusion boundary and because of the residual stress induced by oxidation or the welding process. Seifert et al. [2] conducted an investigation on SCC growth behavior in the transition region of an Alloy 182-SA 508 Cl.2 weld joint heat treated under simulated boiling water reactor normal water chemistry (BWR/NWC) conditions. They conclude that the weld metals are more susceptible to SCC growth and that most cracks are blunted by the fusion boundary. However, they also found that some cracking occurs along the fusion boundary, often in an area with high hardness.

Nelson et al. [3] investigated a DMW of Monel 409 stainless steel and American Iron and Steel Institute (AISI) 1080 alloy and found a type-II boundary, which

exists parallel to the fusion boundary in the dilution zone. They conclude that the type-II boundary is a potential path for crack growth. While there are several theories for the mechanisms of the type-II boundary formation, they conclude that the type-II boundary forms from the allotropic δ - γ transformation at the base metal in the elevated austenitic temperature range. Moreover, many other crack growth experiments conclude that the type-II boundary and fusion boundary region of the weld metal are susceptible to SCC [4]. Hou et al. [4, 5] investigate the microstructure and mechanical properties of the DMW of Alloy 182 and low-alloy steel A533 Gr. B. Using the tensile test, they found that type-II boundaries are high angle grain boundaries, which are more susceptible to SCC than low-angle grain boundaries.

As the operation time of nuclear power plants using DMWs of Alloy 152 and A533 Gr. B increases, these DMWs must be evaluated for their resistance to SCC for long-term operations. However, only few studies have investigated the thermal aging effects induced by long-term operations at high temperature. So thermal aging effect by long-term operation, and existence of type-II boundary must be considered to evaluate the susceptibility to SCC of structural materials.

Purpose of this study is to analyze the detailed microstructure of the type-II boundary region in the DMW of Alloy 152 and A533 Gr. B, after applying heat treatment simulating thermal aging effect of a nuclear power plant operation condition to evaluate the susceptibility of this region to SCC. The microstructure of the type-II boundary region in the DMW of Alloy 152 and A533 Gr. B were analyzed with an energy dispersive x-ray spectroscopy attached to a scanning electron microscope (SEM-EDX), electron backscatter diffraction (EBSD), and a nanoindentation test.

2. Experiments

2.1 Materials

In the former research, DMW samples were fabricated by Argonne National Laboratory, joining Alloy 690 and low alloy steel A533 Gr. B with Alloy 152 filler metal by multi-pass shielded metal arc welding [6]. Latter heat treatment was followed. Specimens for experiments were prepared at weld root of mock-up samples

2.2 Thermal aging

The heat treatment was conducted to simulate thermal aging at LWR environment by long term operation at 320°C. The accelerated temperature for thermal aging simulation is decided to 450°C which is the highest temperature not making any excessive carbides or sigma phase which would not be created after long term operated at 320°C. Equation (1) is used for calculating aging time needed to simulation.

$$\frac{t_{aging}}{t_{ref}} = \exp \left[-\frac{Q \left(\frac{1}{T_{ref}} - \frac{1}{T_{aging}} \right)}{R} \right] \quad (1)$$

t_{aging} is aging time needed to simulate thermal aging at nuclear power plant about t_{ref} . T_{aging} is thermal aging temperature and T_{ref} is actual operation temperature of nuclear power plant. R is gas constant and Q is activation energy for Cr diffusion which is 125 kJ/mol [6]. To simulate service time 30 years, heat treatment is needed about 2750 hours at 450°C. We make 4 types of specimens, which simulated service time 15 years, 30 years, 60 years and non-heat treated.

2.3 Procedure

Every specimen used for experiments was cut off from weld root of fusion boundary region of each heat treated mock-up samples. Specimens are polished up to 0.05- μm with colloidal silica and etched with 20% $\text{HNO}_3 + \text{HCl}$ solution about 3 minutes.

3. Results

3.1 Microstructure analysis with SEM.

As shown in Fig. 1, type-II boundaries were found in the whole specimen. The type-II boundaries seem to be arranged and be shifted away from the fusion boundary as the heat treatment was conducted. Over the type-II boundaries, a dendrite structure was found which is a typical structure found in weld metals, formed by heat diffusion during the welding process.

From the chemical composition analysis with the EDX, no composition trends were detected by the existence of type-II boundary as shown in Fig. 2. However, at the narrow zone between the type-II boundary and fusion boundary, the change of composition was sharp. Furthermore, slight increasing of chromium contents were observed at the narrow zone between the fusion boundary and type-II boundary as the heat treatment progressed.

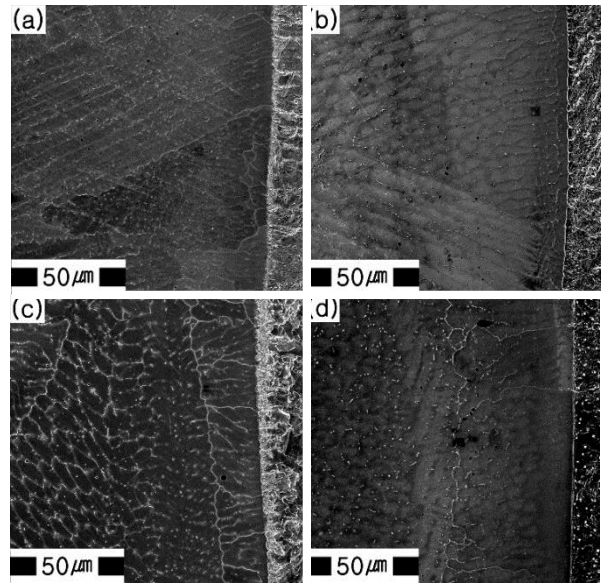


Fig. 1. Microstructures near type-II boundaries of (A) As-welded, (B) HT450_Y15, (C) HT450_Y30, AND (D) HT450_Y60

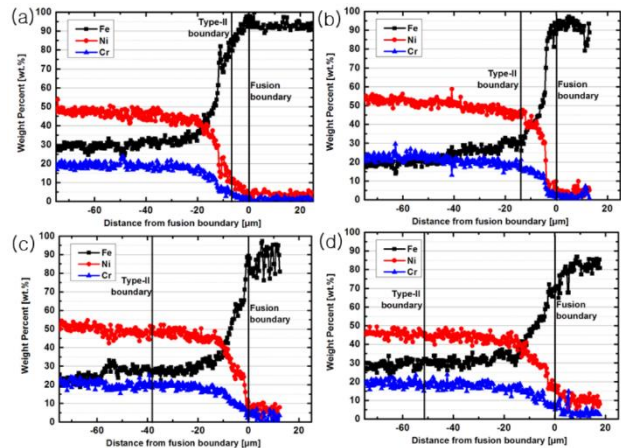


Fig. 2. EDX analysis data of (a) As-welded, (b) HT450_Y15, (c) HT450_Y30, AND (d) HT450_Y60.

3.2 Orientation analysis with orientation imaging microscopy

Orientation imaging microscopy with EBSD was performed near the type-II boundary and fusion boundary. The misorientation of grain boundaries and the grain average misorientation (GAM) were analyzed for each specimen.

The grain boundary characters play an important role in the resistance to SCC growth or initiation. Previous studies often categorize the grain boundaries into three groups: low-angle grain boundaries, coincidence site lattice (with Σ numbers), and high-angle grain boundaries. Generally, low-angle grain boundaries and low-number- Σ boundaries are known to have resistance to SCC. High-angle grain boundaries have high energy that could induce the segregation and formation of precipitates. Because most precipitates formed in nickel-based weld metals are chromium precipitates, the chromium dilution zone could be formed near high-angle

grain boundaries, which could increase the susceptibility of alloys to SCC [3-5].

GAM is a local misorientation parameter defined by the average misorientation within neighboring grains. It is noted that previous studies about cold worked metals conclude that GAM is proportional to the local strain [4]. Therefore, residual strain induced by welding or oxidation could be analyzed with the GAM value of the EBSD analysis. The residual strain is plays a key role in the susceptibility of alloys to SCC [4, 5].

As shown in Fig. 3, a slight increase of low-angle grain boundaries was observed as the heat treatment progressed. The GAM was highest at the narrow zone between the type-II boundary and fusion boundary as shown in Fig. 4, and no significant changes occurred by heat treatment. Several studies noted that GAM is proportional to the residual strain, which could increase the susceptibility of alloys to SCC [4, 5].

Therefore, the narrow zone between the type-II boundary and the fusion boundary could be more susceptible to SCC than any other region.

3.3 Nanoindentation

Nanoindentations were performed in the specimen 1) within the narrow zone between the type-II boundary and fusion boundary, as shown in region (a) of Fig. 5 or 2) within the region about 80 μm away from the fusion boundary in the weld metal, as shown in region (b) of Fig. 5. In both tests, the nanohardness was lowest for the non-heat-treated specimen as shown in Fig. 6. The hardness increased for the heat-treated specimen simulating 15 years, and the hardness decreased for heat-treated specimen simulating more than 15 years of service. The influence of the heat treatment was less effective in the region away from the fusion boundary. The hardness was slightly changed by the heat treatment, but this change was much smaller than the change near the type-II boundary.

The average hardness was higher in the narrow zone between the type-II boundary and fusion boundary. This result is compatible with the result from previous studies, concluding that residual strain at the narrow zone between the type-II boundary and fusion boundary was the highest [4, 5].

There are several factors affecting the hardness in the weld region: precipitates, residual strain and grain boundary misorientations near grain boundaries, and so on.

Therefore, factors affecting the hardness at the time region between non-heat-treated and simulating operation of 15 years might be different from the factors affecting the hardness in later stages.

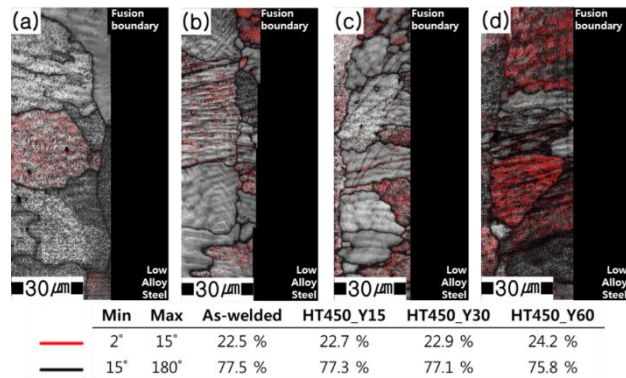


Fig. 3. Map of grain boundary misorientation angle: (a) As-welded, (b) HT450_Y15, (c) HT450_Y30, AND (d) HT450_Y60

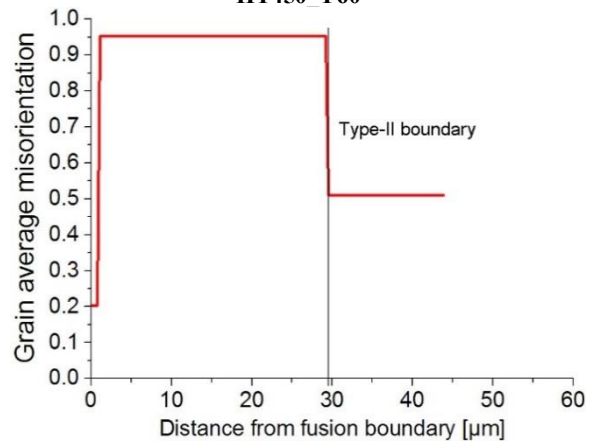


Fig. 4. GAM analysis result at arbitrary line intersecting fusion boundary and type-II boundary of HT450_Y60

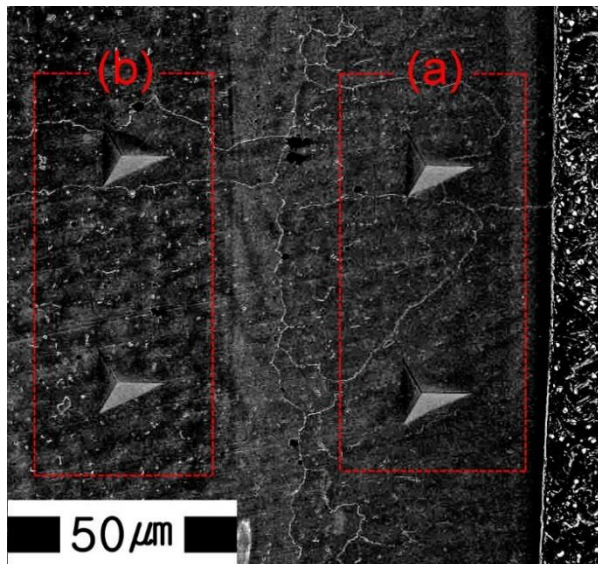


Fig. 5. Nanoindentation tracks

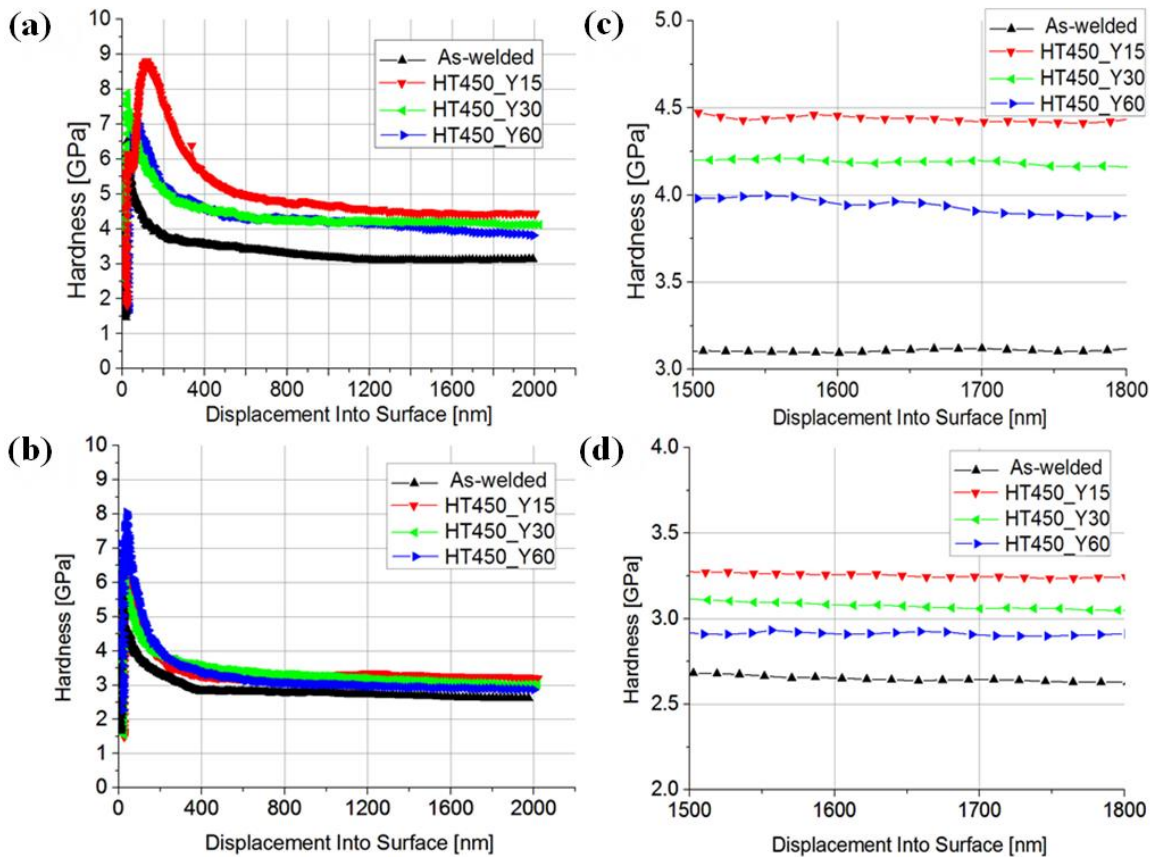


Fig. 6. Hardness-displacement into surface graphs of nanoindentation tests. (a) and (c) are result tested in the narrow zone between type-II boundary and fusion boundary, (b) and (d) are result tested over the type-II boundary. (c) and (d) are a more magnified view of (a) and (b), respectively

4. Discussion

4.1 The effect of thermal aging

The purpose of this study is to analyze the detailed microstructure near the type-II boundary in the weld metal side of the DMW between Alloy 152 and A533 Gr. B with heat treatment to simulate thermal aging.

From scanning electron microscopy analysis, the type-II boundaries were found in the all specimen including as-welded and aged samples. Moreover, the type-II boundaries seem to be aligned and then be shifted away from the fusion boundary as the heat treatment proceeded. From previous studies, crack growth through the type-II boundary was observed; however, the concept of the type-II boundary was established recently, so few studies explicitly discuss the type-II boundary. From the images of the cracks, it is clear that cracking occurs along the type-II boundary [4, 5]. As a result, the migration of the type-II boundary could induce several SCC related problems. Further experiments, including a crack growth test, will be needed to confirm this.

After heat treatment, the EDX result shows increasing chromium content in the dilution zone of weld metal. The chromium dilution zone is formed by the precipitation of carbides with the carbon from the base metal near the fusion boundary. From this result, the precipitates could be formed in the early stages of

thermal aging and coarsen or migrate during later stages of thermal aging. Previous studies found that the precipitates were formed at the early stage of thermal aging, and they only coarsen or migrate to the highly energetic boundary [7, 8]. The coarsening of precipitates is known to reduce the resistivity of alloys to SCC.

From the EBSD analysis, an increase in the low-angle grain boundaries is observed with the heat treatment. Generally, low-angle grain boundaries are known to reduce susceptibility to SCC. Furthermore, a high GAM value is observed at the narrow zone between the type-II boundary and fusion boundary. Previous studies conclude that GAM value is proportional to residual strain which could increase susceptibility of materials to SCC [4].

4.2 Nanohardness based analysis

As seen from the nanoindentation test results, the nanohardness was highest for the heat treatment simulating an operation time of 15 years and was lowest for the non-heat-treated specimen. Based on thermal aging effects described above, the parameters affecting nanohardness should be considered during the heat treatment process.

At the early stage of heat treatment, i.e., until heat treatment simulating an operation time of 15 years, the precipitation of chromium carbide might significantly affect the nanohardness. For this reason, the hardness of

the specimen simulating an operation time of 15 years is the highest. There are corresponding studies concluding that the majority of precipitates are formed in the early stages of thermal aging [7, 8]. Furthermore, these precipitates only coarsen and migrate after formation, so the precipitates are less affects the nanohardness after the early stages of heat treatment.

From the next stage of heat treatment, i.e., heat treatments simulating an operation time of 30 or more years, the characteristics of the grain boundaries and residual strain significantly affect the nanohardness. The increase in low-angle grain boundaries near the type-II boundary could reduce the hardness because precipitates are formed and located at the highly energetic grain boundaries, which are high-angle grain boundaries [7, 8]. Furthermore, the relaxation of residual stress can reduce the hardness.

5. Conclusions

Microstructural, grain boundary orientation, nanohardness analysis were conducted in the type-II boundary and fusion boundary region of the DMW between Alloy 152 and low-alloy steel A533 Gr. B in order to investigate the effect of thermal aging influence.

Type-II boundaries are observed in the whole specimen, which seem to be arranged and then shifted away from fusion boundary as the heat treatment is applied.

Increasing low-angle grain boundaries were observed as the heat treatment proceeded. The GAM has the highest value at the narrow zone between the type-II boundary and fusion boundary.

Chromium contents increased in the weld metal side of the DMW as the heat treatment was applied. And chromium dilution zone of weld metal was decreased as heat treatment applied over that of simulating operation time 30 years.

Nanohardness was the highest for the heat treatment simulating an operation time of 15 years. From the changes that occurred during the heat treatment, we infer that different parameters affect the nanohardness at each stage of the heat treatment.

At the early stage of thermal aging, i.e. until operation time of 15 years, the precipitation of chromium carbide might significantly affect the nanohardness. From the next stage of thermal aging, i.e. operation time of 30 years or more years, the characteristics of the grain boundaries and residual strain might significantly affect the nanohardness.

Acknowledgments

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