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IG Cracking of SA508 by Thermal Treatment at 350-420°C

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1. Introduction

A reactor pressure vessels(RPV) of light water reactor(LWR) become brittle due to exposure to fast neutron(E>1MeV) irradiation during commercial nuclear reactor operation. This embrittlement behavior of the RPV should be evaluated periodically. This phenomenon is called irradiation embrittlement because it is understood that it occurs by fast neutron irradiation. The embrittlement behavior of the pressure vessel material is quantified by evaluating the shift of the reference temperature nil ductility temperature (RT_{NDT}) of Charpy impact energy curve with temperature as shown in Fig. 1 [1]. This evaluation method evaluates the degree to which the left curve shifts to the right in Figure 1.

The premise of irradiation embrittlement in RPV is that it is caused by fast neutron irradiation. However, according to the IAEA report [2], a surveillance specimens installed at locations where there is little fast neutron irradiation on a Russian LWR show a transgranular (TG) and/or an intergranular (IG) fracture as shown in Figure 2. This specimen only has an only thermal history without fast neutron irradiation. IG fracture is a typical of brittle fracture manner in various materials.

Although transgranular cleavage is the predominant mode of brittle fracture in RPV steels, solute (e.g. phosphorus) segregation to grain boundaries can result in another type of brittle fracture known as intergranular (grain boundary) fracture. Figures 1 a) and b) show examples of transgranular and intergranular (IG) fracture, respectively, as viewed in a scanning electron microscope [2-4]. The investigators have interpreted the IG cracking occurs as a result of segregation of sulfur and/or phosphorus at grain boundary [3, 4]. The IG cracking is a kind of symptom of embrittlement. It is reported that the IG cracking occurs in inert (Ar) environment under slow strain rate test [5].

Recently, it has been reported that the cause of aging of materials is that a decrease in entropy in the process of changing the arrangement of atoms at a temperature at which the diffusion of atoms is possible due to short range ordering (SRO), and a contraction of the lattice occurs in this process [6-8]. Recently, it has been reported that the lattice contraction is a rate controlling process in a primary water stress corrosion cracking phenomenon in Alloy 600. The aging process of the material is caused by a phenomenon in which stress is generated inside the material due to lattice contraction due to the decrease in entropy.

This study is to confirm whether the SRO phenomenon exists in the RPV material and this fundamental phenomenon would cause grain boundary fracture. Thus, RPV material was thermally treated up to 66 months at a temperature slightly higher than the operating temperature of the reactor, and the IG fracture of RPV material were to be confirmed. In this thermal treatment, it was confirmed whether the SRO phenomenon accompanied by a decrease in entropy occurred, and based on this, it was confirmed that the cause of embrittlement of the RPV material was caused by a lattice contraction due to the decrease in entropy. It is concluded that the irradiation embrittlement was based on the SRO phenomenon of the RPV material, and it was judged that the neutron irradiation was a secondary factor that changed the rate of SRO kinetics.

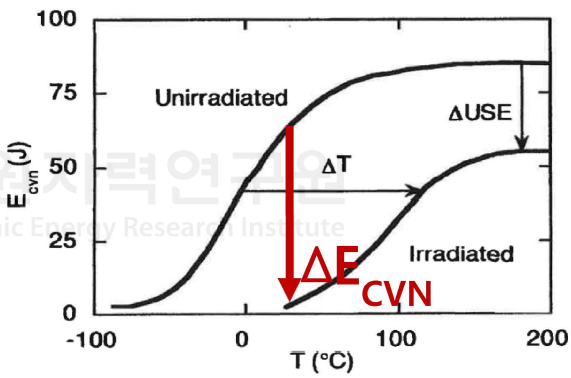


Fig. 1. Illustration of the effect of irradiation on the Charpy impact energy curves in RPV steel [1].

2. Experimental

Chemical composition of SA508 is shown in Table 1. Charpy impact specimens of SA508 were machined in T-L orientation and aged at 350, 400, and 420℃ for 2,250H and for 48,000H (66 months) in air. As received and the aged specimens were tested at 16℃ by a Charpy impact tester. The fracture surfaces of impact specimens were observed by SEM.

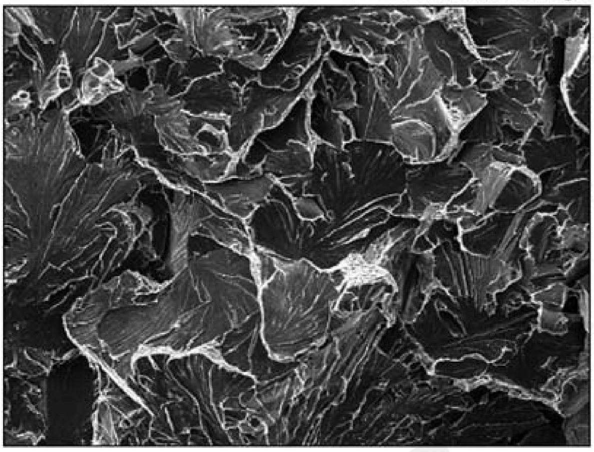
In order to confirm whether the SRO reaction exists in SA508 RPV steel, SA508 specimen is heated to 950oC for 1H and water quenched (WQ). This specimen is subjected to differential scanning calorimeter (DSC) analysis. Whether the exothermic reaction is detected is analyzed.

The lattice variations were determined by XRD using CuKa. The peak is determined by a center gravity of the diffraction. The precise lattice parameter is calculated by Nelson-Riley relationship. The lattice variation during aging was calculated by using a relationship of (Ao_{aged} - $Ao_{as\text{-}received}$)/$Ao_{as\text{-}received}$.

Table 1. Chemical composition of SA508

Elements	C	Si	Mn	P
%	0.18	0.04	1.43	0.0025
S	**Ni**	**Cr**	**Mo**	**V**
0.001	0.85	0.22	0.55	<0.0025

a) transgranular (TG) type



b) intergranular (IG) type

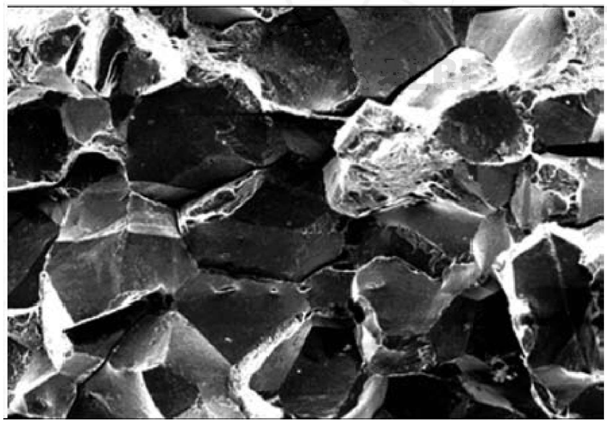


Fig. 2. Fracture surfaces of pressure vessel steel, a) transgranular type and b) intergranular type in Russian LWR [2].

3. Results and Discussions

Figure 3 shows the rate of decrease in Charpy impact energy according to the aging time. At 420°C, the initial decrease appears to be large, but the rate of decrease at 48,000 hours is the lowest. The specimens aged for 2,250 hours at 350, 400, and 420°C showed a 4-25% reduction in impact energy at 16°C.

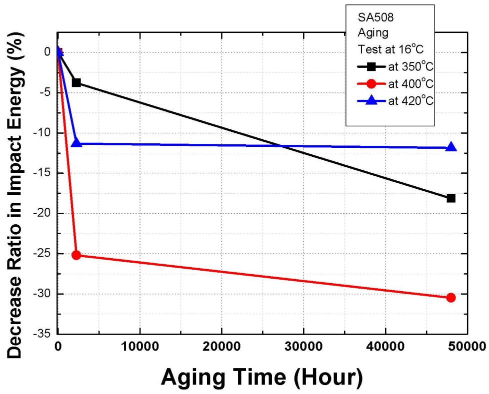


Fig. 3. Decreasing ratio of impact energy at 16°C by thermal treatment at 350-420°C.

On the other hand, the specimens aged for 48,000 hours showed a reduction in impact energy of about 12-30%. What this result means is that the impact energy is reduced only by thermal treatment without any neutron irradiation. This means that the decrease in fracture toughness of the RPV material is essentially unrelated to neutron irradiation.

Fig. 4 shows the fracture surface of the specimen with reduced fracture toughness of about 37% as a result of thermal treatment at 400°C for 48,000 hours after impact test. It can be seen that certain part of fracture surface have undergone IG fracture. This is similar to the IG fracture observed in the surveillance test specimen of a Russian LWR, as shown in Fig. 2.

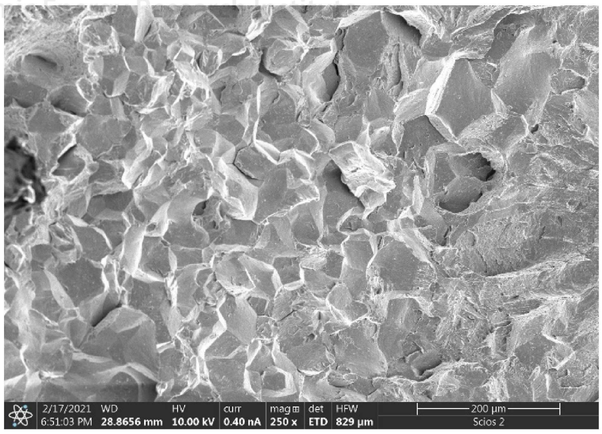


Fig. 4. IG cracking in thermal treatment at 400°C for 48,000H.

In order to investigate the intrinsic phenomena occurring in the pressure vessel material, the SA508 material was maintained at 950° C for 1 hour and then water-quenched (WQ) in order to perform thermal analysis through differential scanning calorimeter (DSC). The results of thermal analysis are shown in Fig. 5, and the WQ SA508 material shows an exothermic peak at below 350°C. The meaning of this heat generation is that the entropy remaining in the lattice decreases as it is quenched at a high temperature.

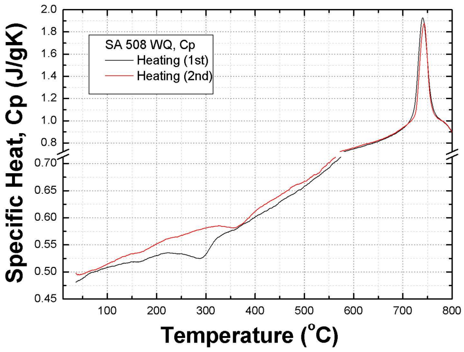


Fig. 5. Specific heat variation with temperature showing an exothermic reaction at 80-370°C.

Entropy reduction according to the SRO process causes lattice contraction. Therefore, XRD was used to investigate the lattice contraction behavior according to the aging treatment, and the relationship between lattice contraction and impact energy reduction is shown in Fig. 6. The amount of impact energy decrease appeared 4-25% according to aging temperature, compared to as-received SA 508. The amount of decrease in impact energy appered 4, 25, 11% in aged 350, 400, 420℃ for 2,250H, respectively. The lattice variation determined by XRD appeared +0.004, -0.022, -0.012% in 350, 400, 420℃ for 2,250H, respectively. It is possible to understand that the decrease in impact energy is proportional to the amount of lattice contraction.

The decreasing rate of Charpy impact energy in the specimens thermally treated for 2,250 hours shows a nearly linear relationship with the decrease in impact energy. In other words, the decrease in the impact energy of the pressure vessel material is due to the lattice contraction due to SRO phenomenon.

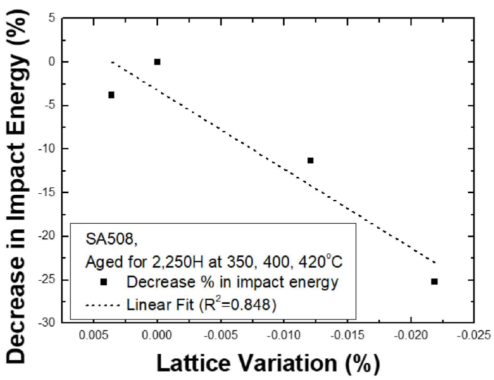


Fig. 6. Decreasing ratio with lattice contraction in aged at 350-420°C for 2,250H.

In observing the change in material properties and identifying the cause of the change, it is more appropriate to explain it as the cause of the fundamental change rather than the macro one. Atomic ordering (SRO) is caused by a decrease in entropy depending on the arrangement of atoms. As a result, the distance between the atoms becomes closer, and this is the concept that stress is generated by itself inside the material. These changes reduce the length of the material and make the grain size smaller, resulting in tensile stresses in grain boundaries. This is the root cause of the IG cracking by aging of structural materials.

Fig. 7 is to conceptually explain what happens at the grain boundary when individual grains of a polycrystalline material shrink. When each grains of the material contract anisotropically, a tensile force that spreads the grain boundaries acts. If this is greater than the attractive force of an atom or grain boundary, the grain boundary is split. Or, vacancy and/or void could be formed at the grain boundary, and this is connected to each other to cause IG cracking or grain boundary destruction. All process explained above will reduce the fracture toughness RPV steel.

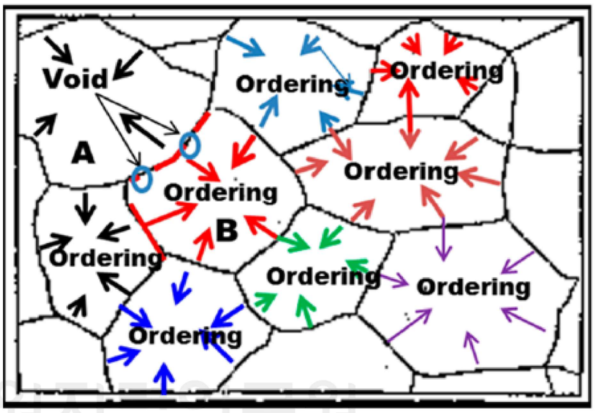


Fig. 7. Conceptual explanation of intergranular (IG) cracking in polycrystalline material due a lattice contraction.

The reduction of the impact energy not only increases the RT_{NDT}, but also has the effect of reducing the energy of the transition region as shown in red arrow in Fig. 1. The surveillance test analyzes the irradiation embrittlement effect based on the transition phenomenon of RT_{NDT}, but in this study, the energy change at 16°C corresponding to the transition region was investigated.

In order to track the RT_{NDT} change, an S curve is derived using a number of specimens over a wide temperature range. However, in this study, it was confirmed whether brittleness appeared by tracking the decrease in impact energy at a specific temperature such as 16°C. Since this experiment only derives the impact test results at 16°C, a small number of pieces were used.

In SA508 pressure vessel materials, the lattice contraction caused by the reduction in entropy due to ordering (SRO) creates a tensile stress at the grain boundaries by itself. This stress creates defects such as vacancy or void at grain boundary. These are connected together to be IG crack. Eventually, IG fracture appeared. This is the root cause of the decrease in fracture toughness. Therefore, the root cause of the aging phenomenon of the RPV material is the SRO phenomenon. The fundamental reason of the irradiation embrittlement is essentially due to the SRO phenomenon, and the neutron irradiation effect is believed to cause the acceleration of the SRO kinetics.

Recently, MnNi nano clusters were observed in ion irradiation experiments on the FeMnNi RPV model alloy, and it was interpreted that this may be the cause of irradiation embrittlement [9]. However, the past paradigm of confirming the differences before and after embrittlement is only correlation; this cannot explain the cause of embrittlement.

This study showed that RPV showed embrittlement regardless of neutron or ion irradiation. It has been shown that RPV materials inherently have thermal embrittlement and this cannot be avoided. The interpretation of the results in this study is a new paradigm of materials science based on a causal relationship.

4. Conclusions

1. The SA508 reactor pressure vessel (RPV) material undergoes a SRO phenomenon accompanied by an exothermic reaction.
2. The cause of embrittlement of the RPV material is a lattice contraction caused by entropy reduction due to SRO reaction.
3. IG fracture was observed in the specimens thermally treated at 400°C for 48,000 hours, and it was confirmed that the lattice contraction due to SRO reaction was the fundamental reason for the decrease in fracture toughness of the SA508 RPV material.

Acknowledgements

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