Transactions of the Korean Nuclear Society Autumn Meeting
Gyeongju, Korea, October 29-30, 2015

Thermal Embrittlement of Reactor Pressure Vessel Steel due to Aging

SungSoo Kim and Duck-Gun Park
Korea Atomic Energy Research Institute,
111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Korea

1. Introduction

It is known that a reactor pressure vessel (RPV) undergoes an irradiation embrittlement under neutron irradiation during operation service in pressurized water reactor (PWR) environment [1]. To estimate the ductile-to-brittle transition temperature of RPV materials during operation of PWR, surveillance specimens (SS) are placed in separate containers immediately before RPV wall at different heights relative to the nuclear core in Russia. Irradiated specimen of SS allows forecasting changes in liability of RPV materials to brittle fracture by periodical extraction of a part of the specimens followed by mechanical tests. Thermal SS sets are located above the nuclear core where a fast neutron flux is negligible and temperature is 320℃ (as opposed to 290℃ in locations of high-irradiated SS). These SS allow monitoring of continuous operation temperature exposure effect on mechanical characteristics of the steels [2].

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 [1-3]. The investigators have interpreted the intergranular cracking occurs as a result of segregation of sulfur and/or phosphorus at grain boundary [2, 3].

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 [4]. In addition, it is proposed that the intergranular cracking occurs due to a lattice contraction in Ni-base alloys without neutron irradiation [5, 6]. The fact that the intergranular cracking is occurred by thermal treatment in RPV steel without neutron irradiation implies there is another damage mechanism decreasing toughness except radiation damage.

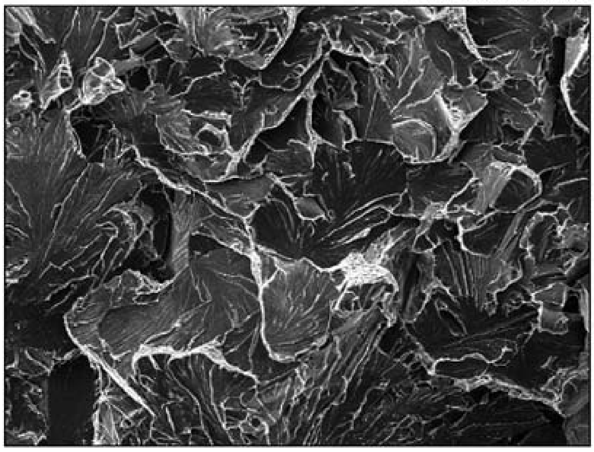
Whether thermal aging without neutron irradiation causes IG cracking and embrittlement is checked in this study. The thermal aging is applied to SA508 RPV steel, the microstructure and lattice variation with thermal aging is examined by electron back scattered diffraction (EBSD), TEM, and XRD, respectively, and a decrease in Charpy impact energy with thermal aging is correlated with lattice variation.

2. Experimental

Charpy impact specimens of SA508 were prepared and aged at 350, 420, and 420℃ for 2,250H. The aged specimens were tested at 17℃ by a Charpy impact tester. The microstructures of aged specimens were examined by EBSD and TEM after electro polishing by 10% perchloric + methanol solution in the transverse normal plane. The fracture surfaces of impact specimens were observed by SEM.

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 $(Ao_{aged} - Ao_{as\text{-}received})/Ao_{as\text{-}received}$.

a) transgranular type



b) intergranular type

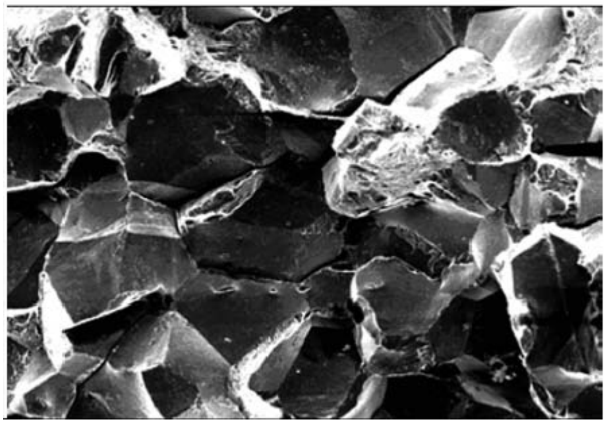
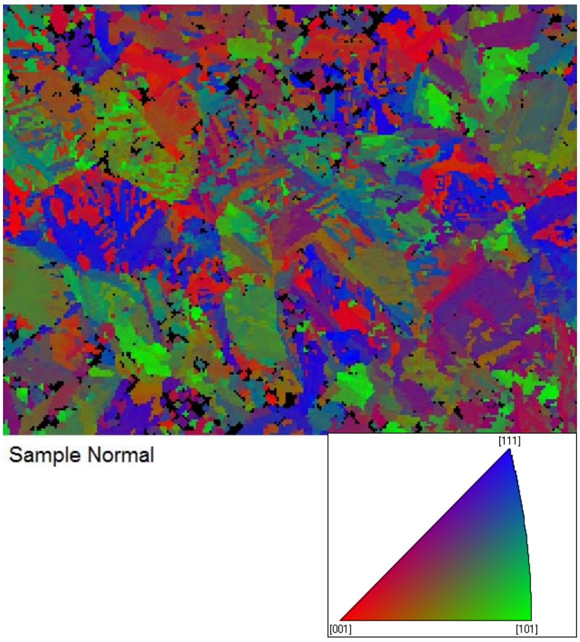


Fig. 1. Fracture surfaces of pressure vessel steel, a) transgranular type and b) intergranular type [1].

3. Results and Discussions

The microstructure and grain orientation examined by EBSD are shown in Fig. 2 a) as-received and b) aged at 400℃ for 2,250H. The grain size is slightly increased by thermal aging. Although the pole figure is not shown here, the grain orientation is slightly changed with aging.

a) as-received



b) 400°C-2,250H

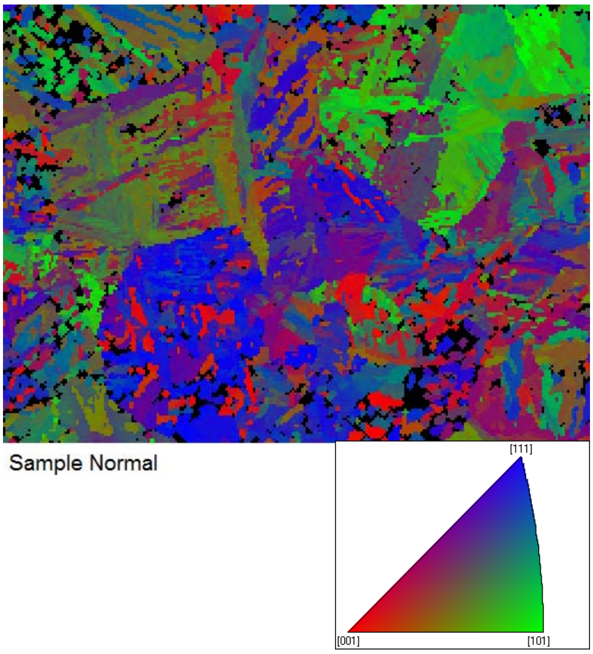


Fig. 2. Comparisons of grain orientation of SA 508 in a) as-received and b) aged at 400℃ for 2,250H.

The fracture surfaces of Charpy impact specimen observed by SEM in low magnification are compared in Fig. 3. The fracture surface in aged at 400℃ for 2,250H is different from that of as-received specimen. The aged at 400℃ for 2,250H specimen shows a certain delamination, compared to as-received one. This suggests that the aged specimen have suffered certain damage.

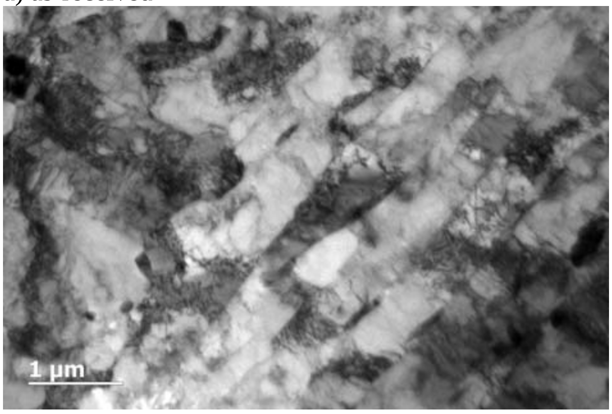
a) as-received

b) aged at 400℃ for 2,250H

Fig. 3. Comparisons of fracture surfaces of SA 508 in a) as-received and b) aged at 400℃ for 2,250H.

The microstructures by TEM in as-received and aged at 400℃ for 2,250H is compared in Fig. 4. The as-received SA 508 shows elongated lath structure and there are little carbides. However, the microstructures reveal that the aged at 400℃ for 2,250H specimen shows a relatively larger lath and carbides frequently.

a) as-received



b) aged at 400℃ for 2,250H

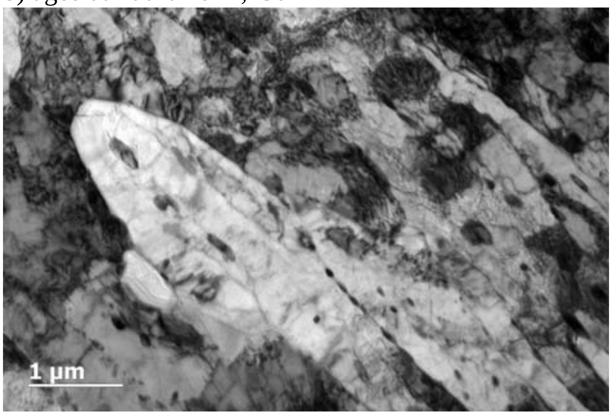


Fig. 4. Comparisons of TEM microstructure of SA 508 in a) as-received and b) aged at 400℃ for 2,250H.

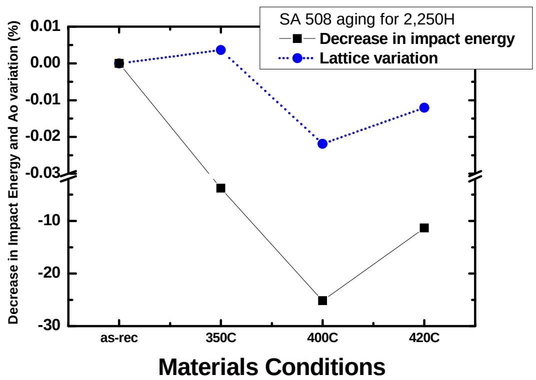


Fig. 5. Comparisons of lattice variations and decrease in impact energy in as-received and aged SA508.

The decrease in impact energy due to thermal aging and the lattice variation are compared in Fig. 5. The amount of impact energy decrease appeared 4-25% according to aging conditions, 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 reason of IG cracking in RPV is suggested that the segregation of P [2] and P, C, Mo and Ni [3] causes IG brittle fracture at grain boundary. The segregation of certain elements on the grain boundary is confirmed; however, how the segregation causes a brittle fracture of grain boundary is still not clear.

The lattice contraction observed in this study may be a reason for segregation of impurities at grain boundary, since the impurities inside grain stay under strain filed due to a state of solid solution and therefore these tend to precipitate or segregate. This means that the lattice contraction may be a main reason for a decrease in toughness and eventually for IG cracking in RPV steel during thermal aging.

The main reason why a lattice contraction occurs during thermal aging is a changing process for a equilibrium state of material. It is generally acceptable that any fabrication process does not allow reaching a stable state.

4. Conclusions

1. The lath grain size in SA508 RPV steel increases slightly due to thermal aging at 350, 420, and 420℃ for 2,250H.
2. The decrease in toughness appeared 4-25% and the lattice contraction appeared to be +0.004% - -0.022% due to thermal aging at 350, 420, and 420℃ for 2,250H.
3. The amount of decrease in Charpy impact energy due to thermal aging is correlated well with the magnitude of lattice contraction.

Acknowledgements

This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korea government (MSIP).

REFERENCES

[1] IAEA report No. NP-T-3.11, 'Intergrity of Reactor Pressure Vessel in Nuclear Power Plants: Assessment of Irradiation Embrittlement Effects in Reactor Pressure Vessel Steels', P. 25, (2009).
[2] Oleg O. Zabusov1, Boris A. Gurovich, Evgenia A., Kuleshova, Michail A. Saltykov, Svetlana V. Fedotova, Alexey P. Dementjev, Key Engineering Materials Vol. 592-593 (2014), pp 577-581
[3] Y. Nishiyama, K. Onizawa, M. Suzuki, J.W. Anderegg Y. Nagai, and T. Toyama, Acta Materialia 56 (2008) 4510–4521
[4] T.M. Angeliu, D.J. Paraventi, G.S. Was, Corrosion 51 (1995) 837.
[5] S. Kim, D. W. Kim, and Y. S. Kim, Met. Mater. Int., Vol. 19, No. 5 (2013), pp. 969~974
[6] Y. S. Kim, W. Y. Maeng, and S. Kim, Acta Materialia 83 (2015) 507-515