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The Primary Water Stress Corrosion Cracking Mechanism of Alloy 600 Steam Generator Tubes: Materials Perspective

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1. Introduction

It is well known that Alloy 600 steam generator tubes are susceptible to primary water stress corrosion cracking (PWSCC) in pressurized water reactors. However, the PWSCC mechanism remains elusive. Either enhanced corrosion or oxidation of grain boundaries has been suggested to be the cause of intergranular stress corrosion cracking of Alloy 600 steam generator tubes. The problem is that intergranular (IG) cracking of austenitic Fe-Cr-Ni alloys occurs even in Ar with no corrosion or oxidation of grain boundaries being accompanied [1,2]. This fact suggests that IG cracking has nothing to do with grain boundary (GB) corrosion or oxidation. Another thing to note is that IG cracking of Alloy 600 occurs after exposure to primary water of 325°C without applied stress for several hundreds to thousands hours [3]. This fact cast a doubt about the current notion that applied stresses are required to initiate IG cracking or PWSCC. These facts indicate that PWSCC is closely related to internal factors of materials, not to external factors such as grain boundary oxidation or corrosion or applied stresses.

Given that austenitic alloys including Alloy 600 are a kind of solid solution alloys with alloying elements dissolved in the matrix as solutes, ordering of alloying elements of Fe, Cr and Ni occur in Alloy 600 during exposure to reactor operating condition. We suggest that atomic ordering is the main internal factor to govern PWSCC or IG cracking of austenitic Fe-Cr-Ni alloys because lattice contraction due to atomic ordering induces internal stresses which are large enough to cause GB cracking. The aim of this work is to provide experimental evidence for our suggestion. To this end, water quenching (WQ) or air cooling (AC) or furnace cooling (FC) was applied respectively to Alloy 600 after solution treatment at 1095°C for 0.5h to make Alloy 600 with either disorder (DO) or different degrees of short range order, respectively.

2. Methods and Results

To simulate atomic ordering of Alloy 600 during reactor operating conditions, the WQ-, the AC- and the FC-Alloy 600 were aged at 400°C for up to 5,500h. To evaluate the degree of atomic ordering, the lattice spacing of three different kinds of Alloy 600 were determined using neutron diffraction after aging at 400°C. As shown in Fig. 1, the WQ-Alloy 600 showed the largest amount of lattice contraction and the FC-Alloy 600 the least amount of lattice contraction. Note that the lattice contraction shown in Fig. 1 represents the relative values when compared to the unaged Alloy 600. The largest amount of lattice contraction for the WQ-Alloy 600 upon aging at 400°C indicates that the WQ-Alloy 600 with disorder experiences the highest amount of atomic ordering, transforming to short range order from disorder. In contrast, the FC-Alloy 600 with a higher degree of short range order experiences the least amount of atomic ordering upon aging at 400°C, leading to the smallest amount of lattice contraction.

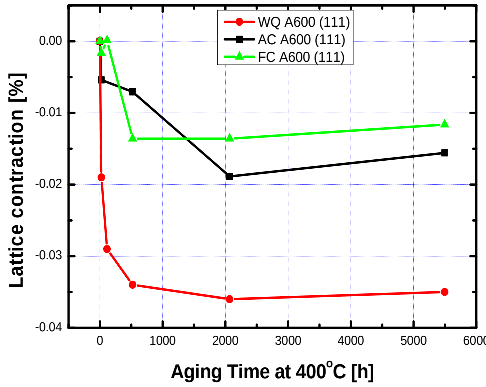

Fig. 1. The amount of lattice contraction of Alloy 600 upon aging at 400°C with cooling rate after solution annealing.

Given our suggestion, the WQ-Alloy 600 with the largest amount of lattice contraction would show the lowest resistance to PWSCC while the FC-Alloy 600 with the lowest amount of lattice contraction the highest resistance to PWSCC. Definitive evidence is provided by Yonezawa's experiment [4] where SCC tests were conducted at 332°C in simulated primary water for 23,939h using the U-bend specimens of the WQ-, the AC- and the FC-Alloy 600 which were given water quenching, air cooling or furnace cooling, respectively, after solution annealing at 1025°C for 2h. As expected, the WQ-Alloy 600 showed 100% failure while the AC- and FC-Alloy 600 had zero failure. Thus, it is evident that the WQ-Alloy 600 with the largest amount of lattice contraction is the most susceptible to PWSCC and the FC-Alloy with the least amount of lattice contraction the least susceptible to PWSCC. This observation definitively demonstrates that the amount of lattice contraction due to atomic ordering governs PWSCC of Alloy 600. Furthermore, this rationale can account for the reason why HTMA (high temperature mill annealed) Alloy 600 steam generator tubes in one of 6 PWR units in Korea showed the most severe IGSCC when compared to the other units. Note that HTMA Alloy 600 steam generator tubes in 6 PWR units showed the carbides precipitated at the grain boundary whose coverage at grain boundaries exceeds 20%. Considering Yonezawa's summary [4] revealing that the crack initiation time of Alloy 600 has no relevance to the coverage ratio of grain boundary carbides as long as it exceeds 20%, the hypothesis that the coverage ratio of grain boundary carbides governs PWSCC of Alloy 600 is evidently invalid.

3. Conclusions

Alloy 600 showed lattice contraction upon aging at 400^{o}C whose extent increased with increasing cooling rate: the water-quenched (WQ) Alloy 600 exhibited the largest amount of lattice contraction than the furnace-cooled (FC) or air-cooled (AC) one. Yonezawa's experiments have indeed shown that the WQ-Alloy 600 with the largest amount of lattice contraction upon aging at 400^{o}C is the most susceptible to PWSCC when compared to the AC- or FC-Alloy 600 with the lesser amount of lattice contraction. These observations demonstrate, for the first time, that PWSCC of Alloy 600 is governed by the amount of lattice contraction due to atomic ordering during reactor operation.

REFERENCES

[1] T. Onchi, K. Dohi, N. Soneda, M. Navas, and M.L. Castano, Mechanism of irradiation assisted stress corrosion crack initiation in thermally sensitized 304 stainless steel, Journal of Nuclear Materials, Vol. 340, p. 219, 2005.
[2] T.M. Angeliu, and G.S. Was, Creep and intergranular cracking of Ni-Cr-Fe in 360^{o}C in Argon, Metallurgical and Materials Transactions A, Vol. 25, p. 1169, 1994.
[3] L. Fournier, O. Calonne, P. Combrade, P. Scott, P. Chou, R. Pathania, Grain boundary oxidation and embrittlement prior to crack initiation in Alloy 600 in PWR primary water, 15th International Conference on Environmental Degradation, p. 1491, 2011.
[4] T. Yonezawa, Private communication.