Transactions of the Korean Nuclear Society Spring Meeting
Jeju, Korea, May 18-19, 2017

PWSCC Behavior of Alloy 600 under Long-term Thermal Aging and Triaxial Stress

Seung Chang Yoo [a], Kyoung Joon Choi [a], Ji Soo Kim[b], Byoung-Ho Choi[b], Yun-Jae Kim[b], Jong-Sung Kim[c] and Ji Hyun Kim[a]*

[a] *Department of Nuclear Engineering, School of Mechanical, Aerospace and Nuclear Engineering, Ulsan National Institute of Science and Technology, 50 UNIST-gil, Ulsan, 44919*
[b] *Department of Mechanical Engineering, Korea University, 145, Anam-ro, Seongbuk-gu, Seoul, 02841*
[c] *Department of Nuclear Engineering, Sejong University, 209, Neungdong-ro, Gwangjin-gu, Seoul, 05006*

**Corresponding author: kimjh@unist.ac.kr*

1. Introduction

Components used in nuclear power plant (NPP) are exposed to very corrosive environment including high temperature, pressure, corrosive coolant, radiation and residual stress, etc. Among this, several parts that manufactured with nickel-based alloys, such as steam generator tubes, heater sleeves and head penetration nozzle for control rod drive mechanisms, are suffer from cracking and leakage of coolant. In particular, Alloy 600 has been reported to be susceptible to primary water stress corrosion cracking (PWSCC) in the primary circuit of NPP, which is caused by a susceptible microstructure, corrosive environment and tensile stress, generally weld residual stress.

Many studies were conducted to find out the mechanism of PWSCC. Scott et al. investigated Alloy 600 in various environments to correlate intergranular SCC (IGSCC) with oxidation phenomenon [1]. The researchers suggested that IGSCC is caused by internal oxidation, induced by diffused oxygen through the grain boundary of nickel-based alloys in redox potential condition. Furthermore, SCC tests evaluated the resistance of nickel-based alloys to crack initiation and crack growth. Alloys 600 and 690 were tested in a primary water coolant environment to determine the influence of the material`s properties on susceptibility to SCC initiation [2].

However, several reports pointed out that there still exists lack of understanding about SCC and more study and effective predictive model is required. In particular, long-term thermal aging during long-term operation of NPP and triaxial stress applied near welding spots of heat penetration nozzle due to weld residual stress and geometrical complexity are important factors related to PWSCC susceptibility, which were not studied enough.

Long-term thermal aging is issued to be an important parameter to material degradation as the operation time of an NPP increases. It is essential to understand the long-term thermal aging effect on the properties of a material, however, only a few studies have investigated this aspect. Also, most of studies considering a heat effect to material were mostly conducted at elevated temperature above 600 ℃ and very short time of several hundreds of hours, which is not close to real case of NPP environment.

In addition, stress state near head penetration nozzle of NPP should be suspected to multi-axial since welding residual stress or geometrical complexity could cause multi-axial stress state at those parts. However, most of SCC studies were conducted in mono-axial stress state with U-bend test or slow strain rate test (SSRT) with smooth bar. It is therefore essential to analyze the effect of triaxial stress in order to understand the nature of PWSCC.

The objective of this study is to investigate the effect of long-term thermal aging at a relatively low temperature (~ 400 ℃) and triaxial stress to PWSCC initiation susceptibility of Alloy 600 through SSRT with active loading which requires relatively short test time compared to other methods such as constant load or U-bend test.

2. Experimental

A representative thick-walled Alloy 600 (74.49Ni, 15.83Cr, 8.4Fe, 0.56Mn, 0.33Si, 0.07C, 0.02Cu, <0.001S) provided by Doosan Heavy Industries & Construction was used in this study. It was fabricated according to ASTM B166 by annealed at 1060 ℃ for 3.5 h and water quenched.

The material was then heat treated in an Ar environment to simulate the effect of actual long-term thermal aging. Thermal aging was applied via heat treatment at accelerated temperature condition since it need too long time to duplicate the actual thermal history of Alloy 600 in a NPP (10 or 20 years at 320 ℃). The temperature used was 400 ℃ which limits excessive formation of carbides or sigma phases that do not form at 320 ℃ according to thermodynamic calculation [3].

The heat treatment conditions were determined and performed based on equation (1), shown below, which is a modified form of the Arrhenius diffusion equation, calculating the required heat treatment time at 400 ℃

to simulate the long-term thermal aging at reactor-operation conditions, i.e., 320 ℃.

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

In this equation, R=8.314 J/mol/K, t_{aging} is the heat treatment time needed at 400 ℃ to simulate long-term thermal aging in a NPP, and t_{ref} is the service time at 320℃ of a NPP or the time that would be simulated. The heat treatment temperature is represented by T_{aging} (400 ℃), and T_{ref} is the actual operation temperature of the NPP (320 ℃). For activation energy Q, the Cr diffusion energy along the grain boundary (180 kJ/mol [4]) was selected since Cr is known to play an important role in the precipitation and corrosion resistance of material.

Specimens were prepared for simulating materials aged in NPP for 0 (as-received state), 10 and 20 years. The heat treatment durations at 400 ℃ (corresponding to 10 and 20 years at 320 ℃) were 1142 and 2284 hours, respectively. Table 2 shows the specimen I.D. (HT + 'heat treatment temperature [°C]' – Y + 'simulated aging time [year]'), simulated aging time and temperature of each specimen.

For PWSCC initiation test, specimens were cut by electric discharge machining to the shape of proportionally reduced ASTM standard according to ASTM E8-E8m. Two types of specimens were subjected to PWSCC initiation test; smooth specimen and 33% reduced cross section specimen with notch to apply triaxial stress on the notched section.

For SSRT test, the strain rate was selected as 1.00×10^{-7}/s according to ASTM G129 and it was controlled via displacement rate control of actuator [3]. Strain rate of smooth and notched specimen was maintained to be equal by applying different displacement rate which were calculated through finite element analysis. Corresponding displacement rate for the test with 6 specimens was 1.30×10^{-5} mm/s for smooth specimens, and 2.66×10^{-6} mm/s for notched specimens. Total 6 specimens were tested in one set of experiment with online monitoring system via direct current potential drop (DCPD) method.

Potential drop due to reduce of cross sectional area due to PWSCC initiation was monitored and PWSCC initiation time of each specimen was calculated by the method used in previous study [3]. Cubic polynomial fitting was conducted for each DCPD curves to find out the inflection point which is defined as PWSCC initiation point and then PWSCC initiation time was measured.

All tests were performed in the recirculating water loop and autoclave system with DCPD probe for in-situ observation of PWSCC initiation. Experiments were conducted in simulated primary water condition with two different temperatures of 315 and 340 ℃. The PWSCC initiated specimen was taken from the autoclave and the surface was investigated with scanning electron microscope.

3. Results and Discussion

The representative DCPD graph is shown in Fig. 1. The averaged PWSCC initiation times of each specimen are summarized in Table 1. There exists electrical noise in the graph, however, the inflection point is clearly observed at graph, which indicates PWSCC initiation time.

From the results of SSRT with smooth specimen, PWSCC initiation time was the shortest at 10 years thermally aged specimen and the longest at As-received specimen. At the surface of each initiated specimen, multiple cracks with length of 100 μm and depth of 4 μm were observed. These cracks were acknowledged to be cracks formed by SCC, which exhibited a zigzag shape along the grain boundaries. The reason for this aspect was suspected to the changes of morphology and number densities of precipitates and corresponding chromium depletion formation along grain boundary which were investigated in previous study [3]. The semi-continuous feature of precipitates and chromium depletion at grain boundary make 10 years thermally aged specimen have the highest susceptibility to PWSCC.

Table 1 Measured SCC initiation time of each specimen achieved in SSRT

SCC initiation time [h]	As -received	10 years aged	20 years aged
Smooth	560.9 ± 34.1	427.9 ± 38.7	518.6 ± 1.4
Notched	307.1 ± 16.1	253.1 ± 6.3	205.8 ± 0.3

The trend of PWSCC initiation time of notched specimen, which consider triaxial stress, was differ from that of smooth specimen. 20 years thermally aged specimen has the highest susceptibility to PWSCC while As-received have the lowest. Critical resolved shear stress (CRSS) was calculated with the parameters obtained from microstructural investigation and same trend was figured out with PWSCC initiation time of notched specimen. Also, cracks were mainly observed at the region where shear stress is focused, therefore, CRSS is suspected to be a reason for the decreasing of PWSCC initiation resistance of 20 years thermally aged specimen.

Triaxial stress was considered in several mechanisms of SCC. Localized plasticity mechanism explains about passive oxide film rupture and induced SCC [5]. Triaxial stress plays an important role in activating the slip system of alloys or precipitates. It could activate many slip systems compared to simple uniaxial stress which in turn influence to the aspects of precipitate hardening [6 ,7]. This phenomenon would decrease the effect of precipitate hardening and the peak hardened region could be moved to further thermal aging time (i.e. peak hardened region at above 20 years of thermal aging).

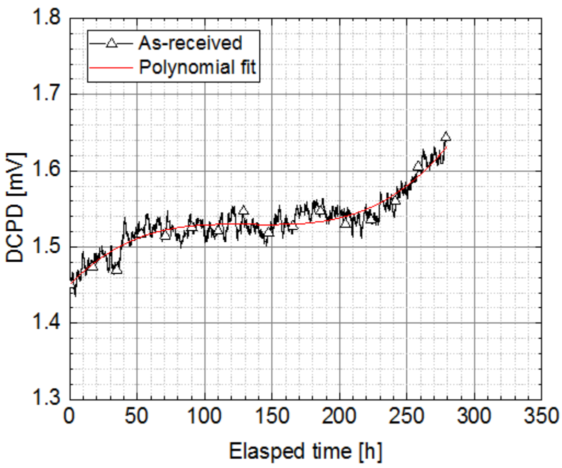

Fig.1 Representative DCPD curve obtained from SSRT.

4. Conclusions

To understand the nature of PWSCC with consideration of the effects of thermal aging and triaxial stress, the PWSCC susceptibility of nickel-based Alloy 600 was investigated via SSRT with in-situ monitoring DCPD system.

At the results of SSRT with smooth specimen, 10 years thermally aged specimen has the highest susceptibility to PWSCC and precipitate morphology of semi-continuous feature and corresponding chromium depletion at grain boundary is suspected to a reason. While 20 years thermally aged specimen has the highest susceptibility in SSRT with notched specimen, and critical resolved shear stress might be responsible to the trend of the results.

Acknowledgment

This work was financially supported by the Nuclear Safety Research Program through the Korea Foundation of Nuclear Safety (KOFONS), granted financial resource from the Nuclear Safety and Security Commission (NSSC), Republic of Korea (No. 1403006) and by the Human Resources Development of the Korea Institute of Energy Technology Evaluation and Planning (KETEP) grant funded by the Korea Government Ministry of Trade Industry and Energy (MOTIE).

REFERENCES

[1] P. Scott, "An Overview of Internal Oxidation as a Possible Explanation of Intergranular Stress Corrosion Cracking of Alloy 600 in PWRs", Proceedings of the 9th International Symposium on Environmental Degradation of Materials in Nuclear Power Systems-Water Reactors pp.3-14 (1999)

[2] M. J. O. M. B. Toloczko, D. K. Schreiber, S. M. Bruemmer, "Corrosion and Stress Corrosion Crack Initiation of Cold-Worked Alloy 690 in PWR Primary Water", Technical Milestone Report, 2013

[3] J. Boursier, F. Vaillant and B. Yrieix, "Weldability, Thermal Aging and PWSCC Behavior of Nickel Weld Metals Containing 15 to 30% Chromium", ASME/JSME 2004 Pressure Vessels and Piping Conference, pp.109-121, 2004

[4] S. C. Yoo, K. J. Choi, T. Kim, S. H. Kim, J. Y. Kim and J. H. Kim, “Microstructural evolution and stress-corrosion-cracking behavior of thermally aged Ni-Cr-Fe alloy”, Corrosion Science, Vol. 111, pp.39-51, 2016

[5] D. Jones, “Localized Surface Plasticity During Stress Corrosion Cracking”, Corrosion, Vol. 52, pp. 356-362, 1996

[6] E. F. Westbrooke, L. E. Forero, and F. Ebrahimi, “Slip Analysis in a Ni-Base Superalloy”, Acta Materialia, Vol. 53, pp. 2137-2147, 2005

[7] D. M. Dimiduk, M. D. Uchic, and T. A. Parthasarathy, “Size-Affected Single-Slip Behavior of Pure Nickel Microcrystals”, Acta Materialia, Vol. 53, pp. 4065-4077, 2005