Effect of Dissolved Hydrogen Concentration on Irradiation Assisted Stress Corrosion Cracking Initiation Properties of Stainless Steel in Primary Water Condition

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1. Introduction

Austenitic stainless steel has been widely used in primary circuits of pressurized water reactor (PWR). Especially type 304 and 316 stainless steels are the major materials of the reactor internal structures supporting the reactor core. According to the screening criteria of the guidance for managing the effects of aging degradation in PWR internals, reactor internals composed of stainless steel have a great resistance to stress corrosion cracking (SCC) except welds [1]. However, when these materials exposed to neutron irradiation, they become susceptible to irradiation assisted stress corrosion cracking (IASCC) [2]. To investigate IASCC phenomenon, neutron-irradiated material should be used, however, it is hard to handle neutron-irradiated materials due to activation. Thus, generating similar effect of neutron-irradiation on test material is essential to investigate IASCC.

According to the several research, there is a clear relationship between the dissolved hydrogen (DH) concentration and SCC behavior of Ni-based alloy in PWR primary water condition [3]. However, the electrochemical potential of austenitic stainless steels is different with that of Ni-based alloy. So that, changing DH may aggravate the degradation of stainless steels. It was reported that DH concentration changes diffusion of iron ion which can affect oxide thickness [4], but the correlation between DH concentration and corrosion behavior of stainless steel has not been clarified so far. Therefore, it should be deeply studied prior to change DH condition of commercial nuclear power plant.

Thus, in this study, proton was chosen as an alternative irradiation source to make 1 and 3 dpa radiation damage on stainless steel 304L sample surface. Furthermore, to investigate combined effect of radiation damage and DH concentration on the IASCC initiation behavior, two different DH levels, 25 and 50 cm3/kg, were applied inside of experimental facility respectively. To detect the crack initiation moment precisely, slow strain rate test (SSRT) combined with direct current potential drop (DCPD) method was conducted. According to the results, the crack initiation time of sample whose radiation damage level is higher become faster than that of lower one. Furthermore, higher DH concentration, in other words lower oxygen partial pressure, makes material more susceptible to IASCC initiation.

2. Experimental

In this section, detailed explanation about the material, proton irradiation, stress corrosion cracking experimental methodology, and the water chemistry condition are described.

2.1 Material

Solution annealed stainless steel 304L mock-up block quenched by water was prepared for this research. Chemical composition of the material is listed below. The block was cut into pieces as the shape of SSRT sample. Prior to proton irradiation, each sample was polished with emery paper up to 800 grits and was polished with diamond suspension up to 1 μm. After the polishing process the samples were rinsed by in order of acetone, ethanol, and distilled water.

Table I: Chemical composition of 304L stainless steel used in this research [wt%]

Fe	C	Si	Mn	P	S	Cr
Bal.	0.024	0.45	1.43	0.033	0.003	18.35
Ni	**Mo**	**N**	**Co**	**Cu**	**Ti**	
8.11	0.18	0.044	0.17	0.28	0.002	

2.2 Proton irradiation

Structural material such as 304L stainless steel can be irradiated by neutron during normal operation period. But neutron irradiation is hard to handle from lab scale experiment. So that, there are several ways to simulate neutron irradiation effect on material. Irradiation using other sources such as proton and heavy ion is the most used method. In this work, proton irradiation was done from the Michigan Ion Beam Laboratory (MIBL) to figure out the effect of irradiation. The test condition was chosen following preceding research done by G. Was et al. and the target depth of irradiation was calculated by stopping and range of ions in materials (SRIM) [5].

Table II: Conditions for proton irradiation done my Michigan Ion Beam Laboratory

Beam energy [MeV]	2.0
Dose rate [dpa/s]	1*10-5
Displacement energy [eV]	40
Temperature [°C]	360
Radiation damage [dpa]	1, 3

2.3 Stress corrosion cracking experiment

SSRT is a common SCC test methodology which can figure out crack initiation time and crack density by stretching sample very slowly. In this work we focused on crack initiation time to check IASCC resistance of each sample. Strain rate was set as $1*10^{-7}$ mm/mm/s.

The experimental facility was filled with simulated primary water. Temperature and pressure were set as following real plant conditions, and chemical ion such as boron and lithium were put into the test solution. Dissolved oxygen (DO) concentration was controlled strictly by injecting hydrogen gas. DH concentration as set as 25 and 50 cm^3/kg. Below table shows the test conditions including SSRT and DCPD system conditions as well.

Table III: Stress corrosion cracking experimental environment including slow strain rate test and direct current potential drop conditions

Pressure [MPa]	**Temperature [℃]**	**Boron [ppm]**	**Lithium [ppm]**
15.5	325	1200	2.2
DO [ppb]	**DH [cm^3/kg]**	**Current [A]**	**Strain rate [mm/mm/s]**
< 5	25, 50	5	$1*10^{-7}$

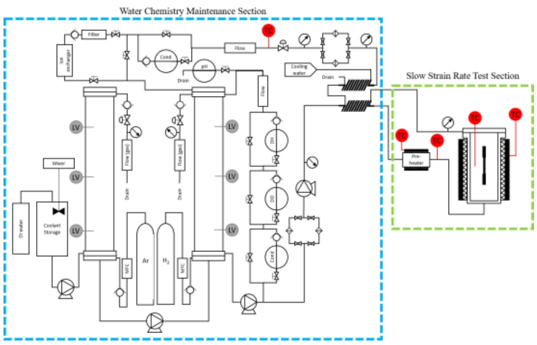

Fig. 1. Schematic diagram of experimental facility which consists of water chemistry maintanence section and stress corrosion cracking experimental section

3. Results

Considering the sample dimension especially gauge section, the electrical resistance of sample will be increased uniformly. But this tendency is going to be changed when crack initiated. The micro crack can reduce cross sectional area of sample which causes rapid increase of electrical resistance, in other words potential drop increase since the current maintains as 5 A during experiment. Below plot (left) is an example of DCPD data (black colored dot) with fitting curve (red colored line). As it shows, the DCPD data fits perfectly with the fitting curve on the stage 1. And the DCPD data starts to fluctuate but it follows the fitting curve well on the stage 2, which represents plastic deformation with crack initiation. And finally, the data escapes from the fitting curve on the stage 3, which means the occurrence of necking. By this method, the crack initiation time was decided as the starting point of the stage 2.

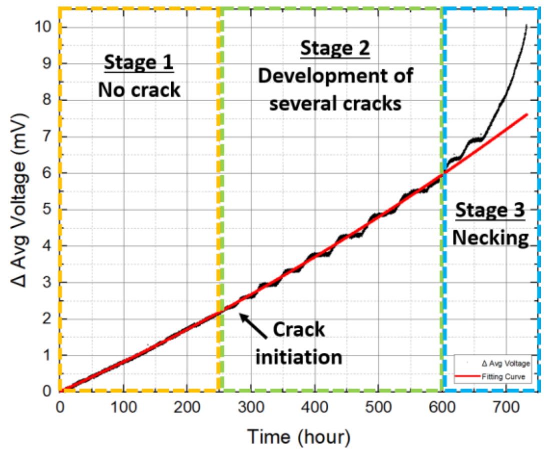

Fig. 2. Detection method of crack initiating moment by interpreting direct current potential drop data

To make the set of control group, as-fabricated samples were tested with two different DH concentration environments. The average crack initiation time of as-fabricated samples with DH concentration 25 cm^3/kg is about 326 hours and that of DH 50 experiment is about 259 hours. Comparing these results, DH decreases crack initiation time about 21 % in case of as-fabricated sample. To figure out the effect of radiation damage on crack initiation, DH 25 experiments using as-fabricated, 1 dpa irradiated, and 3 dpa irradiated sample had been conducted. According to the results, the crack initiation time of 1 dpa irradiated sample is about 205 hours and that of 3 dpa irradiated sample is 170 hours respectively. The reduction rate of crack initiation time for 1 and 3 dpa samples compared to as-fabricated sample are 37 % and 48 % respectively. So that, radiation damage decreases crack initiation time dramatically compared to the effect of DH concentration. The combined effect between DH concentration and radiation damage can be figured out with comparing the result of as-fabricated sample in DH 25 environment to the result of 3 dpa irradiated sample in DH 50 condition. The crack initiation time of 3 dpa irradiated sample is about 140 hours, in other words, the time decreases about 57 %. This reduction rate is larger than the difference between as-fabricated sample and 3 dpa irradiated sample in DH 25 condition. Therefore, it can be noted that DH concentration and radiation damage have synergetic effect on crack initiation behavior of the material.

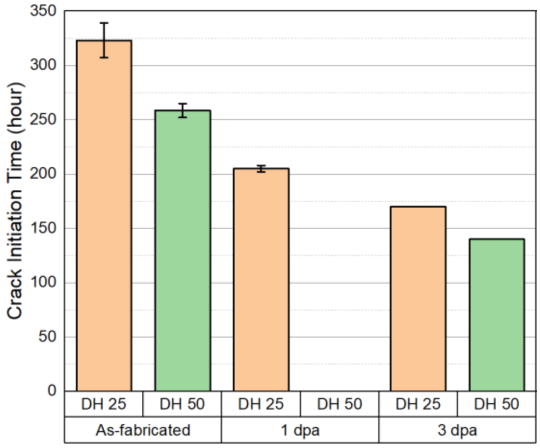

Fig. 3. Summary of crack initiation time measurement with differing the amount of radiation damage and dissolved hydrogen concentration

3. Conclusions

In this research, the effect of dissolved hydrogen concentration changes on irradiation assisted stress corrosion cracking behavior of stainless steels in primary water environment was studied by slow strain rate test combined with direct current potential drop method using as-fabricated, 1 dpa irradiated, and 3 dpa irradiated samples. According to the results from the sets of as-fabricated samples with differing dissolved hydrogen concentration, the crack initiation time decreases with increasing dissolved hydrogen concentration. The crack initiation time from the sets of same dissolved hydrogen concentration, 25 cm^3/kg, the time decreases with increasing the amount of radiation damage. Comparing those reduction rate of crack initiation time, the effect of radiation damage is greater than that of dissolved hydrogen concentration. The combined effect of both changes was investigated using 3 dpa irradiated sample with 50 cm^3/kg dissolved hydrogen condition, and the synergetic effect was figured out.

ACKNOWLEDGEMENT

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