Transactions of the Korean Nuclear Society Virtual Autumn Meeting
October 21-22, 2021

Dissolved Hydrogen Concentration and Proton Irradiation Effect on Crack Initiation Behavior of 304L Stainless Steel

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1. Introduction

Austenitic stainless steel has been widely used in primary circuits of pressurized water reactor. Especially type 304 and 316 stainless steels are the main materials of the reactor internal structures supporting the reactor core. These kinds of structural components are located close to the nuclear fuel of reactor and are irradiated by neutron which causes severe problem irradiation assisted stress corrosion cracking (IASCC) [1]. IASCC, a special case of intergranular stress corrosion cracking, referred as intergranular cracking showing little or no ductility, which can occur in heavily irradiated structural components of nuclear reactor cores and/or under irradiation. However, it is hard to handle neutron-irradiated materials due to activation, thus, simulation of neutron radiation effect is essential to investigate IASCC.

There are several irradiation effects on material degradation which can affect IASCC susceptibility, such as hardening, radiation induced segregation, and dislocation. To simulate abovementioned irradiation effect without using neutron, various attempts have been made so far; heat treatment, warm/cold working, and irradiation with other sources. However, these cannot simulate the exact state of neutron-irradiated material. Thus, research to identify IASCC have been done limitedly and further studies should be conducted.

Most of SCC studies have used U-bend specimen in various environment. But U-bend test has a limitation related to its long experimental period. To overcome this weakness, slow strain rate test (SSRT) or constant elongation rate test (CERT) are used which are a kind of acceleration test observing fracture surface. However, in point of view of IASCC, the effect of irradiation is concentrated in the shallow irradiated surface. Thus, it is hard to investigate the effect of irradiation using fracture surface observation method when it comes to deal with crack initiation.

In this study, IASCC susceptibility of 304L stainless steel was investigated by conducting SSRT experiments combining with direct current potential drop (DCPD) methodology to detect the exact moment of crack initiation using sensitized and 1 dpa proton irradiated samples respectively. Also, dissolved hydrogen concentration effect on IASCC resistance of 304L stainless steel was considered as well.

2. Experimental

This section describes the experimental setup for slow strain rate test (SSRT) using sensitized and proton irradiated samples. The method of proton irradiation and sensitization are also described.

2.1 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. Irradiating using other sources such as proton and heavy ion is the most used method that have been conducted these days. In this work, proton irradiation was done to figure out the effect of irradiation. Irradiation condition was chosen following preceding research done by G. Was et al. [2] and the target depth of irradiation was calculated by stopping and range of ions in materials (SRIM) [3]. Detailed test conditions and the result of SRIM calculation are inserted below.

Table I: Proton irradiation condition

Beam energy [MeV]	2.0
Dose rate [dpa/s]	$1*10^{-5}$
Displacement energy [eV]	40
Temperature [℃]	360
Radiation damage [dpa]	1

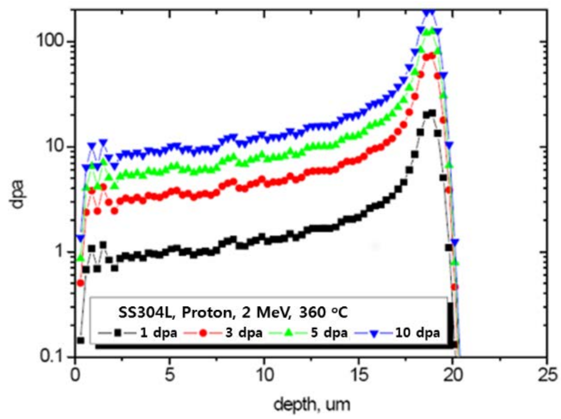

Fig. 1. SRIM calculation for proton irradiation

2.2 Sensitization

To simulate radiation induced segregation effect on material, sensitizing heat treatment was done. Considering carbon content of prepared 304L stainless steel, according to time-temperature transformation diagram of 304L stainless steel, heat treatment under 550 ℃during 200 hours is enough to form carbide along the grain boundary which can simulate irradiation effect. Below table shows the chemical composition of the material that was used for each experiment.

Table II: Chemical composition of 304L stainless steel [wt%]

C	Si	Mn	P	S	Cr
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

2.3 Slow strain rate test & Direct current potential drop

SSRT is a common SCC test methodology which can figure out crack initiation time or crack density of sample during or after stretching the sample very slowly. In this work we focused on crack initiation time to check irradiation assisted stress corrosion cracking (IASCC) resistance of each sample. Strain rate was set as $1*10^{-7}$ mm/mm/s following ASTM G129 [4].

Suppose that sample is going to be elongated continuously without any volumetric change, the electrical resistance of sample will be increased uniformly. However, when some crack occurs on the sample surface the electrical resistance will be changed shortly. So that, detecting this moment by measuring potential drop through the sample gauge section, crack initiation time of sample can be measured. This kind of method is called direct current potential drop (DCPD). Below figure shows that the sample engaged into the test facility and platinum wires; the first set which is welded at both edge side supplies direct current about 5 A uniformly, and the second set which is welded at the middle side measuring potential drop through the gauge section of the sample

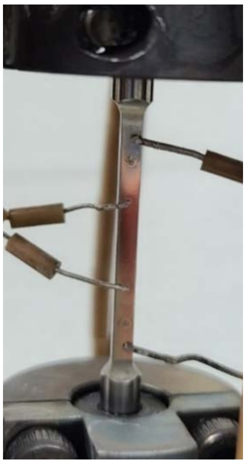

Fig. 2. Platinum probe which supplies direct current and measures potential drop

2.4 Primary water environment

The experimental facility was filled with simulated primary water [5]. 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 was controlled strictly by injecting argon and hydrogen gas. To investigate dissolved hydrogen (DH) effect on IASCC, two different DH concentrations were adapted. Below table shows the test conditions that we developed in this study.

Table III: Experimental conditions

Pressure [MPa]	Temperature [℃]	B [ppm]	Li [ppm]	DO [ppb]	DH [cm^3/kg]
15.5	325	1200	2.2	< 5	25 50

2.5 Test matrix

Two different types of samples were used to investigate irradiation effect on IASCC initiation behavior. The first one is sensitized 304L stainless steel, and the second one is 1 dpa proton irradiated 304L stainless steel. And two different DH concentration were applied to investigate dissolved hydrogen effect. The detailed test matrix is listed in the below table.

Table IV: Test matrix

Sample type	DH [cm^3/kg]	Notation
Sensitized	25	2S
Sensitized	50	5S
Irradiated (1 dpa)	25	21

3. Result

3.1 Loop data

Experiments had been conducted up to 450 hours which was the longest case. During each experiment, loop data such as pressure and temperature inside of test section had been measured and conductivity of test solution had been monitored as well. As below figure shows, test environment was uniformly maintained during the experiment.

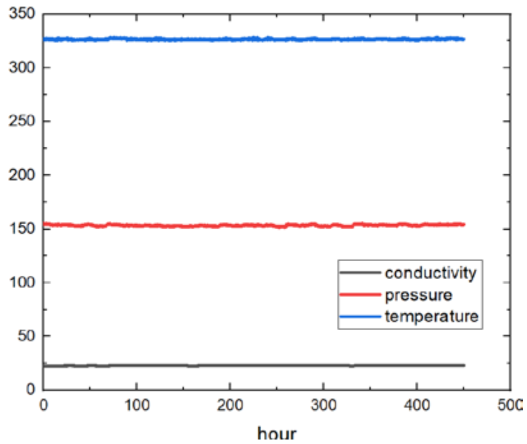

Fig. 3. Loop data during the experiment which had been conducted about 450 hours

3.2 Dissolved hydrogen effect (2S vs 5S)

Considering the sample dimension especially gauge section, the electrical resistance of sample will be increased uniformly by a quadratic function. But this tendency is going to be changed when crack initiated. The micro crack can decrease cross sectional area of sample slightly which causes rapid increase of electrical resistance, in other words potential drop since current maintains as 5 A during every moment. Below plot is DCPD data of one of 2S experiments (black colored line) and second order polynomial fitting curve (red colored line). As it shows, the fitting curve fits almost 99% with measured DCPD data until crack occurs; approximately 320 hours elapsed. After that, the DCPD data fluctuates compared to fitting curve. That means crack increases potential drop and the crack is filled with oxide that can decrease the electrical resistance by increasing cross sectional area.

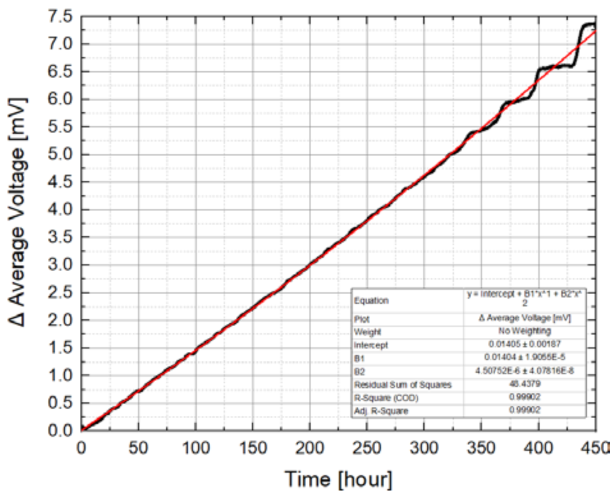

Fig. 4. DCPD data with second order polynomial fitting curve for 2S experiment

To point out the exact crack initiation time from the data, normalizing process was done between measured data and fitting curve. According to the below graph, the signal rises over 1 % near 370 hours elapsed. And this trend becomes greater. So that it can be noted that crack was initiated and grew continuously. From the cross sectional microstructure observation, after normalized voltage reached over 1 %, approximately 10 μ m depth crack was observed, therefore, we decided crack initiation time as when the normalized voltage of each experiment peaked over 1 %.

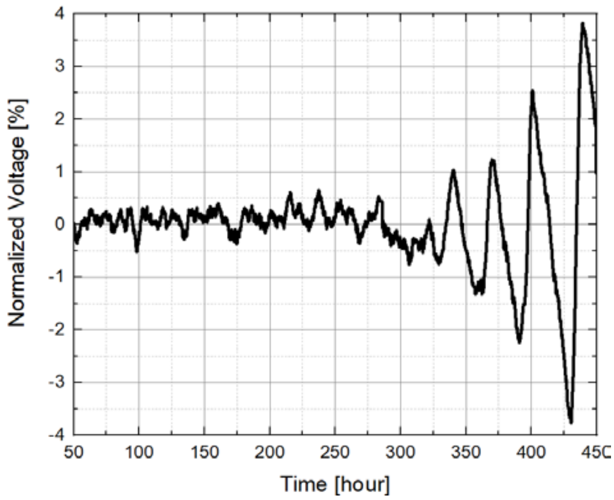

Fig. 5. Normalized DCPD data for 2S experiment

SSRT experiments using 50 cm^3/kg concentration of DH were done following the same manner with 2S experiments. From the DCPD results, the data followed quadratic function well and it fluctuated after 230 hours. According to the normalized voltage plot, the voltage goes over 1 % near 260 hours.

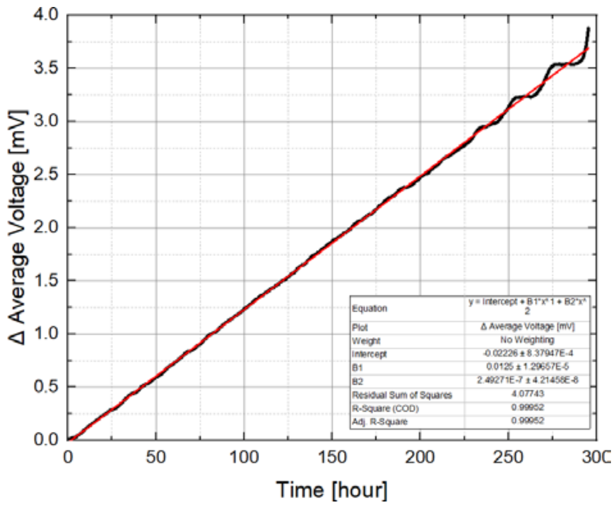

Fig. 6. DCPD data with second order polynomial fitting curve for 5S experiment

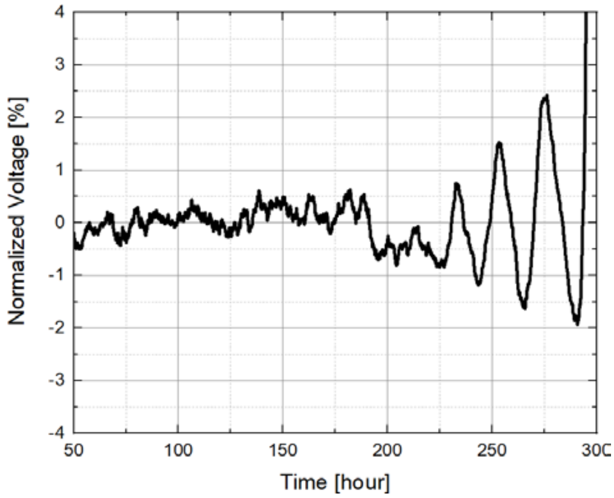

Fig. 7. Normalized DCPD data for 5S experiment

Gathering data from several repeated experiments 2S and 5S respectively, crack initiation time affected by dissolved hydrogen concentration can be summarized as below chart. When the DH concentration set as 25 cm^3/kg the average crack initiation time was about 359 hours and that of 50 cm^3/kg DH was 267 hours. In conclusion when DH concentration increases the crack initiation time was shortened.

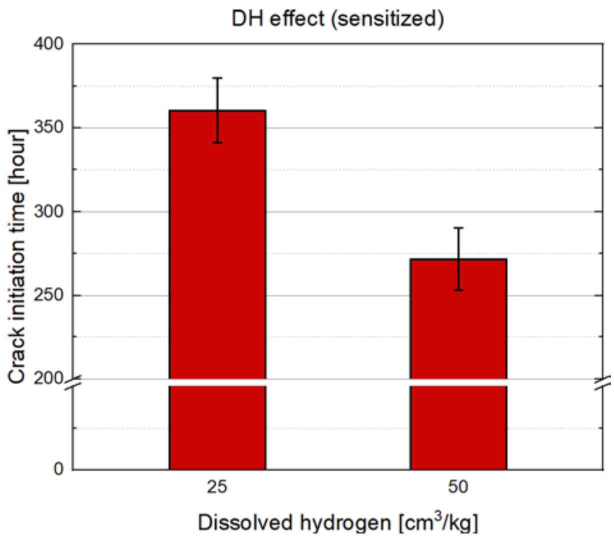

Fig. 8. Dissolved hydrogen effect on crack initiation time

3.3 Irradiation effect (2S vs 21)

SSRT experiments using 1 dpa irradiated sample with 25 cm^3/kg DH concentration were conducted. The DCPD data fits perfectly with second order polynomial fitting curve as well. Although there is a noise near 230 hours, but the trend seems not to be affected by the noise. So that it can be also noted that the crack initiation time of 1 dpa proton irradiated sample was about 250 hours for the given case.

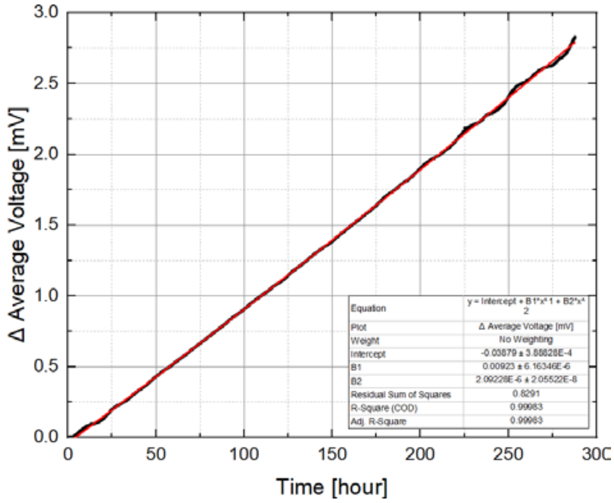

Fig. 9. DCPD data with second order polynomial fitting curve for 21 experiment

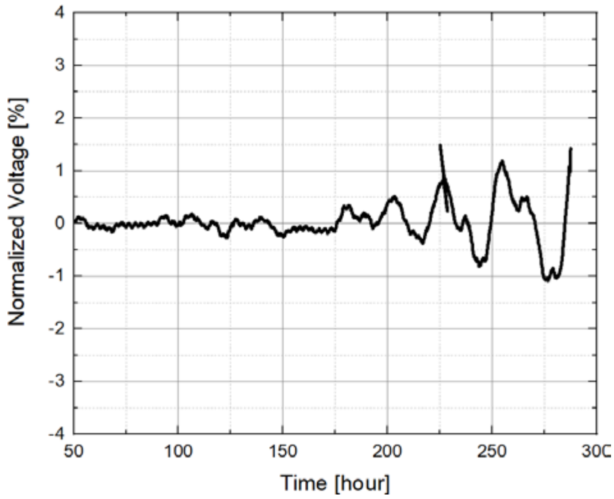

Fig. 10. Normalized DCPD data for 21 experiment

The summarized chart shows the effect of irradiation on IASCC susceptibility. Despite DH concentration in both cases are same, the crack initiation time decreases from 359 hours to 254 hours. The reproducibility of this results was checked by repeating 21 experiment. Thus, it can be said that proton irradiation worsens IASCC resistance of 304L stainless steel.

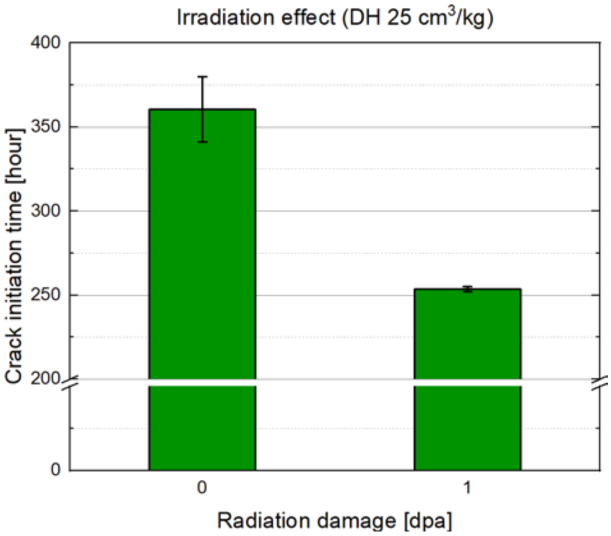

Fig. 11. Irradiation effect on crack initiation time

4. Conclusion

To figure out dissolved hydrogen concentration effect and irradiation effect on irradiation assisted stress corrosion cracking resistance of 304L stainless steel, experiments combining with direct current potential drop methodology were conducted. Each experiment was done in the simulated primary water chemistry condition changing sample type; sensitized and 1 dpa proton irradiated and changing dissolved hydrogen concentration; 25 cm^3/kg and 50 cm^3/kg respectively.

Comparing between measured potential drop and second order polynomial fitting curve, every case fits well with theoretical approach, and they show fluctuated potential drop after certain elapsed hours as well. To find the exact moment when crack initiated, normalizing process was done, and this caught that crack initiated when the normalized voltage peaked over 1 % by empirically.

When dissolved hydrogen concentration increases from 25 cm^3/kg to 50 cm^3/kg, crack initiated faster than before; 359 hours to 267 hours. In the same manner, irradiation effect on crack initiation time was also investigated and it can be noted that irradiation effect seems more dominant than that of dissolved hydrogen concentration; 359 hours to 254 hours.

ACKNOWLEDGEMENT

This work was financially supported by a Human Resources Development of the Korea Institute of Energy Technology Evaluation and Planning grant (No. 20194030202400) funded by the Korea Government Ministry of Trade Industry and Energy, and Nuclear Safety Research Program through the Korea Foundation Of Nuclear Safety (KoFONS) using the financial resource granted by the Nuclear Safety and Security Commission (NSSC) of the Republic of Korea. (No. 2106022).

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