CRACKING OF NEUTRON IRRADIATED AUSTENITIC STAINLESS STEELS IN LIGHT WATER REACTOR ENVIRONMENTS

Y. Chen, and B. Alexandreanu
Argonne National Laboratory
Lemont, IL, USA

Email contact of corresponding author: Yiren_chen@anl.gov

ABSTRACT

The performance of austenitic stainless steels (SS) used for reactor core components is critical for the safe and economic operation of light water reactors (LWRs). Exposed to corrosive coolant and neutron irradiation simultaneously during service, SS in the LWR core can undergo significant microstructural and microchemical changes. To evaluate the cracking behavior of reactor internal materials, an experimental study was performed on neutron-irradiated SS samples. Slow strain rate tests were performed in simulated LWR coolants with different levels of dissolved oxygen (DO). The crack growth rates (CGRs) of various SSs were also measured in LWR environment at different doses. It was found that, at low doses, the cracking susceptibility of irradiated SS depended strongly on the DO level in LWR coolant. At high doses however, the effect of DO was not so clear. High CGRs were observed on the highly irradiated samples even in hydrogen water chemistry.

INTRODUCTION

Stress corrosion cracking (SCC) is a degradation mechanism involving the synergistic interactions of environment, microstructure, and stress, and is a critical issue for stainless steels (SS) and nickel alloys used in aqueous environment. Irradiation-assisted SCC (IASCC) is a special form of SCC induced or accelerated by irradiation. Subject to neutron irradiation, the microstructure and service environment of reactor materials can undergo significant changes, giving rise to elevated cracking susceptibility and deteriorated service performance. Premature failures resulting from IASCC have been reported in a wide range of reactors core internal components [1][2]. With the aging of light water reactors (LWRs) worldwide, IASCC has become an increasingly important issue and could seriously impair the long-term operation of LWRs if not addressed adequately.

Irradiation plays a key role in IASCC, and SS can become vulnerable to SCC after extended neutron exposure. A sharp rise in cracking susceptibility has been observed above 0.75 dpa in BWRs [3], and 3 dpa in PWRs [4]. Because of the complex nature of multi-variable dependence, a true dose threshold may not exist for IASCC. In a simulated LWR environment with high dissolved oxygen (DO), cracking has been observed as low as 0.3 dpa [5]. Nonetheless, exposure to irradiation is a necessary condition for IASCC.

In addition to irradiation, IASCC is also sensitive to local metallurgical conditions. Early mechanistic studies of IASCC focused mainly on the grain boundary depletion of Cr resulting from radiation-induced segregation (RIS), and its influence on the passivation process at grain boundaries [6]. The roles of impurity elements such as Si, P, and S on IASCC have also been investigated [7]. While the redistribution of impurities does occur under irradiation, a clear dependence of IASCC susceptibility on the bulk impurity content has yet emerged. More recently, the connection between the deformation mode and IASCC has attracted considerable attention. The development of localized plastic flow in irradiated materials can be linked to IASCC [8], suggesting an underlying mechanism associated with irradiation embrittlement.

Furthermore, as a special form of SCC, IASCC depends strongly on the corrosion potential between –100 and 0 mV with respect to a standard hydrogen electrode (SHE), a low potential in BWR hydrogen water chemistry (HWC) or PWR primary water chemistry is considered beneficial to mitigate SCC [9]. However, low corrosion potential does not provide complete immunity to IASCC. Intergranular cracking has also been observed in cold-worked and irradiated SS baffle bolts in PWRs [10].

In this study, the cracking behavior of neutron-irradiated SS samples were studied with slow strain rate tensile (SSRT) and crack growth rate (CGR) tests in simulated LWR environments. The effect of water chemistry on the cracking susceptibility was evaluated at different doses.

EXPERIMENTAL

As shown in Fig. 1a, the SSRT specimen used in this study was a flat tensile sample with a gauge dimension of 7.6 x 1.5 x 0.8 mm. The overall length of the sample was about 25 mm. Several heats of wrought SS (304, 316, 347) and cast SS (CF-3 and CF-8) were included. Both cold-worked (CW) and solution-annealed (SA) samples were also included in the study. The SSRT samples were irradiated in the BOR-60 reactor at ~320°C to 5, 10 and ~47 dpa. After irradiation, the samples were tested in simulated BWR normal water chemistry (NWC) or PWR primary water chemistry with a nominal strain rate of $7x10^{-7}$ s^{-1}. During the tests, water was circulating in the autoclave at a rate of 10-20 ml/min. After the tests, fractographic examination was performed on the fractured samples to assess their cracking susceptibility.

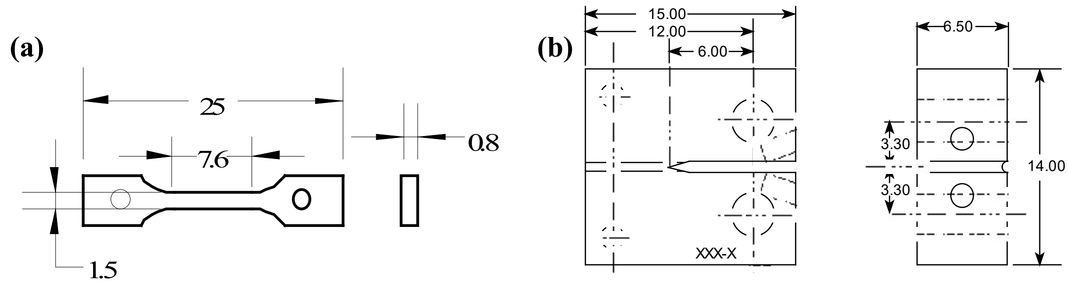

Fig. 1. SSRT (a) and CT (b) specimens used in this study (dimensions in mm).

CGR tests were conducted with small compact tension (CT) specimens as shown in Fig. 1b. The sample had a nominal thickness of 6.5 mm, and was side grooved to ~10% of its thickness. The samples were various wrought, cast or weld 304 and 316 SS irradiated in the Halden reactor to various doses up to 3 dpa. After irradiation, the samples were tested in BWR NWC, or BWR hydrogen water chemistry (HWC), or PWR water. While BWR NWC contained high DO (>300 ppb), both BWR HWC and PWR water contained less than 20 ppb DO and were low-corrosion-potential environments beneficial for SCC resistance [9]. During the tests, water was also circulating in the system to refresh the autoclave volume once every 30-45 minutes. The CGR tests were conducted with two servo-hydraulic test systems inside "hot" cells. Each CGR test was started with cyclic loading with a triangle waveform and a low load ratio (i.e., min load/max load) around 0.2 or 0.3. Once the sample was pre-cracked, the test would be transitioned to a SCC test with progressively increased rise time and load ratio. The SCC CGRs were then evaluated at several stress intensity factors (Ks), with or without periodical partial unloading (PPU).

RESULTS AND DISCUSSION

A total of 31 SSRT tests were performed on the irradiated samples. Most of the tests at ~5 dpa were performed in BWR NWC, and the rest tests were conducted in PWR water. Overall, a significant effect of irradiation hardening was observed with these tests regardless their test environments. Fig. 2 shows the yield strength (YS) and total elongation (TE) as a function of irradiation dose. To illustrate the behavior at lower doses, the SSRT results from a previous study [11] using samples irradiated in the Halden reactor was also included in the figure. Note that the gauge section of the Halden samples were longer and wider than that of the BOR-60 samples, making their results not directly comparable. Nonetheless, an increase trend of YS and a decrease trend of TE can be seen with the dose in the figure. The saturation of irradiation hardening appears to be above 3 dpa. Above ~10 dpa, the changes in YS and TE reduced considerably.

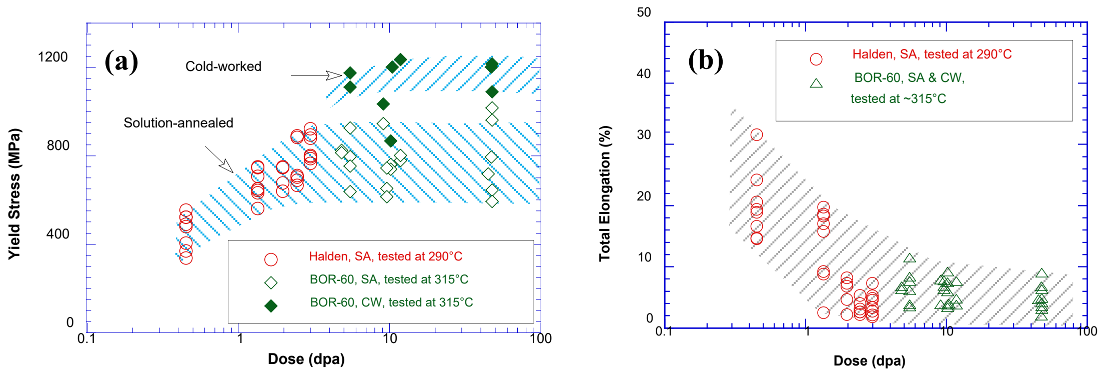

Fig. 2. SSRT yield strength (a) and total elongation (b) for specimens tested in simulated LWR water

For the tests performed in BWR NWC, extensive intergranular (IG) cracking was observed in several samples with elevated oxygen and sulfur contents. Similar IG cracking was also observed in the study using the Halden samples in BWR NWC [11]. In contrast, the samples tested in PWR water showed mainly a ductile dimple fracture. An example of the different fracture morphology is shown in Fig. 3. While large IG cracking areas can be seen on the fracture surface of a high-purity 304L SS tested in BWR NWC (Fig. 3a), the same heat exhibited little brittle features when tested in PWR water (Fig. 3b), despite being irradiated to a higher dose. Evidently, the overall fraction of brittle fracture was lower in PWR water than in BWR NWC. This lower cracking susceptibility in PWR water can be inferred from the effect of corrosion potential. It has been shown that the crack tip dissolution rate can vary considerably with the corrosion potential in oxygenated water [9]. By lowering the DO level, and therefore the corrosion potential of a material-environment system, the oxidization rate is reduced significantly. However, a low corrosion potential does not provide a complete immunity to IASCC. In this study, two highly irradiated samples at ~47 dpa did show IG cracking when tested in PWR water. Nonethless, the cracking suspectibility of these irradiated SS was generally lower in PWR water than in BWR NWC.

Different cracking responses in BWR NWC and HWC were also observed in the CGR tests. Fig. 4 shows the results from some samples irradiated in the Halden reactor. The open and closed symbols represent the tests conducted in NWC and HWC, respectively. In Fig. 4a, the measured CGRs were plotted against the anticipated fatigue crack growth rate in air under the same loading condition. As expected, the cyclic CGRs measured in the test environments were mostly higher than the fatigue growth rates in air, especially in the low-CGR region. With the decrease of the fatigue growth rate (moving from the top right to lower left of the figure), the extent of environmental enhancement becomes more evident. While the two doses (1.35 and 3 dpa) did not affect the cyclic CGRs significantly, the different test environments did lead to different cyclic responses. The cyclic CGRs obtained in NWC were much higher than that obtained in HWC, suggesting a beneficial effect of HWC to reduce the degree of environmental enhancement under cyclic cracking.

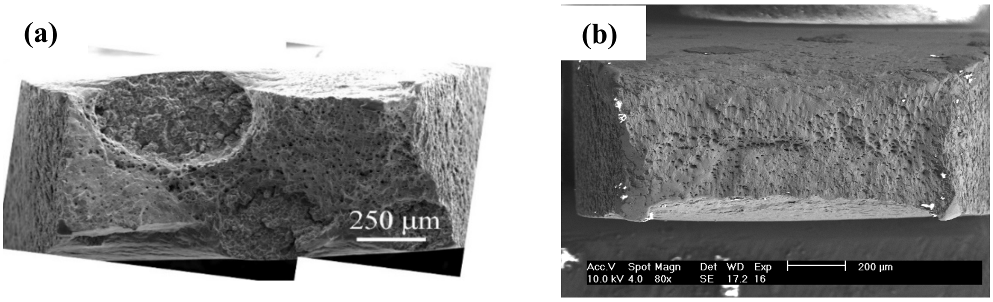

Fig. 3. Fracture surfaces of a high-purity Type 304L SS tested in (a) BWR NWC (5 dpa), and (b) PWR water (10 dpa).

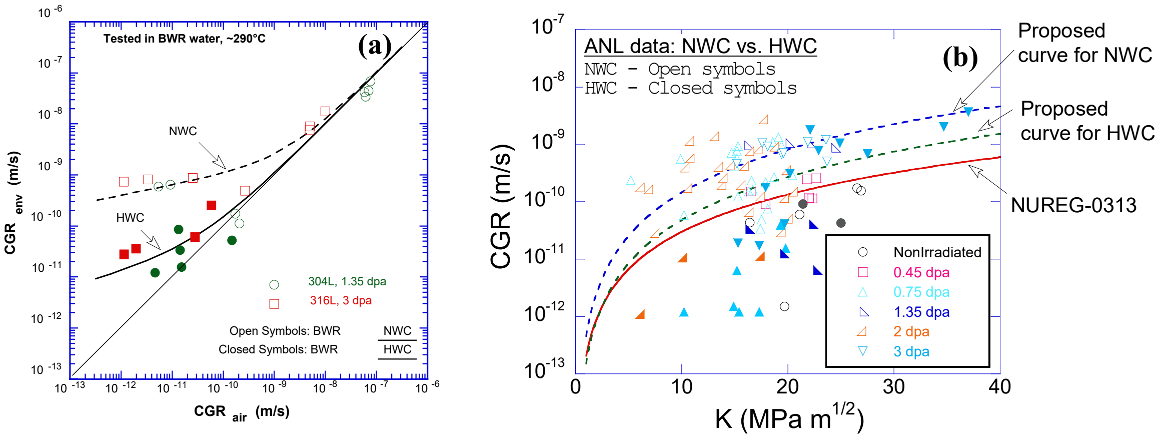

Fig. 4. (a) Cyclic CGRs, and (b) SCC CGRs of irradiated SS in BWR NWC and HWC environments.

A similar effect of HWC can also be seen in the SCC CGR results. In Fig. 4b, the measured SCC CGRs were plotted as a function of applied K. Again, the open and closed symbols represent NWC and HWC, respectively. The disposition curves provided in References [12] and [13] were also included in the figure. While the SCC CGR data scatter significantly, they should generally follow a power-law relationship with the increasing K according to the slip-dissolution model [9]. As shown in Fig. 4b, the SCC CGRs obtained in NWC were much higher than those obtained in HWC at the low-K region. The irradiation dose did not play a significant role, and all CGR data clustered into two separated regions, above and below the NUREG-0313 line. Above ~20-30 MPa $m^{1/2}$ however, some CGRs in HWC measured on the highly irradiated samples (~3 dpa) increased rapidly with the increasing K. The CGRs measured in HWC became similar to those obtained in NWC. Evidently, the effect of HWC in mitigating SCC vanished when the applied K and the irradiation dose were high. This is consistent with that observed in the SSRT tests where IG cracking can still prevail with high-dose samples in low-corrosion potential environments.

CONCLUSION

SSRT and CGR tests were performed on irradiated samples in simulated LWR environments to study the effect of water chemistry on the IASCC of SS. While extensive IG cracking can be seen in the SSRT tests conducted in BWR NWC, brittle morphology resulting from the tests in PWR water was much less, suggesting a beneficial effect of low-DO environment in suppressing IASCC. However, the low-DO environments did not guarantee a complete immunity to IASCC, and some IG cracking were indeed observed in highly irradiated samples tested in PWR water. Similarly, the effect of the test environments can be seen in the CGR tests. Under the cyclic loading mode, the extent of environmental enhancement was lower in BWR HWC than in BWR NWC. Under the constant-load test mode, the SCC CGRs measured in BWR HWC and PWR water are also much lower than those measured in BWR NWC. However, the beneficial effect of the low-DO environment could be diminished considerably with the increasing applied K for high-dose samples. High CGRs were observed for the highly irradiated samples even in PWR water or BWR NWC.

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