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Crack initiation behavior of neutron irradiated model and commercial stainless steels in high temperature water

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HIGHLIGHTS

- Environmental constant extension rate tensile tests were performed on neutron irradiated steel.
- Percentage of intergranular cracking quantified the cracking susceptibility.
- Cracking susceptibility varied with test environment, solute addition, and cold work.
- No singular microstructural change could explain increases in cracking susceptibility with irradiation dose.
- The increment of yield strength due to irradiation correlated well with cracking susceptibility.

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ABSTRACT

The objective of this study was to isolate key factors affecting the irradiation-assisted stress corrosion cracking (IASCC) susceptibility of eleven neutron-irradiated austenitic stainless steel alloys. Four commercial purity and seven high purity stainless steels were fabricated with specific changes in composition and microstructure, and irradiated in a fast reactor spectrum at 320 °C to doses between 4.4 and 47.5 dpa. Constant extension rate tensile (CERT) tests were performed in normal water chemistry (NWC), hydrogen water chemistry (HWC), or primary water (PW) environments to isolate the effects of environment, elemental solute addition, alloy purity, alloy heat, alloy type, cold work, and irradiation dose. The irradiated alloys showed a wide variation in IASCC susceptibility, as measured by the relative changes in mechanical properties and crack morphology. Cracking susceptibility measured by %IG was enhanced in oxidizing environments, although testing in the lowest potential environment caused an increase in surface crack density. Alloys containing solute addition of Ni or Ni + Cr exhibited no IASCC. Susceptibility was reduced in materials cold worked prior to irradiation, and increased with increasing irradiation dose. Irradiation-induced hardening was accounted for by the dislocation loop microstructure, however no relation between crack initiation and radiation hardening was found.

1. Introduction

Irradiation-assisted stress corrosion cracking (IASCC) is a complex, multivariable issue that is a critical problem for austenitic stainless steels used in light water reactor (LWR) core internal components. Observations of IASCC have been made both in service and in laboratory settings [1], and are consistent in showing that IASCC is due to persistent changes to the alloy caused by irradiation. This degradation mode is one of the highest priority issues affecting the longevity of existing nuclear reactors. Twenty-year life extensions have been granted to numerous plants and the consideration of further extensions is likely to depend on progress understanding the mechanism of IASCC and the development of mitigation strategies.

Similar to stress corrosion cracking, IASCC is caused by the combination of an applied stress, a corrosive environment, and a susceptible material; however, irradiation adds another element that alters both the material and environment. The difficulty in understanding the mechanism of IASCC in stainless steel stems from the fact that irradiation causes simultaneous microstructural changes (dislocation loop, precipitate, and void formation) and microchemical changes (radiation induced segregation (RIS) of solute elements). These micro-scale changes lead to changes in mechanical properties (radiation hardening), deformation mechanisms (dislocation channeling), and enhanced susceptibility to intergranular stress corrosion cracking (IGSCC).

This study takes advantage of an opportunity to gain significant insight into the IASCC problem by leveraging prior investment by the CIR (Cooperative IASCC Research) I and II programs [2–9]. In these programs, four commercial purity austenitic stainless steels were selected for irradiation along with ten high purity solute addition alloys created from the same base material, each with

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specific solute element additions. Irradiations were conducted in the BOR-60 liquid sodium fast reactor to doses ranging from 4.4 to 47.5 dpa. The selection of the alloys in that program provided an opportunity to assess of the effects of elemental solute additions, alloy purity, alloy heat, alloy type, cold work, and irradiation dose on IASCC. Constant extension rate tensile (CERT) tests performed on these materials were used to assess susceptibility to IASCC crack initiation and compare the effects of each aforementioned variable.

2. Experiment

2.1. Materials

A total of eleven different stainless steel (SS) alloys were analyzed in this study. Four of these alloys were commercial purity (CP), and seven were high purity solute addition (HP) alloys, each made from a single heat of model type 304L stainless steel with single elemental additions.

The four CP steels had designations A, B, C, and SW. Alloy A was type 304L SS from a LWR core shroud, alloy B was type 316L SS from a PWR baffle bolt, alloy C was type 316L + Ti baffle bolt material, and alloy SW was a Swedish heat of type 304L SS. In the alloy designation scheme, the second letter in the sample ID corresponds to the starting state; S indicates a solution annealed microstructure created by treatment at 1050 °C for 0.5 h followed by a water quench, and R indicates a cold worked (CW) starting microstructure. Several samples of alloy B were designated with no second letter (three digits only), and were in the cold worked condition identical to BR. The amount of added cold work was not reported for commercial steels [8]. Alloy SW was in a solution annealed state, although pre-irradiation processing of this alloy was unknown. Alloys AS, BS, BR or B, CR, and SW were available for testing. Alloy BS was reported to be cold worked to an even higher degree than alloy BR or B, despite the intent of a solution annealed microstructure [6]. Chemical compositions of CP alloys are given in Table 1.

The seven HP solute addition alloys had designations E, F, G, H, K, L, and P. Alloys G, H, K, and L were created from a single heat of model type 304L SS designated alloy E, which served as the reference condition. Alloys G, H, K and L were re-melted for the purpose of adding solute elements Mo, Si, Ni, and Ni + Cr, respectively. Alloy F was created from a separate starting material to produce a low C content. Alloy P was created from a separate type 316L SS base alloy with the oversize solute element Hf added. Chemical compositions of solute addition alloys are also given in Table 1, with each solute addition element indicated in bold. As-cast billets of each solute addition alloy were cold rolled from ~17 mm to ~8 mm, and ~8 mm to ~4 mm, applying approximately 13% reduction on each pass through the rolling mill. Solution annealing treatments between 900 °C and 1200 °C for 0.5 to 1 h were applied after each ~50% reduction to achieve a final ASTM grain size between 6 and 8 for each condition. Details of the cold work and solution annealing were presented by Arey et al. [8]. All solute addition alloy IDs contain the second letter S, indicating a solution annealed starting microstructure.

2.2. Sample fabrication, irradiation, and post-irradiation sample preparation

Dog bone type tensile bar samples were created from the alloys with the geometry shown in Fig. 1. Each sample had a 2 mm diameter cylindrical gage section with gage length of 12 mm and a pin hole size of 3.1 mm. Several of the tensile bar samples had a reduced pin hole diameter of 1.5 mm. This modification was inconsequential to CERT experiment results because all samples were shoulder loaded to avoid premature IASCC adjacent to the pin hole. Samples were fabricated at Pacific Northwest National Laboratory (PNNL) and shipped to Dimitrovgrad, Russia, where irradiation was performed in the BOR-60 reactor.

Irradiations in the BOR-60 liquid sodium fast reactor were completed between 2001 and 2004. The irradiation temperature was 320 °C. Neutron flux was $\sim 1.8 \times 10^{15}$ n cm^{-2} s^{-1} ($E > 0.1$ MeV), corresponding to a damage rate of 9.4×10^{-7} dpa s^{-1} ($E > 0.1$ MeV) calculated using the Norgett, Torrens and Robinson (NRT) model [6]. Total irradiation doses were between 4.4 and 47.5 dpa, Table 2. After irradiation, samples were shipped to Oak Ridge National Laboratory (ORNL) for post-irradiation sample preparation.

Each sample was both mechanically and electrochemically polished at the ORNL Low Activity Materials Development and Analysis (LAMDA) Laboratory. Mechanical polishing was performed manually using 320 grit SiC sandpaper to remove sodium and surface contaminants left by the reactor coolant during irradiation. Each sample was then electropolished using the A2 solution supplied by Struers™ (percentages by volume: 73% ethanol, 10% ethylene glycol monobutyl ether, 9% distilled water, and 8% perchloric acid), cooled to below 20 °C in an ice-bath. A potential of 30 V was applied for four 15 s periods per sample to achieve a mirror finish.

After the final polish, ~1 mm thick slices were removed from the heads of each tensile bar with a low speed diamond saw for Vickers hardness testing with a 500 g applied load. The finished tensile bars were then shipped from ORNL to the University of Michigan (UM) for testing.

Prior to testing, sample surfaces were imaged using a JEOL JSM-6480 scanning electron microscope (SEM) to characterize any surface defects with the potential to affect crack formation. Only minor imperfections were noticed on several samples, with the exception of sample HS13 which had grooves along the gage section parallel to the tensile direction. None of the minor imperfections were associated with cracking, however the grooves on the HS13 sample did interact with cracks and likely affected crack formation.

Table 1
Chemical compositions of tested alloys in wt%. Alloys FS–PS have the solute addition element indicated in bold.

Heat ID	Material	Fe	Cr	Ni	Mn	Mo	Si	N	C	P	S	Nb	Ti	O	Co	Cu	Hf
AS	SA CP304L	Bal.	19.95	10.80	1.82	0.53	0.56	0.0720	0.023	0.023	0.015	<0.001	0.02	0.011	0.22	0.29	–
BS/BR/B	SA/CW/CW CP316	Bal.	16.84	10.54	1.13	2.25	0.73	0.0210	0.056	0.022	0.022	0.008	0.01	0.009	0.12	0.25	–
CR	CW CP316 + Ti	Bal.	16.77	12.78	1.40	2.18	0.56	0.0080	0.070	0.013	0.007	<0.010	**0.38**	0.004	0.10	0.06	–
SW	SA CP304L	Bal.	18.42	10.45	1.07	0.00	0.24	0.0250	0.022	0.015	<0.002	–	–	–	–	–	–
ES	SA HP304L	Bal.	18.76	12.37	0.94	0.04	0.04	0.0003	0.021	<0.010	0.003	0.005	0.01	0.004	0.01	<0.01	–
FS	SA HP304L (−C)	Bal.	18.17	12.06	0.98	0.02	0.03	0.0005	**0.008**	<0.010	0.003	0.002	0.01	0.013	0.01	<0.01	–
GS	SA HP304L (+Mo)	Bal.	18.26	12.15	0.97	**2.36**	0.03	0.0004	0.020	<0.010	0.002	0.003	0.01	0.005	0.01	0.01	–
HS	SA HP304L (+Si)	Bal.	18.17	12.45	1.01	0.02	**1.05**	0.0005	0.020	<0.010	0.002	<0.001	0.01	0.007	0.01	<0.01	–
KS	SA HP304L (+Ni)	Bal.	18.21	**25.08**	1.00	0.02	0.03	0.0005	0.020	<0.010	0.002	<0.001	0.01	0.003	0.01	<0.01	–
LS	SA HP304L (Cr + Ni)	Bal.	**25.22**	**25.07**	1.02	0.02	0.03	0.0005	0.020	<0.010	0.002	<0.001	0.01	0.009	0.01	0.01	–
PS	SA HP316L (+Hf)	Bal.	17.03	13.60	1.01	2.18	0.10	–	0.028	0.010	0.007	–	–	–	–	–	**1.17**