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Comparison of the microstructure, deformation and crack initiation behavior of austenitic stainless steel irradiated in-reactor or with protons

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HIGHLIGHTS

- Dislocation loops were the prominent defect, but neutron irradiation caused higher loop density.
- Grain boundaries had similar amounts of radiation-induced segregation.
- The increment in hardness and yield stress due to irradiation were very similar.
- Relative IASCC susceptibility was nearly identical.
- The effect of dislocation channel step height on IASCC was similar.

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ABSTRACT

The objective of this study was to compare the microstructures, microchemistry, hardening, susceptibility to IASCC initiation, and deformation behavior resulting from proton or reactor irradiation. Two commercial purity and six high purity austenitic stainless steels with various solute element additions were compared. Samples of each alloy were irradiated in the BOR-60 fast reactor at 320 °C to doses between approximately 4 and 12 dpa or by a 3.2 MeV proton beam at 360 °C to a dose of 5.5 dpa. Irradiated microstructures consisted mainly of dislocation loops, which were similar in size but lower in density after proton irradiation. Both irradiation types resulted in the formation of Ni–Si rich precipitates in a high purity alloy with added Si, but several other high purity neutron irradiated alloys showed precipitation that was not observed after proton irradiation, likely due to their higher irradiation dose. Low densities of small voids were observed in several high purity proton irradiated alloys, and even lower densities in neutron irradiated alloys, implying void nucleation was in process. Elemental segregation at grain boundaries was very similar after each irradiation type. Constant extension rate tensile experiments on the alloys in simulated light water reactor environments showed excellent agreement in terms of the relative amounts of intergranular cracking, and an analysis of localized deformation after straining showed a similar response of cracking to surface step height after both irradiation types. Overall, excellent agreement was observed after proton and reactor irradiation, providing additional evidence that proton irradiation is a useful tool for accelerated testing of irradiation effects in austenitic stainless steel.

1. Introduction

Irradiation-assisted stress corrosion cracking (IASCC) is a complex, multivariable materials degradation phenomenon that affects core components in light water reactors (LWRs). IASCC, similar to SCC, is caused by the combination of an applied stress, a corrosive environment, and a susceptible material; however, irradiation adds complexity by altering both the material *and* environment. As reactors age, austenitic stainless steel (SS) core internal components will reach high neutron irradiation doses approaching 80 dpa [1], yet IASCC has been observed to occur in both service applications and laboratory experiments at relatively low irradiation doses between approximately 1–3 dpa [2]. Due to the high cost of reactor shutdowns for maintenance and the desire to safely extend the lifetimes of operating reactors, understanding the mechanism of IASCC and developing a mitigation strategy are of high importance.

Despite the substantial database on IASCC and significant research efforts aimed at improving understanding, the mechanism

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of IASCC remains unknown. One of the major barriers to solving this problem is the difficulty of acquiring and working with neutron irradiated materials. Irradiation programs are both time consuming and costly, and special requirements are necessary for shipping, handling, and characterizing radioactive samples. Proton irradiation is an attractive alternative for creating similar irradiation effects without the complications of working with neutron irradiated material. Proton irradiation also has the ability to cause moderately high irradiation doses in short amounts of time, while reducing the cost of irradiation and characterization as compared to working with reactor irradiated materials [3–5].

Several previous studies have compared the effects of neutron and proton irradiation, showing that in general, good agreement is observed. Was et al. [3] was the first to directly compare microstructures, microchemistry, and IASCC susceptibility of the same heats of commercial purity 304 SS and 316 SS after neutron and proton irradiation to doses between 1 and 5 dpa. Excellent agreement in microstructure and IASCC susceptibility was observed for both alloys. Microstructures consisted primarily of dislocation loops, grain boundaries were enriched in Ni and Si and depleted in Cr and Mo, the magnitude of irradiation hardening was nearly identical, and the dose to induce IASCC susceptibility was in excellent agreement. Gan and Was [5] compared microstructures of neutron and proton irradiated austenitic stainless steels, showing that the dose and irradiation temperature dependence of dislocation loop and void evolution were similar, regardless of irradiation type. In this study, the microstructural similarities were also observed to cause similarity in irradiation hardening [5]. Victoria et al. [6] observed dislocation loops of similar size in type 304 SS, although variations in loop size distribution were attributed to different neutron and proton irradiation doses. Was et al. [4] also compared the ratio of Ni and Cr radiation-induced segregation (RIS) between proton and neutron irradiated austenitic alloys, and observed similar trends in alloys with varying bulk Ni and Cr content.

Other studies have compared the effects of neutron and proton irradiation on materials other than austenitic stainless steel, further validating proton irradiation as a method for creating similar defect structures as neutron irradiation. Fournier et al. [7] and Tournadre et al. [8] observed Zircaloy-4 microstructures after proton irradiation to doses of 2 and 11.8 dpa, respectively. *a* type dislocation loops were the dominant microstructural feature observed at 2 dpa [7] while *c* type loops were observed at 11.8 dpa [8], indicating a threshold dose exists to cause *c* loop formation, similar to what has been observed after neutron irradiation [8]. The observed dislocation loop character, size, and density at both doses were comparable to neutron irradiated Zircaloy-4 microstructures [7,8]. Zu et al. [9] also studied irradiated Zircaloy, and noticed good agreement in dislocation loop structure as well as irradiation hardening. At low doses (less than 3 dpa), ferritic/martensitic steel was observed to form dislocation loops of similar size, although the loop density was inconsistent after proton and neutron irradiation [10]. Further, Victoria et al. [6] compared dislocation loops in neutron and proton irradiated Fe and Mo–5%Re, observing similar average loop size and loop size distribution in both materials, as well as a similar dependence of loop density with dose in irradiated Fe.

This study adds a new and sizable set of data to the database comparing the effects of neutron and proton irradiation on austenitic stainless steels. The recently completed Cooperative IASCC Research (CIR) I and II programs [11–18] subjected a variety of commercial purity (CP) and high purity (HP) solute addition alloys to either neutron irradiation in the BOR-60 fast reactor or proton irradiation at the Michigan Ion Beam Laboratory, with the objective of developing a mechanistic understanding of IASCC initiation and growth. After irradiation, characterization was conducted on alloy microstructures (quantifying dislocation loop, precipitate, and void formation), microchemistry (quantifying RIS of solute elements at grain boundaries), and mechanical properties (quantifying radiation hardening). Existing results are compared with regard to the difference in irradiation type. The IASCC susceptibility of neutron irradiated alloys are also compared with existing data from the same alloys irradiated with protons [11,12], and finally, localized deformation observed in neutron irradiated alloys is compared with that from proton irradiated alloys.

2. Experiment

2.1. Materials

A total of eight austenitic stainless steel (SS) alloys were compared after proton and neutron irradiation. Two were commercial purity (CP), and six were high purity (HP) solute addition alloys, each made from a single heat of a model type 304L stainless steel. All alloys were selected during the previous CIR program.

The two CP stainless steels had alloy designations A and B. Alloy A was type 304L steel from a boiling water reactor (BWR) core shroud and alloy B was type 316 steel from a pressurized water reactor (PWR) baffle bolt. In the alloy designation, a second letter 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 exact amount of added cold work was not reported for the commercial heats [17]. Alloys AS, BS, and BR or B were available for testing. Neutron irradiated alloy BS was reported to be cold worked to an even higher degree than alloy BR or B, despite the intent of this condition to be solution annealed [15]. Chemical compositions of CP alloys are given in Table 1.

The six HP solute addition alloys had designations E, F, G, H, K, and L. Alloys G, H, K, and L were created from a single heat of a model type 304L SS alloy, designated alloy E, which served as the reference condition. Small amounts of alloy E were re-melted for the purpose of adding solute elements Mo, Si, Ni, and Ni + Cr to create alloys G, H, K and L, respectively. Alloy F was created from a separate starting material due to the need to produce a low C content. Chemical compositions of each alloy are shown 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 to break up the as-cast dendritic structure. Solution annealing treatments between 900 °C and 1200 °C were applied for 0.5 to 1 h 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. [17]. All solute addition alloy designations contain the letter S, indicating a solution annealed starting microstructure.

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

A different tensile bar geometry was used in the reactor and proton irradiation experiments. All samples were fabricated and irradiated as part of the previous CIR program.

Tensile bars for neutron irradiation were fabricated with a 2 mm diameter cylindrical gage section and gage length of 12 mm, shown in Fig. 1a. The nominal pin hole diameter was 3.1 mm, but several of the neutron irradiated tensile bar samples had a reduced pin hole diameter of 1.5 mm. This modification was inconsequential, as neutron-irradiated samples were shoulder