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# Micro-mechanical investigation for effects of helium on grain boundary fracture of austenitic stainless steel

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## HIGHLIGHTS

- We investigate effects of helium on grain boundary fracture of stainless steel.
- We conduct micro-tensile tests on helium ion-implanted type 316 stainless steel.
- Brittle fracture occur on grain boundaries on which small bubbles formed densely.
- Formation of bubbles both on grain boundary and in matrix promotes brittle fracture.
- Grain boundary segregated helium atoms may have a role in grain boundary fracture.

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## ABSTRACT

Effects of helium (He) on grain boundary (GB) fracture of austenitic stainless steel were investigated by micro-tensile tests. Micro-bicrystal tensile specimens were fabricated for non-coincidence site lattice boundaries of He ion-irradiated 316 stainless steel by focused ion beam (FIB) micro-processing. Micro-tensile tests were conducted in a vacuum at room temperature in the FIB system. Specimens containing more than 2 at.% He fractured at GBs. The criteria for brittle fracture occurrence on GBs were: (1) He concentrations higher than 2 at.%; (2) formation of He bubbles on the GBs with less than a 5 nm spacing; and (3) matrix hardening to more than 4.6 GPa (nano-indentation hardness). The fracture stress of GB brittle fracture was lower for a specimen with higher He concentration while the size and areal density of the GB He bubbles were the same. The specimens that contained 10 at.% He and had been annealed at 923 K after irradiation fractured at the GB nominally in a brittle manner; however the inter-bubble matrix at the GB experienced ductile fracture. The annealing caused He bubbles to grow but decreased the areal density so that the spacing of the GB He bubbles widened and the hardness decreased, therefore the fracture mode changed from brittle to ductile. The findings revealed that He promotes GB fracture by weakening the GB strength and hardening the matrix due to the formation of He bubbles both on GBs and in the matrix. In addition, the findings suggested that GB segregated He atoms may have a role in GB fracture.

## 1. Introduction

Austenitic stainless steels are widely used as structural materials for core internals in fission reactors, that is both light water reactors (LWRs) and fast breeder reactors (FBRs). Neutron irradiation induces various changes in material properties of the stainless steels, such as increase in yield strength, ductility loss and degradation of corrosion resistance of grain boundaries (GBs). These changes are attributed to the formation of radiation-induced defects and GB segregation. The radiation-induced microstructural and microchemical changes also relate to the mechanism of irradiation assisted stress corrosion cracking (IASCC) [1]. IASCC is a primary concern for baffle former bolts. The bolts are mostly fabricated from cold-worked 316 stainless steel (316SS). Highly neutron-irradiated, cold-worked 316SS specimens, which had been obtained from pressurized water reactor (PWR) core thimble tubes, fracture intergranularly during tensile tests conducted even in an inert environment [2]. This suggests that GB cohesive strength might be weakened by neutron irradiations without environmental effects. The reduction of GB strength may promote the occurrence of IASCC. Fujimoto et al. [3] have reported that IASCC susceptibility was higher for 316SS components irradiated in a commercial PWR than those irradiated in an FBR, while the hardness and GB composition were the same. They have suggested that

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higher production rates of hydrogen and helium (He) in the PWR-irradiated steels, due to the softer neutron spectrum compared with the FBR, and formation of GB cavities in the PWR-irradiated steels play an important role in the occurrence of IASCC for PWR conditions. He atoms might accumulate at GBs and weaken the GB strength by formation of cavities. However, the relationships between both microstructure and microchemistry of GBs and GB cohesive strength or fracture behavior have not been well examined. Recently, it has been suggested that local stress concentrations at GBs, due to radiation-induced deformation localization, is important to initiate intergranular stress corrosion cracking (IGSCC) [4]. Clarification of radiation effects on GB strength and fracture behavior would lead to better understanding of the IASCC mechanism.

It is well known that He causes ductility loss during creep tests at elevated temperatures [5,6] and IG cracking after welding of neutron-irradiated austenitic stainless steels [7,8]. Several researchers have proposed both accumulation of atomistic He at GBs due to self-diffusion (or sweeping by moving dislocations) and formation of GB cavities as a mechanism of He embrittlement [9–12]. The He embrittlement becomes less significant as test temperatures decrease [10,12–14]. The reason for the weaker He effects at lower temperatures might be the greater difficulty for He atoms to accumulate at GBs, and for GB cavities to grow, at the lower temperatures. Bennetch and Jesser [15] have suggested that formation of small GB cavities with small spacing is favorable to occurrence of IG cracking at temperatures below 823 K. He is a possible factor that reduces GB cohesive strength; however the effects of He on GB fracture of austenitic stainless steels at lower temperatures have been poorly understood compared with effects at elevated temperatures.

To investigate the effects of He on GB fracture of austenitic stainless steels, He ion irradiation is a useful technique because it can achieve accurate injections of various He concentrations in a broad temperature range with formation of radiation defects. However, the irradiated region is limited to a few μm below the surface due to the short penetration range of He ions. In addition, it is difficult to examine the relationships between fracture of an individual GB and microstructure (or microchemistry) of the GB at the same time by using conventional testing methods. Recently, some micro-mechanical testing techniques have been developed to accompany technological minimization [16]. These techniques are used to evaluate mechanical properties of micron-sized structures such as micro-electro-mechanical systems (MEMS). Focused ion beam (FIB) micro-processing is one of the major techniques to fabricate micron-sized specimens, and it can be used to fabricate the specimen from an ion-irradiated region [17,18]. Micro-bending testing is well used to estimate interface strength; cantilever type specimen containing an interface are used in the tests [19]. The bending tests would be suitable for brittle materials, but not for ductile materials because most ductile materials do not fracture by bending. Fujii and Fukuya [20] have reported a micro-tensile testing method for micron-sized specimens with a GB; the specimens are prepared by FIB micro-processing. They have applied a load on the specimens in FIB system using a micro-cantilever and discussed fracture behavior of phosphorus segregated GB in Fe–Mn–P alloy. The relationships between fracture behavior (fracture mode, stress and strain) and GB microstructure (or microchemistry) can be discussed by using the micro-cantilever method, even for ion-irradiated materials. In the present study, the micro-tensile testing method was applied for micro-specimens of He ion-irradiated, cold-worked 316SS to investigate the effects of He on GB fracture of austenitic stainless steels. The relationships examined between the fracture behavior of micro-specimens were: (1) He concentration, (2) the characteristics of GB cavities, and (3) the hardness.

## 2. Experimental procedure

### 2.1. Material and He ion irradiation

An unused PWR core thimble tube (316SS) with an average grain size of approximately 30 μm was used. The tube was solution annealed at 1350 K and finally cold-worked to approximately 15% by drawing. Table 1 shows the chemical composition. Several plates of 5 × 4 × 1 mm were cut from the tube, and their surfaces were mechanically polished using colloidal silica.

Ion irradiations were carried out with 75 and 190 keV $He^+$ ions at room temperature or 573 K. The maximum irradiation depth was 800 nm, calculated by the SRIM-2013 code [21] using the Kinchin-Pease option with the displacement threshold energy of 40 eV. Fig. 1 shows the calculated depth distributions of implanted He atoms in Fe–17.3Cr–12.5Ni alloy. Average He concentrations in the whole irradiated region ranged from 0.05 to 6 at.%. Some plates were annealed at 923 or 1023 K in a vacuum for 1000 s to grow He bubbles after the ion irradiations. The He ion irradiation and post-irradiation annealing conditions are summarized in Table 2. The plates are designated A to F according to He concentrations and post-irradiation annealing. Plates C to E were as-irradiated plates and plates A, B and F were post-irradiation annealed plates. It is noted that 190 keV $He^+$ ions were only irradiated for plates A to C. In this paper, both He concentration and dose due to the ion irradiation in each plate were defined as average values in the whole irradiated region; these values are shown in Table 2.

Cross-sectional microstructures of GBs were observed by using a transmission electron microscope (TEM) equipped with a field emission gun of 300 kV (Hitachi HF-3000). The TEM specimens were prepared from non-coincidence site lattice (CSL) boundaries by using an FIB system (Hitachi FB2000A) with 30 kV gallium ions. The damage introduced in the TEM specimens during FIB micro-processing was removed by using a precision ion polishing system (Gatan PIPS 691) with acceleration voltages less than 2 kV. The TEM specimen was set to tilt the GB plane parallel to the beam direction (edge-on condition). For He bubbles on the GB, diameter and number were measured. The observations were carried out using defocus values of approximately 1 μm. Some He bubbles would overlap with other bubbles and would not be counted. The TEM sample was then tilted between 5° and 15° from the edge-on condition to count the overlapped GB He bubbles. After

**Table 1**
Chemical composition of 316SS (wt%).

| C | Si | Mn | P | S | Ni | Cr | Mo | Fe |
|---|---|---|---|---|---|---|---|---|
| 0.023 | 0.44 | 1.67 | 0.022 | 0.009 | 12.51 | 17.31 | 2.05 | Balance |

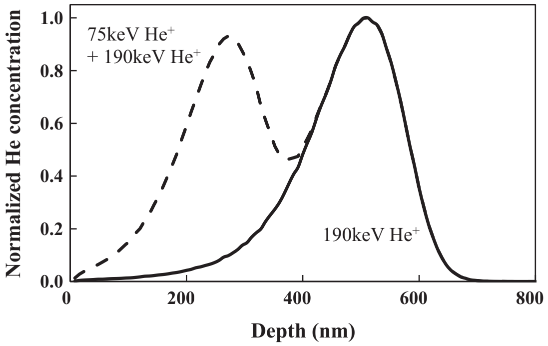

**Fig. 1.** Depth distribution of implanted He atoms in Fe–17.3Cr–12.5Ni alloy calculated by the SRIM code.