Evolution of cellular dislocation structures and defects in additively manufactured austenitic stainless steel under ion irradiation

Shilei Li [1, 2, +], Jing Hu [2, +], Wei-Ying Chen [2], Jingyue Yu [1], Meimei Li [2, *], Yandong Wang [1, *]

[1] Beijing Advanced Innovation Center for Materials Genome Engineering, State Key Laboratory for Advanced Metals and Materials, University of Science and Technology Beijing, Beijing 100083, China
[2] Nuclear Science and Engineering Division, Argonne National Laboratory, Argonne, Illinois 60439, USA

[+] These authors contributed equally to this work.
[*] Corresponding authors.
E-mail addresses: Meimei Li (mli@anl.gov), Yandong Wang (ydwang@ustb.edu.cn).

Abstract

The evolution of irradiation-induced defects in additively manufactured (AM) austenitic stainless steel was investigated *in situ* by 1 MeV Kr ion irradiation at 450 and 600 °C in a transmission electron microscope. Cellular dislocation structure in AM steel act as sink/trap sites for the irradiation-induced defects, resulting in the lower density and smaller dislocation loops in AM steel than conventional forged (CF) steel at 450 °C. The lower stacking fault energy along with the interaction between cellular dislocation structures and defects in AM steel promotes the unfaulting process and the formation of network dislocation at 600 °C.

Key words: Additive manufacturing, austenitic stainless steel, ion irradiation, irradiation-induced defect, cellular dislocation structure

Additive manufacturing (AM) has attracted much attention over the past ten years due to its immanent advantages, such as unrivalled design freedom and short lead times [1-7]. Selective laser melting (SLM), one of the most well-developed processes for AM metals, allows the fabrication of components with high structural integrity at a low cost [8-14]. By reducing residual porosity, insufficient layer bonding, and hot cracking, typically achieved using hot isostatic pressing, several AM alloys with mechanical properties comparable to conventionally manufactured alloys have been produced [1, 15]. AM 316L stainless steels exhibit a combination of high yield strength and tensile ductility that surpasses their conventional forged (CF) counterpart [16].

Conventional forged (CF) austenitic stainless steels used in light water reactors and sodium fast reactors were found to be susceptible to irradiation-assisted stress corrosion cracking [17] and void swelling [18]. By introducing a high density of dislocations as the sink sites of irradiation-induced defects, cold-worked austenitic stainless steels can achieve significantly improved radiation resistance [19-20]. The cold-worked materials usually sacrifice some of the plasticity and may be not applicable for the components with complex shapes. The excellent combination of strength and ductility of AM 316L is attributed to its unique hierarchical microstructures [16, 21]. Recently, the behavior of AM 316L under ion irradiation was investigated under 2 MeV proton ions at 360 °C [22] and 1MeV Kr ions at 400 °C [23]. Much work is still needed to evaluate the performance of AM materials in nuclear environments, especially at temperatures above 400 °C.

In this study, we evaluated the irradiation response of AM 316L steel using *in situ* heavy ion irradiation in a transmission electron microscope (TEM) under 1 MeV Kr ion irradiation at 450 and 600 °C. The evolution of irradiation-induced defects and their interaction with the cellular dislocation structures were investigated. The irradiation behavior of AM 316L was compared with a CF 316L steel to understand the irradiation response of this innovative material and gain insights into the development of advanced alloys with superior radiation resistance.

The AM 316L sample was fabricated by a SLM facility EOS M290 (EOS GmbH, Krailling, Germany) equipped with a 400 W Yb-fiber laser. Spherical gas atomized powders were selected and the following parameters were used: laser power (340 W), beam size (100 μm), layer thickness (40 μm), scanning speed (1300 mm/s) and line spacing (150 μm). A pillar (10 mm of length and width, 50 mm of height) was built and no post-built heat treatment was applied. The bulk chemical compositions of the as-built AM 316L and CF 316L, measured by electron spectroscopy for chemical analysis, are listed in Table 1.

Thin foil disks for TEM were sliced from the AM 316L pillar with their normal direction parallel to the building direction. TEM samples of CF 316L were cut from a wrought bar. These TEM samples were mechanically thinned to 0.06 mm and electropolished to perforation using 95% ethanol 5% perchloric acid electrolyte at –40 °C. The microstructure and elemental distribution in AM 316L were investigated using a FEI Talos F200X TEM at 200 kV equipped with a four-quadrant energy dispersive X-ray spectrometer (EDS) using a step size of ~5 nm and acquisition time of 45 minutes.

In situ irradiation experiments were performed at Intermediate-Voltage Electron Microscopy (IVEM)-Tandem Facility of Argonne National Laboratory. The microstructural evolution under irradiation was observed in a Hitachi-9000 TEM with 300 kV electrons. Both AM and CF specimens were irradiated with 1 MeV Kr ions (6.3×10^{11} ions/cm^2/sec) at 450 and 600 °C. The dpa values were estimated using the calculation method in Ref. [24]. Real-time videos of defect formation and evolution was captured on a high-speed CCD camera, and the size and number density of irradiation-induced defects was measured using TEM images. The size and density of dislocation loops were measured manually and at least one hundred loops were used for quantitative analysis in each condition. The density of the loops was calculated by dividing the count with the foil thickness estimated from thickness contours.

Figures 1a-c show the microstructure under different magnifications of the AM 316L. The as-built AM 316L steel has a grain size of 3~5 microns. Each grain is composed of dozens of dislocation cells with a mean size of approximately 500 nm (Figure 1d). Comparing with the cellular boundaries (CBs), the dislocation density is very low inside the cell. The EDS elemental mapping are shown in Figure 1e. The semi-quantitative composition of both CB and cell interior were also detected by EDS and are listed in Table 1. The CBs are enriched in ferrite-forming elements of Cr, Si and Mo. This micro-segregation on CBs was related to the rapid solidification process [25]. Nanoparticles enriched in Si, O and Mn elements were also found in AM 316L steel.

The AM 316L TEM samples were *in situ* irradiated in a TEM to observe the evolution of cellular dislocation structures and defects. Figure 2a shows the microstructures and defects at the scale of a cellular structure in AM 316L after irradiation to different doses at 450 °C. All images were taken with g_{200} close to <110> zone. While the cellular dislocation structure in AM 316L was still visible at 2 dpa, the dislocation density at CB significantly decreased after irradiation to 0.5 dpa, and remained relatively unchanged upon continued irradiation. High density of irradiation-induced "black-dots" (small dislocation loops) were observed inside the cell after irradiation to 0.5 dpa. Due to the high dislocation density near CBs, it is difficult to count the loops near CBs. Quantitative analysis of dislocation loop size and density was performed in the cell interior, and the results are given in Figures 2c and e. With increasing irradiation dose, their sizes increased and their density decreased. The faulted interstitial Frank loops resulting from clustering of self-interstitial atoms are the primary type of irradiation-induced dislocation loops at lower temperatures in austenitic stainless steels [26]. These sessile Frank loops are lying on the {111} planes with a Burgers vector of 1/3 <111> [27].

During irradiation at 600 °C, the cellular dislocation structures underwent a dramatic change, as shown in Figure 2b. The pre-existing dislocations on the CBs gradually disappeared with increasing irradiation dose. Comparing with irradiation at 450 °C, the irradiation microstructure at 600 °C in AM 316L changed from the small loops-dominated microstructure to a mixture of small and large dislocation loops and even network dislocations. Dislocation loops larger than 10 nm can be observed in AM 316L after irradiation at 600 °C to 0.5 dpa, and both the size and density of loops increased when the dose reached 1.0 dpa. The density of dislocation loops in AM316L after irradiation at 600 °C to 2.0 dpa was much lower than that at 0.5 and 1.0 dpa. The dominated irradiation-induced microstructure at 2.0 dpa becomes network dislocation, which implies the growth and unfaulting of dislocation loops.

The size distribution and density of loops at 450 and 600 °C are compared in Figures 2c-e. The average loop size is 4.2 ± 1.0, 4.8 ± 1.3, and 6.0 ± 1.2 nm after irradiation to 0.5, 1.0, and 2.0 dpa, respectively, at 450 °C. The loop size at 600 °C for the same doses is about 4 times larger than that at 450 °C. Most of the dislocation loops at 450 °C are smaller than 10 nm, while the loops at 600 °C exhibit a bimodal size distribution over a much wider range. The loop density in AM 316L decreases with increasing irradiation dose at 450 °C, but the density at 600 °C shows no significant change with increasing dose.

CF 316L samples were also irradiated under the same irradiation conditions for comparison with AM 316L. Figure 3a shows the defect evolution in CF 316L during irradiation at 450 °C. The dominant defect in CF 316L is dislocation loops after irradiation up to 2.0 dpa. Larger dislocation loops can be clearly observed in CF 316L at 2.0 dpa and most of them are faulted loops. Figure 3b shows the evolution of defects at 600°C. The loop size in CF 316L at 600 °C is larger than that at 450 °C after irradiation to the same dose. The loop size increased significantly with increasing dose. “Black-dots” changed to larger loops when the irradiation dose exceeded 0.5 dpa. Most loops in CF 316L after irradiation at 600 °C for 2.0 dpa were faulted 1/3 <111> loops and no dislocation network was observed.

Quantitative analysis of dislocation loop size and density in CF 316L is shown in Figures 3c-e. The loop size in CF 316L is larger than that in AM 316L after irradiation to the same dose at 450 °C. The loop density at 450 °C in CF 316L is higher than that in AM 316L under the same irradiation condition, and both of them decrease with increasing dose. In contrast, the average loop size in CF 316L is smaller than that in AM 316L at 600 °C, while the loop density in CF 316L is much higher. Moreover, the dislocation loops in CF 316L after irradiation for 2.0 dpa at 600 °C still remain a unimodal size distribution.

The video captured during *in situ* irradiation helps understand the dislocation sink effect of irradiation-induced defects in AM 316L. Figure 4a shows the snapshots of the interaction process of irradiation-induced dislocation loops (red arrows) and the pre-existing dislocations (blue arrows) inside a dislocation cell during irradiation from 1.22 to 1.36 dpa at 450°C. At 1.22 dpa, two dislocation loops were separate and had a distance of ~ 15 nm from the pre-existing dislocation lines near a CB. When the dose increased to 1.26 dpa, these two loops and the dislocation lines became linked with each other. The two dislocation loops evolved into a large dislocation loop at 1.30 dpa. This large dislocation loop continued to interact with the pre-existing dislocation lines, gradually became smaller and finally disappeared after irradiation to 1.36 dpa. The above observations provide direct evidence that the pre-existing dislocations in AM 316L are effective defect sinks under ion irradiation. The interaction between loops and the pre-existing dislocation was also observed in AM 316 steel under irradiation at 400 °C [23].

Dislocations, as biased sinks for interstitial defects due to the drift of interstitials down the stress gradient near the dislocation core, can be considered as an unsaturable sink because the absorption of interstitials can be accomplished through dislocation climbing [27]. As CBs have much higher dislocation density, the preferential sink for interstitial defects coupled with the relative fast diffusion, causes more defect annihilation near the CBs, thus the lower concentration of interstitial defects in the cell interior of AM 316L comparing with CF 316L when irradiation at 450 °C.

The interaction between defects and cellular dislocation structures becomes more obvious at 600°C, as marked by the blue circles in Figure 4b. The irradiated (600 °C/0.5dpa) AM 316L sample show a mixed irradiation microstructure, including "black-dots" (small Frank loops), larger Frank loops, perfect loops (marked by red arrows), and dislocation network. The evolution of irradiation defects in AM316L at 600 °C can be inferred as: "black-dots" → larger Frank loops → perfect loops → dislocation network. The microstructure of AM 316L after annealing at 600 °C for 1 h was also characterized by TEM, and the cellular dislocation structures remained after annealing, indicating that the microstructural evolution in Figure 4b was not induced by thermal recovery.

The sessile Frank loops have a tendency to transfer to perfect loops with lower energy by the unfaulting process after they grow up to a critical size at higher does or at high temperatures. In austenitic stainless steels, the unfaulting process from faulted 1/3 <111> loops to perfect 1/2 <110> loops usually occurs at about 600 °C [28]. The formed perfect loops are glissile and can glide to interact and form network dislocations [29]. The transition from a loop-dominated dislocation microstructure to a network-dominated dislocation microstructure depends synergistically upon several parameters, including alloy composition, irradiation spectrum and dose [27, 29].

As the unfaulting process involves the removal of stacking faults, the prerequisite for its spontaneous occurrence is that the elastic energy of perfect loop becomes less than the sum of the elastic energy of Frank loop and the stacking fault energy (SFE), even though different unfaulting mechanisms may exist. Thus, the material with lower SFE is expected to experience unfaulting earlier. The SFE of Fe-Cr-Ni steel can be estimated using the relationship between SFE and composition of major alloying elements reported in Ref. [30].

The SFE values of the studied CF 316L (bulk) and AM 316L (bulk, CB, and cell interior) were calculated as 42.54, 56.15, 64.02 and 51.6 mJ/m^2, respectively. The region of CBs in AM 316L has the highest SFE comparing with the cell interior in AM 316L and the conventional 316L steel, which can influence the evolution of irradiation-induced defects, especially for the loop unfaulting process.

In CF 316L, no perfect loops formed after irradiation at 450 °C to 2 dpa, and only few perfect loops were observed after irradiation to 2.0 dpa at 600 °C. Figure 4c shows the bright field and dark field images using g{200} near the [110] zone axis and the corresponding rel-rod dark field images of Frank loops in the irradiated (600 °C/2dpa) CF 316L sample. This Frank loop-dominated microstructure indicates that the unfaulting process did not start on a large scale in CF 316L at 600 °C after irradiation to 2dpa.

In AM 316L only Frank loops were observed after irradiation at 450 °C to 2 dpa, while perfect loops and dislocation network appeared in after irradiation to only 0.5 dpa at 600 °C. The comparison of defect evolution in AM 316L between 450 and 600 °C suggests that the unfaulting process has a strong temperature dependence and this process controls the transition of irradiation microstructures from loop-dominated to network-dominated. The evolution kinetic of dislocation loops in AM 316L at 600 °C appears to be faster than conventional 316L, resulting in early formation of dislocation network. Its influence on the mechanical properties deserves further systematical investigations for its application in harsh environments such as high temperatures and/or high irradiation doses.

In summary, the as-built AM 316L steel has a unique cellular dislocation structure and Cr, Si and Mo elements are segregated on the CBs. The irradiation microstructures in CF 316L and AM 316L irradiated up to 2 dpa at 450 °C are Frank-loop-dominated. The irradiation microstructures in CF 316L irradiated to 2 dpa at 600°C are still Frank-loop-dominated, while the AM 316L samples irradiated under the same conditions have a microstructure mixed with Frank loops, perfect loops, and dislocation network. *In situ* observation reveals that the cellular dislocation structure in AM 316L acts as sink/trap sites for the irradiation-induced defects. The higher SFE of CBs and cell interior in AM 316L promotes the unfaulting process, results in early formation of network dislocation in AM 316L at 600 °C.

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Author contributions

S.L. and Y.W. designed the research project; J.H., W.-Y. C. and M.L. performed the *in situ* irradiation experiments; J.Y. prepared the TEM samples; S.L. analyzed the data and wrote the manuscript; all the authors contributed to the discussions and commented the manuscript.

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