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Void swelling behavior in electron irradiated Fe–Cr–Ni model alloys under temperature variation

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Abstract

We examined the effect of short HVEM pre-irradiation at lower or higher temperature on void swelling behavior in electron irradiated austenitic model alloys, Fe–15Cr–xNi ($x = 15$, 20, 25 or 30 wt%). By 6 dpa irradiation at constant temperature, voids were formed at 200–400 °C in 15, 20, and 25Ni alloys but did not form above 350 °C in the 30Ni alloy. In the temperature variation irradiation, an initial 0.6 dpa irradiation at 200–500 °C was followed by irradiation of 5.4 dpa at 400 °C. The effect of pre-irradiation at lower temperature was a slight increase in the void number density, and that at higher temperature was a suppression of void formation. SFT induced by irradiation at lower temperature were not effective void nuclei in the subsequent irradiation at higher temperature.

1. Introduction

The effect of temperature variation during irradiation on defect structure appears with a large variation depending on the materials and the irradiation conditions, such as temperature history and damage level [1–3]. To understand the mechanism of the effect is important for developing materials for future nuclear applications. Electron irradiation using high-voltage electron microscope (HVEM) yields the advantage of in situ observation of the defect structure development, easy change of irradiation conditions such as irradiation temperature, and high damage rate ($>10^{-3}$ dpa/s). In model alloys of austenitic stainless steel Fe–Cr–Ni, void swelling is well-known to show a clear nickel dependence upon irradiation with various particles, ions, neutrons, and electrons: void swelling is suppressed at intermediate nickel levels of 30–60% [4–6]. The present study examines the effect of short pre-irradiation at lower or higher temperature on void swelling behavior in electron irradiated austenitic model alloys, Fe–15Cr–xNi ($x = 15$, 20, 25, 30 wt%).

2. Experimental procedure

The specimens examined were four austenitic model alloys, Fe–15Cr–xNi ($x = 15$, 20, 25 or 30 wt%). Disc specimens of 3 mm diameter were punched from sheets after rolling to 0.1 mm thick. The specimens were then annealed in vacuum

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of about 10^{-4} Pa at 1050 °C for 3 h. Thin foil specimens for electron microscopy were prepared by jet polishing in a solution of $CH_3COOH:HClO_4$ (19:1) followed by final polishing in $H_2O:H_2SO_4:H_3PO_4$ (3:3:4).

Electron irradiation and in situ observation were performed with a JEM-ARM1250 high-voltage electron microscope at Tohoku University, operating at 1250 kV with current density of about 3×10^{24} electrons $m^{-2}\,s^{-1}$. In the temperature range 200–500 °C, irradiation effects were examined under constant temperature and with a two-step temperature variation. The typical damage level was 6 dpa after irradiation for 20 min. The irradiation was made on wedge shape specimens of thicknesses from 0 to 250 nm. The irradiation and observation were performed along a direction between [1 0 0] and [1 1 0] with 002 systematic reflections excited. The number density of voids were measured, with the thickness determined using the extinction distance of 52.6 nm for 002 reflection with 1250 kV electrons.

3. Results and discussion

3.1. Constant temperature irradiation

Fig. 1(a) shows the typical defect structure in Fe–15Cr–xNi alloys irradiated to 6 dpa at four temperatures. Voids decrease in number density and reach larger size at higher irradiation temperatures, as seen in Fig. 2. Voids started to form after an incubation period of several minutes and nucleation was completed before 20 min (i.e., 6 dpa), except at lower temperatures (e.g., 200 °C). From the figure, the nickel content dependence can be observed as follows. For alloys of 15, 20, and 25Ni, voids formed in the temperature range from 200 °C to 400 °C with the swelling peak around 350–400 °C. The void number density was slightly higher for 20 and 25%Ni than that for 15%Ni, when compared at the same temperature. For Fe–15Cr–30Ni, void formation did not occur above 350 °C.

The specimen foil was considerably damaged by heating to 600 °C in HVEM. Yoshiie et al. reported

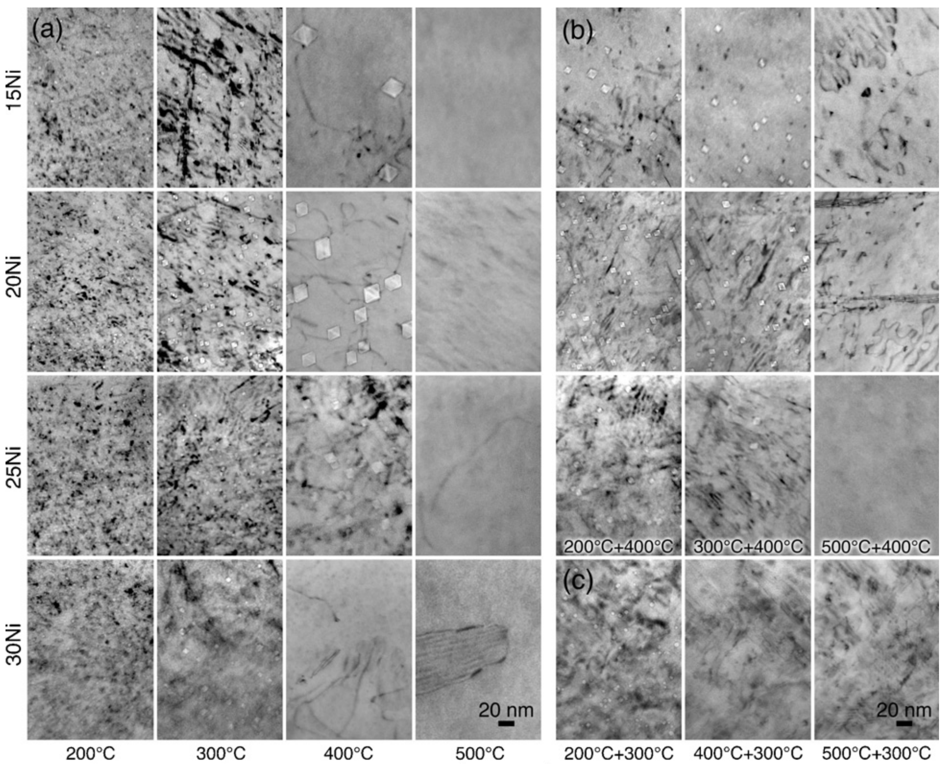

Fig. 1. Typical defect structure observed in electron irradiated Fe–15Cr–xNi (x = 15, 20, 25, 30 wt%), 1250 kV, 3×10^{24} electrons $m^{-2}\,s^{-1}$. (a) Irradiation at constant temperature for 6 dpa, (b) two-step irradiation, T_1 °C × 0.6 dpa + 400 °C × 5.4 dpa for x = 15, 20, 25 wt%, and (c) T_1 °C × 0.6 dpa + 300 °C × 5.4 dpa for x = 30 wt%.

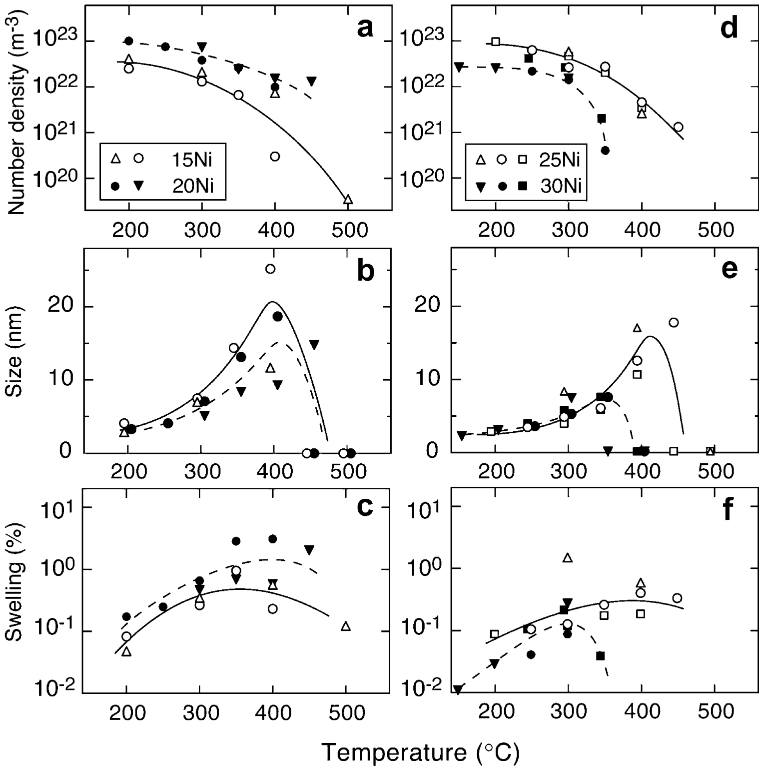

Fig. 2. Temperature dependence of the void number density, average void size, and swelling in Fe–15Cr–xNi ($x = 15, 20, 25, 30$ wt%) after electron irradiation for 6 dpa, 1250 kV, 3×10^{24} electrons $m^{-2} s^{-1}$. Each symbol corresponds to an individual experimental run.

that the heating to 600 °C changed the dislocation loop formation in the subsequent irradiation at lower temperature [7]. In the present experiment, the heating suppressed void formation in irradiation at lower temperatures. Because a heat-treatment for 40 min at 500 °C did not cause any considerable changes on the formation of voids and dislocation loops, this is the upper limit temperature in the present experiments.

3.2. Temperature variation irradiation

The standard irradiation condition employed was an initial irradiation of 0.6 dpa in the temperature range from 200 °C to 500 °C, and subsequent irradiation of 5.4 dpa at 400 °C. Voids were not observed during the first-step irradiation at any temperature. Fig. 1(b) compares typical examples of defect structure induced by temperature variation. Fig. 3 shows the void number density, average void size, and the swelling as a function of the first-step irradiation temperature. The reference experiment is 6 dpa irradiation at constant temperature, 400 °C, which is shown by shading in the figure.

Voids were observed in the alloys with 15, 20, and 25Ni. Although the data are rather scattered, the first-step irradiation temperature affects void formation in these alloys. The effect of first-step irradiation at lower temperature is a slight increase in the void number density. For example, the first-step irradiation at 200 °C increased the void number density by a factor of about two compared to that of constant temperature irradiation at 400 °C. Simultaneously the void size was decreased, which may be explained by the fact that the increase in void number density decreased the number of vacancies assigned per void. On the other hand, the effect of first-step irradiation at higher temperature was the opposite; the void number density decreased approximately an order of magnitude by the first-step irradiation at 500 °C.

No voids were observed in the Fe–15Cr–30Ni alloy after the standard temperature variation irradiations. When the first-step irradiation at 200 °C was prolonged to 3 dpa, voids grew larger during the second-step irradiation at 400 °C. Because small voids (2–3 nm in size) were formed during the long first-step irradiation, the growth of voids was shown

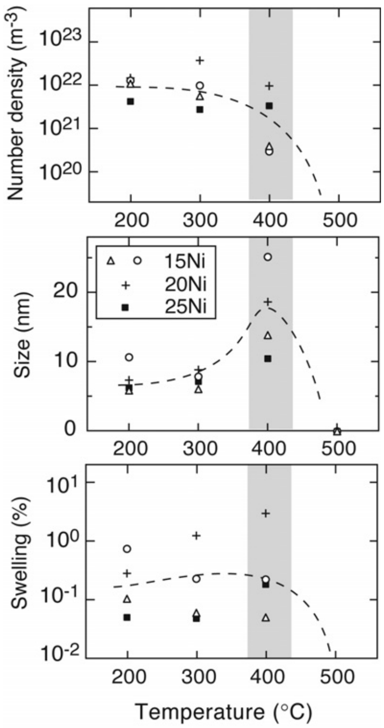

Fig. 3. First-step irradiation temperature (T_1) dependence of the void number density, average void size, and swelling in Fe–15Cr–xNi ($x = 15$, 20, 25 wt%) after electron irradiation, T_1 °C × 0.6 dpa + 400 °C × 5.4 dpa, 1250 kV, 3×10^{24} electrons m^{-2} s^{-1}. The data at $T_1 = 400$ °C shown with shading corresponds to the reference result, i.e., 400 °C × 6 dpa.

to be possible at 400 °C. Then we examined the 30Ni alloy under another two-step irradiation condition with a low second-step temperature: 0.6 dpa at 200–500 °C followed by 5.4 dpa at 300 °C. First-step irradiation at 200 °C increased the void number density by a factor of 1.4, while that at 400 °C and 500 °C suppressed the void formation (see Fig. 1(b)). These results suggest that the effect of first-step irradiation of 30Ni is similar to that of the other alloys except for the difference in the temperature range.

3.3. Mechanism of effects of temperature variation irradiation

The enhanced formation of defect clusters after a short irradiation at lower temperature has been reported under several irradiation experiments [1,2]. This effect can be understood from the general nature of the defect structure development: the nucleation of point defect clusters is enhanced at lower temperatures and the growth of clusters is faster at higher temperatures. In the present experiment stacking fault tetrahedra (SFT) induced by the irradiation at low temperature can act as void nuclei, according to the process of 'conversion from SFT to void' reported by Kojima et al. [8]. For example, the number density of SFT formed in the 20Ni was 2×10^{23} m^{-3} at 200 °C, while that of voids was 2×10^{22} m^{-3} at 400 °C. If some fraction of SFT converts to voids, an increase of void number density is expected. We examined this mechanism in the following way.

We induced SFT by irradiation to 1.5 dpa at 200 °C, and traced them during the second-step irradiation at 400 °C. A large number of SFT were observed in the initial stage of the second-step irradiation (Fig. 4(a)); and after irradiation for about 700 s (Fig. 4(b)), most SFT disappeared and voids were observed. From a careful comparison of these two figures, about 5% of newly appeared voids were found to form adjacent (mutual distance smaller than defect cluster size) to some SFT that had disappeared. Even though their mutual distance along the depth in the specimen foil was unknown, it is clear that the contribution of the conversion process involves less than 5% of the newly appeared voids. Accordingly, the enhanced formation of voids is not due to the conversion from SFT, but probably from some nuclei that are too small to be observed with TEM.

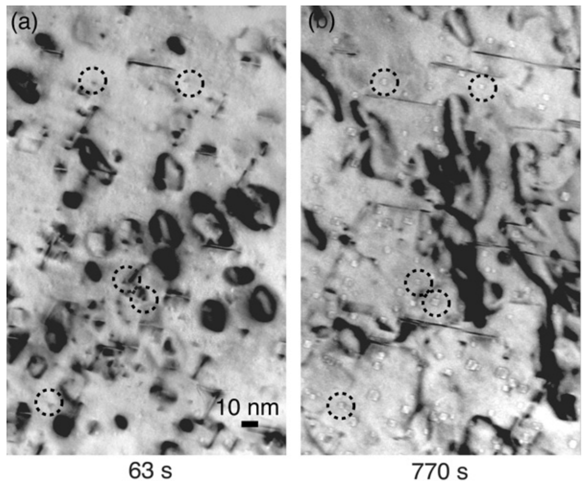

Fig. 4. Defect structure of identical area in Fe–15Cr–20Ni during the second-step irradiation at 400 °C. The first-step irradiation was 1.5 dpa (300 s) at 200 °C. 1250 kV, 3×10^{24} electrons m^{-2} s^{-1}. The circles show defect clusters that were possible to convert from SFT to void.

The suppression of void formation by the first-step irradiation at high temperature has not been reported previously. The effect is difficult to be explained by reactions between point defects and defect clusters, because the first-step irradiation at 500 °C induced very few defect clusters (sometimes no defect clusters were observed). We propose one possible mechanism. The 'radiation-induced purification' has been first proposed by Kiritani et al. [9] to explain a suppression of interstitial-type dislocation loops after the irradiation at high temperature. For example, pure Cu showed a remarkable decrease (reduced to 1/30) in the number density of interstitial loops formed by the irradiation at room temperature when the area had been pre-irradiated at 300 °C for 30 s (though no defect clusters were observed with TEM after the pre-irradiation) [10]. The interpretation of the result is (1) some impurity atoms greatly enhanced the loop nucleation and (2) the effect of the enhancement was lost when the impurities were transported from the matrix to specimen surfaces by point defects induced by pre-irradiation at higher temperatures [9,10]. In Fe–Cr–xNi, impurities are regarded to enhance void formation and to be transported to specimen surfaces during irradiation at high temperatures. We note that this mechanism is characteristic of irradiation of thin foil specimens, and probably for materials with higher sink concentrations.

4. Conclusion

We examined the effect of short HVEM pre-irradiation at lower or higher temperature on void swelling behavior in electron irradiated austenitic model alloys, Fe–15Cr–xNi ($x = 15$, 20, 25 or 30 wt%). In 15, 20, and 25Ni alloy, the short irradiation at 200 °C and 300 °C increased slightly the void number density in subsequent irradiation at 400 °C, and that at 500 °C considerably reduced the void formation. A similar effect was observed in 30Ni, though swelling level was lower and temperature range of void formation shifted to lower temperatures.

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