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Journal of Nuclear Materials 255 (1998) 34–43

journal of nuclear materials

Void swelling in Fe–15Cr–xNi ternary alloys under proton irradiation

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Received 24 April 1997; accepted 2 January 1998

Abstract

Void swelling and microstructure in Fe–15Cr–xNi ($x = 20$, 25, 30 wt%) ternary alloys irradiated with 180 keV protons at 723–873 K (450–600°C) were examined by TEM. The displacement damage levels were 5–20 dpa for the alloys preinjected with 10 appm He at room temperature, and 5 dpa for the alloys without He. The introduced H seems to play the same role as He in enhancing void nucleation at the lower temperatures (723, 773 K). Although the preinjected He can suppress the Ni-dependent incubation dose at all the irradiation temperatures examined, the Ni-dependent swelling rate may be responsible for the Ni influence on swelling at the higher temperatures (823, 873 K). The turning point of the Ni influence on swelling lies around 823 K in the present proton irradiation.

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1. Introduction

The radiation-induced swelling caused by high energy neutrons has been recognized as one of the serious problems for fusion and fast breeder reactor materials. Numerous studies have been focused on the swelling behavior in the candidate materials such as Fe–Cr–Ni austenitic alloys. It is known that void swelling and microstructure are dependent on Ni content in Fe–Cr–Ni ternary alloys irradiated with fast neutrons [1–3], electrons [4,5] and nickel-ions [6,7]. Void number density, as well as swelling, generally decrease with increasing Ni content up to about 30 wt% in Fe–15Cr–xNi alloys. Some explanations [3,8] for the decrease in void number density have been proposed on the basis of an increase in vacancy diffusivity with increasing Ni content.

It has been well-recognized that helium (He) significantly modifies void swelling and microstructure. The preinjected He was reported to be effective in suppressing the Ni influence on swelling under 1 MeV electrons at 773 K [5], although no significant effect of the preinjected He was observed at 873–973 K in another case of 1 MeV electron irradiation [6]. The effect of gaseous elements on swelling have been considered as an important issue for fusion reactor first wall materials, because the 14-MeV neutrons will produce gaseous products from (n, α) and (n, p) transmutation reaction. Recent studies have reported that not only He but also hydrogen (H) may influence microstructure development in cases of Ni-ion [9] and electron [10] irradiation.

In the present experiment, void microstructure and swelling were measured by transmission electron microscope (TEM) for Fe–15Cr–xNi ($x = 20$, 25, 30 wt%) alloys irradiated with 180-keV protons at temperatures from 723 to 873 K. The effect of the introduced H on void microstructure will be discussed from a comparison between the alloys with and without the preinjected 10 appm He. The main objective is to investigate the Ni influence on swelling under the condition with not only the preinjected He but also a high level of the introduced H during the irradiation.

2. Experimental procedure

The materials used in this study were Fe–15Cr–xNi ($x = 20$, 25, 30 wt%) ternary alloys. The chemical compositions of the alloys are shown in Table 1. Cold-rolled

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PII S0022-3115(98)00016-6

Table 1
Chemical composition of Fe–15Cr–xNi alloy (wt%)

Alloys	Cr	Ni	C	Si	Mn	P	S	N	O	Cu	Fe
20Ni	15.04	20.12	0.0061	0.0020	< 0.001	0.0009	0.0020	0.0017	0.0087	< 0.001	bal.
25Ni	14.92	25.09	0.0049	< 0.005	< 0.001	0.0045	0.0022	0.0012	0.0073	< 0.001	bal.
30Ni	14.91	30.21	0.0068	0.008	< 0.001	0.0009	0.0021	0.0015	0.0053	< 0.001	bal.

sheets of the alloys were annealed for 120 s at 1293 K and spark-cut into 3-mm diameter disks. Surface layer of the disk was removed by mechanical grinding and electropolishing, and the final thickness was about 120 μm at the central area of the disks prior to the He-preinjection. Grain size and dislocation density of the unirradiated disks were about 10 μm and 1×10^{13} m^{-2}, respectively. Disk specimens were irradiated with 50–200 keV He^{+} at room temperature so that a He level of about 10 appm was distributed uniformly in the range of 200–600 nm. The total injected He^{+} ions at 50, 70, 100, 150 and 200 keV were 3.0, 4.8, 6.8, 9.8 and 9.8×10^{16} ions/m^2, respectively. The displacement damage level estimated with the TRIM-85 code was 0.01 displacement per atom (dpa) in this depth range. After the He preinjection, the specimens were irradiated with 180-keV protons at temperatures from 723 to 873 K. The dose levels were 5, 10 and 20 dpa for 20Ni and 25Ni alloys with He, 10 dpa for the He-preinjected 30Ni alloy, and 5 dpa for 20Ni and 25Ni alloys without He. Fig. 1 shows the profiles of displacement damage and total injected H concentration in 20Ni alloy calculated with the TRIM-85 code. The profiles are quite similar in all three alloys. The amount of implanted H was 6.9, 13.8 and 27.5×10^{22} ions/m^2, corresponding to 5, 10 and 20 dpa at a depth of 400 nm from the irradiated surface, respectively. Displacement damage rate was 2.0×10^{-4} dpa/s at 400 nm in depth, and the rate varies less than ±30% at the depth ranging from 300 to 500 nm from the irradiated surface. The materials were removed by electropolishing from the irradiated surface by 300-nm thick in order to avoid the surface effect on microstructure development, and then the disks were backside thinned to perforation for transmission electron microscope (TEM) observation. The TEM observation was performed in the foil area of 100 to 200-nm thick. Foil thickness was evaluated by a stereo-graphical technique [11]. When the foil area with more than 200 nm in thickness was obtained due to the coarsening of void microstructure, data calibration was carried out by re-counting voids in the area of 200-nm thick with addressing the spatial void distribution.

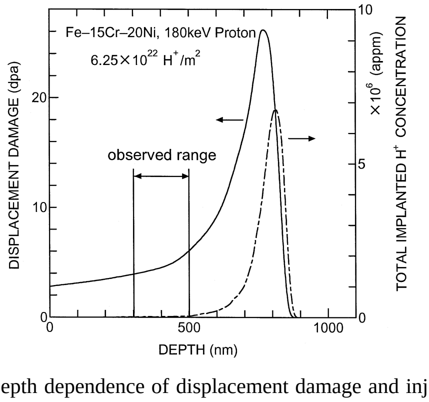


Fig. 1. Depth dependence of displacement damage and injected H concentration in 20Ni alloy calculated with the TRIM-85 code.

Substantial amount of H is introduced to the alloys during the proton irradiation. Since the implanted H diffuses over the entire range and escapes from the surfaces, the H concentration reaches to the saturation during the irradiation. The saturated value is estimated as about 10-appm H at a depth of 400 nm from the irradiated surface, applying to the simplified models for ion permeation at steady state [12]. Actual H concentration, however, appears to be higher than the estimated value due to the interaction with cavities and He. In particular, the preinjected He would increase the H concentration by one or two orders of magnitude in the present proton irradiation, because Keefer and Pard [13] have reported about 1000 appm of H content in the He-preinjected 316 SS after proton irradiation at 773 K.

The observed range in the present experiment was selected at the depth of 300–500 nm from the irradiated surface, and was distant by about 300 nm from the proton range (780 nm). The redistribution of alloy composition in the direction of depth has been investigated by Kimoto and Shiraishi [14] for nominal Fe–15Cr–25Ni alloy irradiated with 200 keV protons at 873 K. The Ni depletion was observed in the regions within about 200 nm of the peak damage position as well as within 300 nm from the irradiated surface. Accordingly, the effect of Ni depletion could be eliminated in the present observed range.

3. Results

Typical void microstructures in the He-free and He-preinjected specimens at 5 dpa are shown in Fig. 2. Fig. 3 shows the Ni dependence on void microstructure in the He preinjected specimens at 10 dpa. The data of the He-prein-

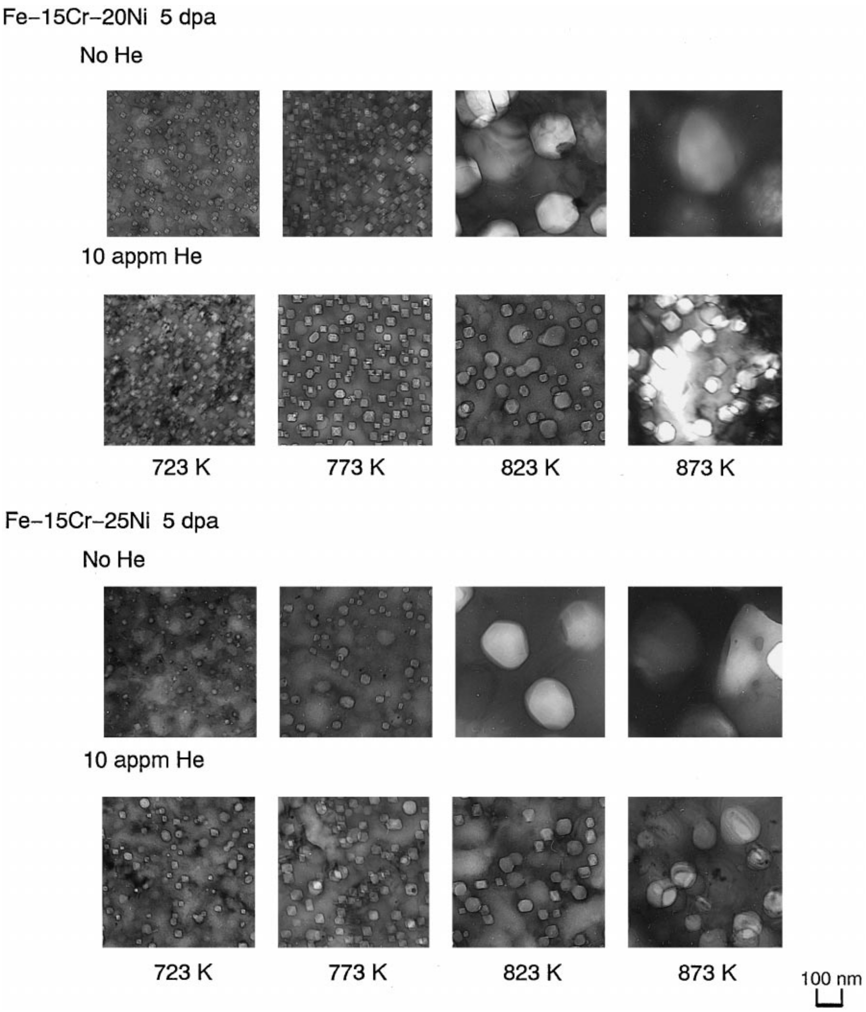


Fig. 2. Typical void microstructures in Fe–15Cr–20Ni and Fe–15Cr–25Ni alloys at 5 dpa with and without He.

jected 20Ni alloy at 10 dpa and 723 K were not available, because the thin foil area was accidentally destroyed. Voids and dislocation network were observed in all the alloys examined by TEM, but faulted dislocation loops were not found even at 5 dpa. Larger voids over 200 nm in diameter were observed not only in the He-preinjected 25Ni alloy at 20 dpa and 873 K but also in 20Ni and 25Ni alloys without He at 5 dpa and 873 K. Since a substantial number of voids were not distinguished clearly due to the coarsening of void microstructure, quantitative TEM measurements were not carried out in such conditions. Results of quantitative TEM observation on microstructure in the He-free alloys and the He-preinjected alloys are summarized in Tables 2 and 3, respectively.

Void microstructure was compared between the alloys with and without He at dose level of 5 dpa. Fig. 4 shows the temperature dependence of (a) average void diameter, (b) void number density and (c) swelling in both 20Ni and 25Ni alloys with and without He. Similar trend of He effect on void nucleation and growth was observed in both alloys. Although void microstructure was similar in specimens with and without He at 723 and 773 K, larger and fewer voids were observed in the He-free specimens for both alloys at 823 K (Fig. 4a,b). As shown in Fig. 4c, the amount of swelling in the He-preinjected specimen was higher than that in the He-free specimen for both alloys at 723 and 773 K. At 823 K, the preinjected He significantly decreased swelling for 20Ni alloy, but increased swelling for 25Ni alloy. The amount of swelling was similar in both alloys with He at all the temperatures, while higher amount of swelling was observed in the lower Ni alloy (20Ni) at 823 K under the He-free condition.

Microstructure development with dose was summarized in the He-preinjected condition. Fig. 5 shows the dose dependence of (a) average void diameter, (b) void number density and (c) swelling for the He-preinjected 20Ni and

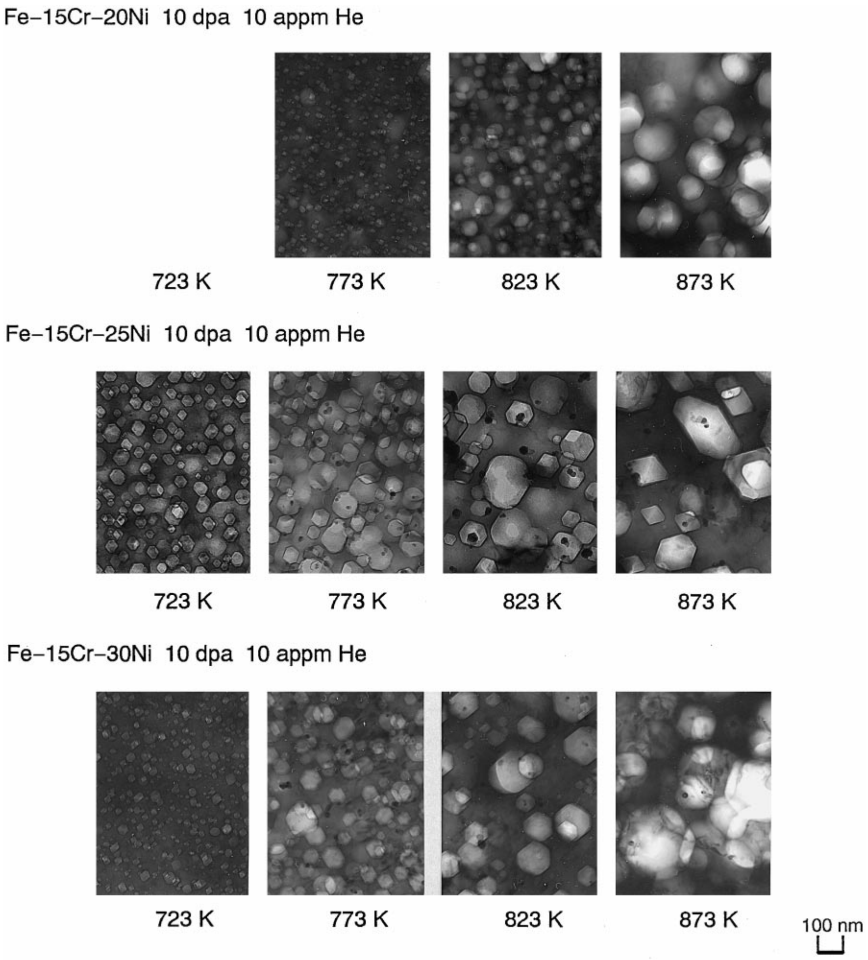


Fig. 3. Typical void microstructures in the He-preinjected Fe–15Cr–xNi ($x = 20$, 25, 30 wt%) alloys at 10 dpa.

Table 2
Microstructure and swelling data of 20Ni and 25Ni alloys without He

Irradiation temperature (K)	Dose (dpa)	Swelling (%)	Average void diameter (nm)	Void number density ($\times 10^{21}$ m^{-3})	Dislocation density ($\times 10^{14}$ m^{-2})
Fe–15Cr–20Ni; no He					
723	5	3.4	21	6.3	4.1
773	5	4.1	36	1.5	2.2
823	5	18.9	175	0.046	0.56
873	5	–	–	–	–
Fe–15Cr–25Ni; no He					
723	5	1.1	18	2.5	2.6
773	5	2.5	37	0.73	1.7
823	5	6.4	176	0.022	0.32
873	5	–	–	–	–

–: Reliable data were not available because the thin foil of 200-nm thick necessary for TEM observation was not obtained by using the electropolishing apparatus.

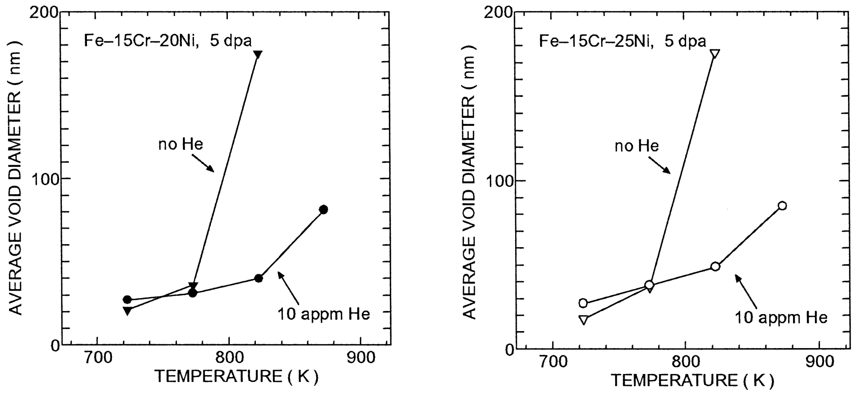
Table 3
Microstructure and swelling data of Fe–15Cr–xNi ($x = 20$, 25, 30 wt%) alloys with He

Irradiation temperature (K)	Dose (dpa)	Swelling (%)	Average void diameter (nm)	Void number density ($\times 10^{21}$ m^{-3})	Dislocation density ($\times 10^{14}$ m^{-3})
Fe–15Cr–20Ni; 10 appm He					
723	5	5.1	27	4.2	3.8
	10				
	20	11.4	31	4.3	6.8
773	5	5.2	31	2.6	1.8
	10	9.0	34	4.0	2.0
	20	16.8	64	0.78	2.4
823	5	7.7	40	1.6	0.90
	10	25.7	62	1.3	1.8
	20	46.7	83	0.42	2.0
873	5	11.9	81	0.30	0.83
	10	24.8	105	0.23	1.2
	20	37.7	115	0.25	1.4
Fe–15Cr–25Ni; 10 appm He					
723	5	3.2	27	2.7	3.1
	10	6.7	31	3.0	3.9
	20	12.0	42	1.9	7.1
773	5	5.7	38	1.4	1.7
	10	9.8	61	0.59	2.3
	20	17.2	104	0.17	2.4
823	5	9.9	49	0.96	1.4
	10	22.0	99	0.31	2.0
	20	38.0	122	0.17	2.7
873	5	12.2	85	0.25	0.68
	10	21.0	135	0.093	0.99
	20	−	−	−	−
Fe–15Cr–30Ni; 10 appm He					
723	10	5.1	23	3.3	3.5
773	10	10.8	59	0.67	1.5
823	10	18.9	109	0.19	1.4
873	10	16.5	146	0.074	0.70

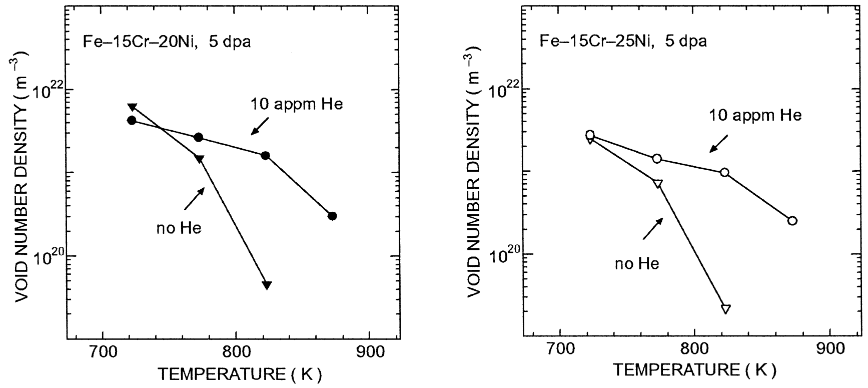
−: Reliable data were not available because the thin foil of 200-nm thick necessary for TEM observation was not obtained by using the electropolishing apparatus.

25Ni alloys. Rapid void growth was observed at the higher temperatures (823, 873 K) for both alloys (Fig. 5a). Void growth rate in 25Ni alloy was higher than that in 20Ni alloy at all the temperatures. Void number density decreased with increasing dose from 5 to 20 dpa for both alloys at almost all the temperatures except in 20Ni alloy at 723 K, as seen in Fig. 5b. This indicates that void nucleation was almost complete at 5 dpa and void density decreased due to the void coalescence at dose levels above 5 dpa. Fewer voids was observed in 25Ni alloy at 5 dpa, and this difference increased with dose due to more frequent void coalescence in 25Ni alloy, especially at 823 and 873 K (Fig. 5b). The plotting of swelling with dose, shown in Fig. 5c, appears to be running through the origin for both alloys at all the temperatures. Swelling rate was similar in both alloys at the lower temperatures (723, 773 K), but the slightly higher rate of swelling was observed in the lower Ni alloy (20Ni) at the higher temperatures (823, 873 K).

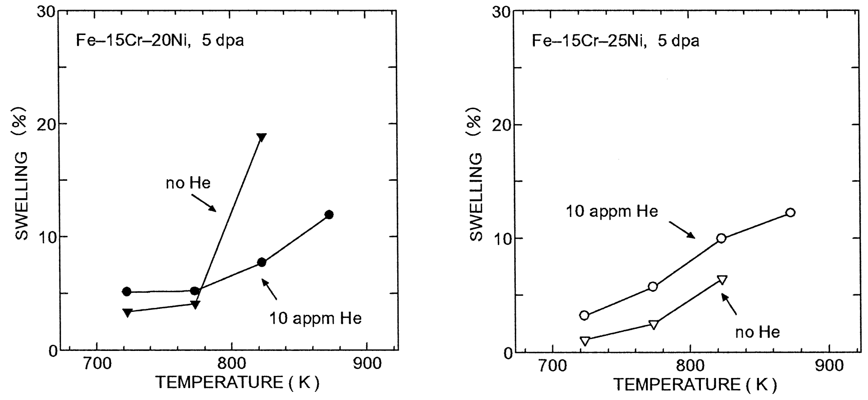
Effect of Ni content on microstructure development was examined for the He-preinjected alloys, as shown in Fig. 2. Fig. 6 summarizes the Ni dependence of (a) average void diameter, (b) void number density and (c) swelling in the He preinjected Fe–15Cr–xNi ($x = 20$, 25, 30 wt%) alloys irradiated to the level of 10 dpa. Although larger and fewer voids were observed in 25Ni alloy as compared with 20Ni alloy, the difference in average void diameter and void number density between 25Ni and 30Ni alloys was less pronounced at all the temperatures (Fig. 6a,b). The amount of swelling shown in Fig. 6c appeared to be little dependent on Ni content at the lower temperatures (723, 773 K), while swelling slightly decreased with increasing Ni content in the range from 20 to 30 wt% at the higher temperatures (823, 873 K).



(a) average void diameter

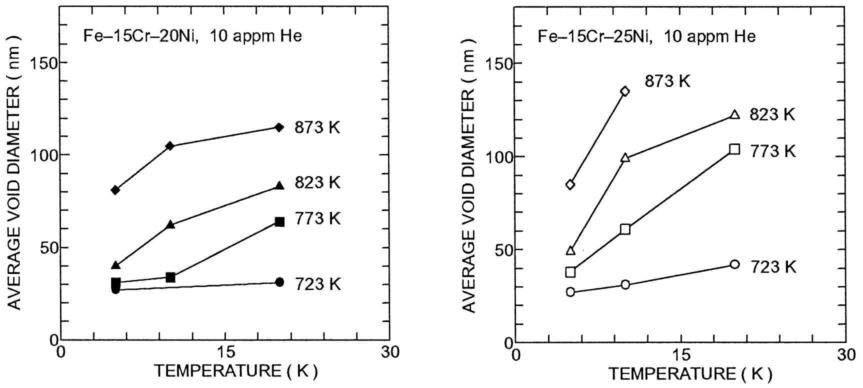


(b) void number density

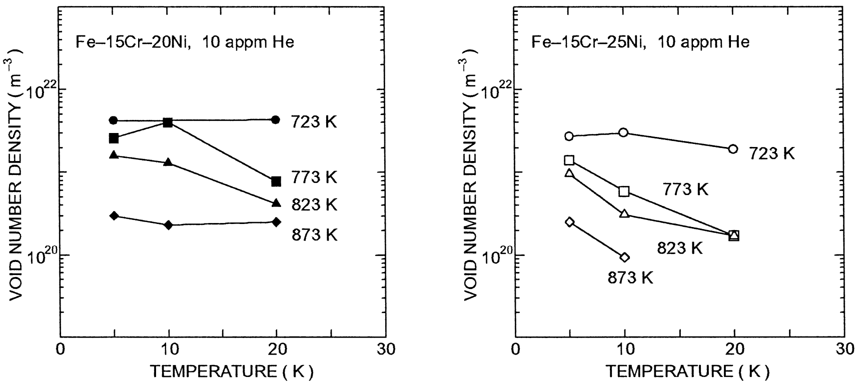


(c) swelling

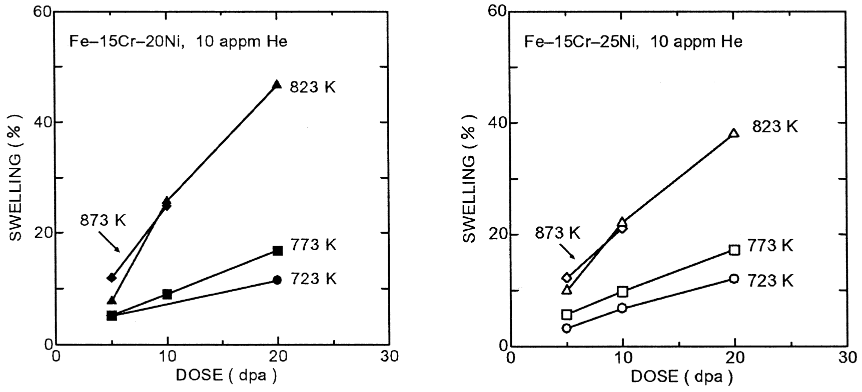
Fig. 4. Temperature dependence of (a) average void diameter, (b) void number density, (c) swelling in 20Ni and 25Ni alloys with and without He.



(a) average void diameter

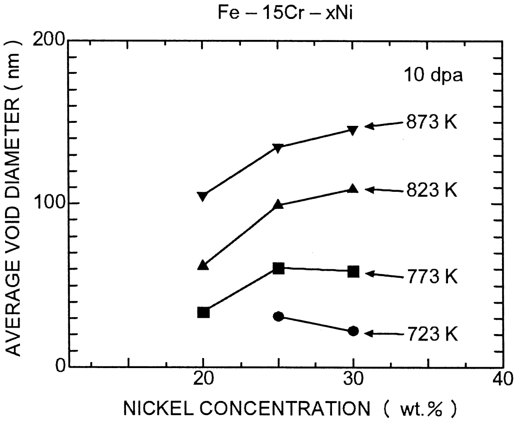


(b) void number density

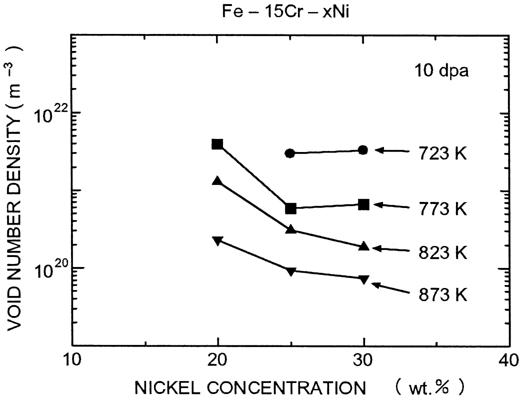


(c) swelling

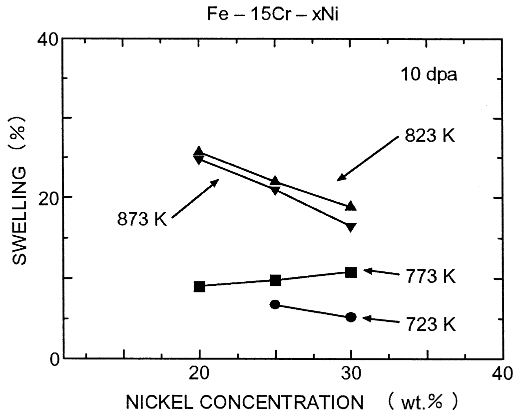
Fig. 5. Dose dependence of (a) average void diameter, (b) void number density, (c) swelling in the He-preinjected 20Ni and 25Ni alloys.



(a) average void diameter



(b) void number density



(c) swelling

Fig. 6. Nickel dependence of (a) average void diameter, (b) void number density, (c) swelling in the He-preinjected Fe–15Cr–xNi ($x = 20$, 25, 30 wt%) alloys at 10 dpa.

4. Discussion

4.1. H effect

It is well-known that void nucleation is strongly promoted by He. The present results showed that the preinjected 10 appm He increased void number density for both 20Ni and 25Ni alloys at 823 K, as presented in Fig. 4a. At the lower temperatures (723, 773 K), however, void microstructure was similar in specimens with and without He. This implies that another factor different from He is working to control microstructure development at such lower temperatures. Recent studies [9,10] suggested that H can also influence void microstructure. Bullen et al. [9] showed an increase in void number density in pure Ni irradiated with 14-MeV Ni-ions at 798 K when H was preinjected to a fluence of 1.5×10^{19} H_3^+/m^2 at room temperature. Ohnuki et al. [10] examined void microstructure in Ti-modified 316 steel (JPCA-2) preinjected with H to a fluence of 1×10^{22} H^+/m^2 at room temperature prior to 1-MeV electron irradiation. They reported that the void number density increased at temperatures from 720 to 770 K in H-preinjected specimens but voids were not observed at 820 K in specimens with and without H preinjection. These results would be asserted to the present proton irradiation. The dose level of 5 dpa was corresponding to a fluence of 6.9×10^{22} H^+/m^2, and at least about 10 appm H was expected at the observed range even in the He-free specimens during the irradiation. The introduced H may play the same role as He in enhancing void nucleation at the temperatures (723, 773 K), because void microstructure was similar in specimens with and without He (Fig. 4b). The drastic decrease of the void number density in the He-free specimens at 823 K appears to be associated with less significant role of H in enhancing void nucleation than that of He at the high temperatures (823, 873 K). The H, however, was considered to promote void growth in the presence of He through the interaction with He-vacancy clusters at 873 K [15]. Although the H effect cannot be clarified from the present results at 823 K, the introduced H may play some role in microstructure development, especially for the He-preinjected specimens, even at the higher temperatures.

4.2. Ni influence

A cross comparison shown in Fig. 4c presents the swelling behavior with and without He for both 20Ni and 25Ni alloys at 5 dpa. The preinjected He slightly increased swelling in 20Ni alloy at 723 and 773 K but decreased swelling at 823 K, while the preinjected He increased swelling in 25Ni alloy at all three temperatures. Since the amount of swelling was similar in both alloys with He at all the temperatures, the preinjected He appeared to reduce the Ni influence on swelling at a dose levels of 5 dpa. However, at the higher dose levels (10, 20 dpa), swelling in the He-preinjected 20Ni alloy was higher than that in the He-preinjected 25Ni alloy at the higher temperatures (823, 873 K), as shown in Fig. 5c. Fig. 6c shows that the Ni influence on swelling in the He-preinjected (20, 25, 30Ni) alloys at 10 dpa. The Ni influence was not pronounced at the lower temperatures (723, 773 K), whereas gradual decrease of swelling with increasing Ni content was observed at the higher temperatures (823, 873 K). These results will be discussed, especially in the light of the Ni influence on swelling under the He-preinjected condition and in comparison with other irradiation results.

The role of the preinjected He in controlling the Ni influence on swelling has been reported in the 1-MeV electron irradiation conducted by Rotman and Dimitrov [5]. They examined void swelling of Fe–16Cr–xNi ($x=$ 20, 25, 45, 75 wt%) ternary alloys under 1-MeV electron irradiation in HVEM at 643–773 K. The He preinjection was carried out to the level of 70 appm at room temperature. As for the He-free alloys, very significant increase in the incubation dose from 3.6 dpa for 20Ni alloy to 90 dpa for 45Ni alloy was observed at 773 K, and the swelling rate decreased drastically with increasing Ni content. However, in the He-preinjected alloys, no incubation was observed and the stationary swelling rate was similar in the Ni range of 20–45 wt% at 773 K. They concluded that the preinjected He strongly suppressed the Ni-dependent incubation dose and swelling rate, namely the Ni influence on swelling at 773 K.

A wider temperature range (673–973 K) was adapted in another electron irradiation [4]. Walters has performed 1-MeV electron irradiation in HVEM at 673–973 K for Fe–17Cr–xNi ($x=$ 15, 25, 40, 50, 60 wt%) ternary alloys with and without 10 appm He preinjection. The preinjected He gave lower incubation doses in the alloys with Ni content of 25–60 wt% at temperatures up to 823 K, but had no effect at 873–973 K. There were some data inconsistent with Rotmans' results in the lower range of Ni content. The incubation dose was similar in both He-free and He-preinjected alloys in the Ni range of 15–25 wt% for all the irradiation temperatures. A possible origin of the difference between Walters' and Rotman's data was discussed in Rotman's paper [5]. One of the causes would be resulted from the larger amount of gas impurities (nitrogen and oxygen) by one or two order of magnitude in Walters' alloy than that in Rotman's alloy.

In the present proton irradiation, the amounts of gas impurities shown in Table 1 were as low as a fourth or tenth of that in Walters' alloys [4], and the effect of gas impurities on microstructure development could be considered as minimal. The amount of swelling was little dependent on Ni content at lower temperatures (723, 773 K), as presented in Fig. 6c. The plotting of swelling as a function of dpa shown in Fig. 5c implies the absence of incubation period prior to the onset of steady swelling and similar swelling rate in both the He-preinjected 20Ni and 25Ni alloys at 723 and 773 K. This is the same as in the electron irradiation conducted by Rotman and Dimitrov [5], and the existence of He prior to the irradiation appears to suppress the Ni dependence of incubation dose and swelling rate at the lower temperatures (723, 773 K). Therefore, it is suggested that the preinjected He suppresses the Ni influence on swelling at the lower temperatures. However, at the higher temperatures (823, 873 K), swelling slightly decreased with increasing Ni content (Fig. 6c). Since the Ni-dependent incubation revived at temperatures above 823 K in the electron irradiation performed by Walters [4], the Ni content would be an important factor to control swelling at such higher temperatures. Although the dose dependence of swelling (Fig. 5c) indicates no influence of Ni on incubation dose even at the higher temperatures (823, 873 K), the Ni-dependent swelling rate may be responsible for the revival of the Ni influence on swelling.

Dominant Ni influence as well as no remarkable He effect on swelling has been reported for Fe–15Cr–xNi ($x = 20$, 25 wt%) ternary alloys irradiated with 2.8–5-MeV Ni ions at 948–973 K [16]. The amount of swelling in Fe–15Cr–20Ni alloy at 948 K was insensitive to the presence or absence of the preinjected He up to the level of 20 appm. The increase from 20 to 25 wt% in Ni content caused a pronounced increase of the incubation period, while the preinjected He had no effect on the incubation dose.

It is noted that more detailed assessment like the difference in cascade production is definitely needed to clarify the role of the preinjected He in controlling the Ni influence on swelling between the proton and other particle irradiations. Furthermore, the synergistic effect of He and H is an important problem to be clarified in the proton irradiation, because the role of He may be modified by the introduced H, especially at higher temperatures. However, in this study, it is generally concluded that the preinjected He can suppress the Ni influence on swelling at the lower temperatures. The Ni influence on swelling would revive at temperatures above 823 K in the present proton irradiation.

5. Conclusion

The 180-keV proton irradiation to 5, 10 and 20 dpa was conducted on solution annealed Fe–15Cr–xNi ($x = 20$, 25, 30 wt%) ternary alloys with and without 10 appm He in the temperature range of 723–873 K. The results of the TEM measurement can be summarized as follows: (1) void microstructure was similar in specimens with and without He for both alloys at 723 and 773 K, although the drastic decrease of void number density was observed in the He-free specimens at 823 K. This indicates that the introduced H during the proton irradiation plays the same role as He in enhancing void nucleation at 723 and 773 K. (2)

In the He-preinjected alloys, swelling was little dependent on Ni content at the lower temperatures (723, 773 K), while gradual decrease of swelling with increasing Ni content was observed at the higher temperatures (823, 873 K). Although the preinjected He appears to suppress the Ni influence on swelling at the lower temperatures, the Ni influence may revive at the higher temperatures.

Acknowledgements

The authors wish to thank Dr I. Shibahara of PNC for the alloy fabrication and chemical analysis.

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