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Void denuded zone formation for Fe–15Cr–15Ni steel and PNC316 stainless steel under neutron and electron irradiations

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HIGHLIGHTS

- Austenitic stainless steel developed to improve void swelling was used.
- Void denuded zone formed near grain boundary can be affected by vacancy mobility.
- Vacancy migration energy was estimated from void denuded zone width in the steel.

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ABSTRACT

Irradiation-induced void denuded zone (VDZ) formation near grain boundaries was studied to clarify the effects of minor alloying elements on vacancy diffusivity during irradiation in practical PNC316 stainless steel developed for nuclear reactor core materials. The test materials were Fe–15Cr–15Ni steel without additives and PNC316 stainless steel; the latter contains minor alloying elements to improve the void swelling resistance. These steels were neutron-irradiated in the experimental fast reactor JOYO at temperatures from 749 K to 775 K and fast neutron doses of 18–103 dpa, and electron irradiation was also carried out using 1 MeV high voltage electron microscopy at temperatures of 723 K and 773 K and doses up to 14.4 dpa. VDZ formation was analyzed by TEM microstructural observation after irradiation by considering radiation-induced segregation near the grain boundaries.

VDZs were formed near random grain boundaries with higher misfit angles in both Fe–15Cr–15Ni and PNC316 steels. The VDZ widths in the PNC316 stainless steel were narrower than those for the Fe–15Cr–15Ni steel for all neutron and electron irradiations. The VDZ width analysis implied that the vacancy diffusivity was reduced in PNC316 stainless steel as a result of interaction of vacancies with minor alloying elements.

1. Introduction

A large number of theoretical and experimental investigations on irradiation-induced evolution of dislocations and voids have been carried out to develop high-performance nuclear reactor core materials. These studies have focused mainly on understanding the fundamental behavior of point defects (vacancies and self-interstitial atoms) induced by higher energy particles [1–6]. As a consequence of point defect accumulation, irradiation effects such as void swelling [1,3,7] and radiation hardening [8,9] occur, which affect the performance of nuclear materials during long-term neutron irradiation in service. To suppress void swelling, minor elements can be added to a base alloy; this approach extends the incubation period required to reach a steady state of void swelling, and thus reduces the extent of void swelling [10,11]. This result is achieved by a change in point defect diffusivity resulting from interactions between point defects and added elements.

Polycrystalline materials are composed of random grain boundaries (GBs), which act as strong neutral sink sites for point defects. When such materials are irradiated, the point defect concentration distributions near the random GBs differ from those in the matrix because the excess point defects flow toward the GBs [12]. These differences in point defect concentration mean that the point defect accumulation behavior near the GBs will differ from that in the matrix, and result in different radiation-induced

Abbreviations: VDZ, void denuded zone; HVEM, high voltage electron microscope; GB, grain boundary; CSLB, coincidence site lattice boundary; RIS, radiation induced segregation.

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phenomena. Denuded zones, where defects such as dislocation loops and voids do not accumulate [12–18], are induced near the random GBs. At the random GBs, radiation induced segregation (RIS) of solutes also occurs [2,19].

In particular, the width of the void denuded zone (VDZ) formed near the GBs will be influenced by factors which affect vacancy diffusion [14,16,17]. Thus, it is expected that the changes of vacancy diffusion caused by the interactions between the added minor elements and irradiation-induced vacancies will alter the VDZ widths, as indicated in the authors' previous research [18]. However, there has been little research on VDZ width with the objective of investigating the effects of minor alloying elements on vacancy diffusivity, especially for actual steels in use. To clarify the effects for these steels are very important for future alloying design and specification of nuclear core materials, because it would allow us to evaluate the effects of minor alloying elements more accurately and could suggest the most appropriate minor elements to suppress void swelling.

In the present study, the VDZ widths near random GBs were analyzed by microstructural observations of neutron-irradiated and electron-irradiated steels to clarify the effects of minor alloying elements in practical PNC316 stainless steel (modified stainless steel). The widths in the PNC316 stainless steel were compared to those in Fe–15Cr–15Ni steel (without additives) and the vacancy diffusivity in the PNC316 stainless steel was evaluated.

2. Experimental

Polycrystalline Fe–15Cr–15Ni steel and PNC316 stainless steel were prepared. The chemical compositions are given in Table 1. The PNC316 steel used was a modified 316 stainless steel containing additional elements such as P, Ti, B, and Si, and was developed to improve void swelling resistance [20]. Some of the PNC316 steel used in this study was cold-worked by 20% (to get PNC316CW steel). The Fe–15Cr–15Ni and PNC316CW steels were irradiated in the experimental fast reactor JOYO using the Core Material Irradiation Rig (CMIR): for the Fe–15Cr–15Ni steel, the irradiation temperature was 749 K and the fast neutron irradiation dose was 18 dpa, while for the PNC316CW steel, the irradiation was carried out at 775 K to a fast neutron dose of 103 dpa. These neutron irradiation conditions are collated in Table 2. On the other hand, foils of Fe–15Cr–15Ni steel and solution-annealed PNC316 steel (PNC316SA steel) were prepared by electro-polishing and were then electron-irradiated at 1 MeV using a high voltage electron microscope (HVEM). The electron irradiations were carried out with a displacement rate of approximately 2.0×10^{-3} dpa/s for the Fe–15Cr–15Ni steel at 723 K or 773 K to 10.8 dpa, and for the PNC316SA steel at 723 K to 14.4 dpa (Table 2). The electron irradiation areas were chosen to include random GBs, as identified previously from selected area electron diffraction patterns. All irradiated areas had a foil thickness greater than 400 nm, to minimize the effect of the surface sink.

For steels irradiated by neutrons or electrons, transmission electron microscope (TEM) observations were carried out focusing the void distributions near the random GBs, and the widths of VDZ were measured. In the present study, the VDZ width was determined as the average distance between the GB position and voids closest to the GB. The average VDZ width in one steel was estimated from widths near three random GBs.

3. Experimental results

3.1. VDZ formation after neutron irradiation

Fig. 1(a) shows a typical void distribution observed near a random GB with a high misfit angle in the Fe–15Cr–15Ni steel, which was neutron-irradiated to 18 dpa at 749 K. An area of no void formation was observed near the random GB, namely the VDZ was formed. The white solid and black dotted lines in Fig. 1(a) indicate the random GB and VDZ width, respectively. The average VDZ width of this steel was determined to be 141 ± 14 nm, and it was confirmed that the standard deviation was not large. On the other hand, a typical void distribution formed near a coincidence site lattice boundary (CSLB) in this steel is shown in Fig. 1(b). Voids formed near the CSLB were homogeneously distributed over the whole area. Thus, it was confirmed that the VDZs were formed preferentially near random GBs. This suggests that the excess vacancy concentration introduced during irradiation was lowered to the critical concentration level to form voids near random GBs, because excess vacancies induced near the GB could flow toward the GB and be absorbed at the GB. On the other hand, no VDZ formation near the CSLB would be caused by less vacancy flow toward the CSLB which has low sink strength. Therefore, in order to observe VDZ formation, void distribution near a random GB with a high misfit angle must be observed rather than near a CSLB with a low misfit angle.

VDZ formation was also observed near the random GB in the PNC316CW steel that was neutron-irradiated to 104 dpa at 775 K; in this case the average VDZ width was about 62 ± 3 nm (Fig. 2). This width was narrower than that of the Fe–15Cr–15Ni steel, although the PNC316CW specimen was irradiated with a larger dose at a higher irradiation temperature. Garner [4] reported that the numbers of dislocations introduced by cold work were reduced during neutron irradiation to higher doses, and voids would be nucleated only after decreasing the dislocation density. Therefore, it is considered that the void distribution and VDZ formation in the PNC316CW steel could only be observed after weakening the dislocation effect (which extends the incubation period of void swelling).

3.2. VDZ formation and segregation near GBs under electron irradiation

To follow the VDZ formation behavior observed in neutron-irradiated steels, steels of the same types as used in the neutron irradiation experiments were electron-irradiated at irradiation conditions capable of inducing void distributions, and their VDZ formation was investigated in combination with RIS behavior.

Fig. 3(a) and (b) shows the void distributions with VDZ formation in the Fe–15Cr–15Ni steel after electron irradiation at 723 K or 773 K to 10.8 dpa. VDZ formation was observed clearly in the grains along both sides of the random GBs. GB migration often occurred during electron irradiation; however, voids were nucleated during the relatively early stages of irradiation, before the occurrence of GB migration. The initially-nucleated voids

Table 1
Chemical compositions of the Fe–15Cr–15Ni steel and the PNC316 stainless steel.

	Chemical composition (wt.%)										
	C	Si	Mn	P	Fe	Cr	Ni	B	Mo	Ti	Nb
Fe–15Cr–15Ni	0.009	<0.005	0.002	0.0006	Bal.	15.14	14.99	–	–	–	–
PNC316	0.051	0.79	1.8	0.026	Bal.	16.41	13.86	0.0038	2.48	0.075	0.074