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Fluence dependence of defect evolution in austenitic stainless steels during fission neutron irradiation

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Abstract

To understand microstructural evolution during fission neutron irradiation, a pure Fe–Cr–Ni ternary alloy, phosphorus-containing model austenitic stainless steels and SUS316 were irradiated in a Japanese Material Testing Reactor (JMTR) at 493 and 613 K. At 493 K, the density of defect cluster increased with the irradiation dose, but there was no significant change in loop density and loop size among all the materials. At 613 K, on the other hand, interstitial type dislocation loops and phosphides were formed in pure ternary and phosphorus-containing alloys, respectively, by an early stage of irradiation. These results suggest that the defect cluster formation at 493 and 613 K is mainly controlled by the cascade damage and long-range migration of free point defects, respectively.

1. Introduction

The authors have earlier studied the fundamental behavior of point defects in Fe–16Cr–17Ni pure ternary alloy and the role of phosphorus atoms in Fe–Cr–Ni–P alloys utilizing a high voltage electron microscopy (HVEM) [1,2]. It is concluded that the nucleation and growth processes of interstitial type dislocation loops were strongly affected by phosphorus additions. In these studies, the interstitial migration energy and binding energy between interstitial and phosphorus were estimated to be 0.9 and 0.5 eV, respectively. Recently, the irradiations with well-controlled conditions, such as irradiation temperature and irradiation time, have become possible at the Japanese Material Testing Reactor (JMTR) by using the technique developed by Kiritani et al. [3,4].

In this study, to understand the fundamental behavior of point defects during fission neutron irradiation, defect cluster formation and radiation-induced phosphide formation of phosphorus-modified stainless steel specimens under JMTR irradiation are examined and compared with the results obtained by the previous HVEM experiments.

2. Experimental procedures

The model stainless steel specimens (Fe–16Cr–17Ni, Fe–16Cr–17Ni–0.024P, Fe–16Cr–17Ni–0.1P, Fe–16Cr–17Ni–0.25Ti–0.1P) and a commercial austenitic stainless steel (SUS316) were used in this study. The model alloys were melted from Johnson–Matthey high purity starting materials in a flowing hydrogen atmosphere. The specimens were solution treated at 1323 K for 1.8 ks and air-cooled.

Fission neutron irradiation was carried out in the JMTR under improved temperature control condition at 493 and 613 K. To study the microstructural evolution, a new irradiation temperature controlled rig, which allows the removal of some specimens out of the reactor, was used [3]. The two specimens were removed at the scheduled irradiation time (40.5 and 188.0 h) and the rest of the specimens were kept until an end of the reactor cycle (591.5 h). The total neutron dose of full cycle irradiation (591.5 h) was $1.03 \times 10^{24}/m^2$ (>1.0 MeV), which corresponds to 0.15 dpa for stainless steels. This experiment made it possible to derive the fluence

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dependence of microstructures at exactly constant temperature and damage rate.

3. Results

Fig. 1 shows the microstructure of all specimens after the end of the reactor cycle (0.15 dpa). The upper and lower photos show the microstructure at 493 and 613 K, respectively. At 493 K, a high density of defect clusters was observed in all materials, without significant difference in loop density. At 613 K, on the other hand, interstitial type dislocation loops were observed in pure ternary alloy. Needle-like phosphides which were oriented along ⟨1 0 0⟩ directions, were observed in phosphorus-containing alloys. However, no visible defect was formed in SUS316.

Fig. 2 shows the microstructural evolution of pure ternary alloy at 493 and 613 K. At 493 K, density of the defect clusters increased with irradiation dose, with slight increase (from 3 to 5 nm) in loop size. At 613 K, on the other hand, the density of interstitial type dislocation loop had saturated at an early stage of irradiation (40.5 h) and the loop size increased with irradiation time. In phosphorus-containing alloy and SUS316, the microstructural evolution of 493 K was almost same as that of pure ternary. But at 613 K, phosphides nucleated and grew instead of dislocation loops in in phosphorus-containing alloys. Figs. 3 and 4 show the measured defect cluster density and size of each specimen at 493 and 613 K, respectively. Since dislocation loops were not formed in phosphorus-containing alloy and SUS316 at 613 K, the measured phosphide density and size at each irradiation dose for these specimens were shown in Fig. 4. Fig. 3 shows that the defect cluster density in all materials increased almost proportional to the square root of the irradiation dose (fluence).

4. Discussions

At 493 K, high number densities of very fine defect clusters are observed. In the pure ternary alloy, they are vacancy type clusters with the shape of stacking fault tetrahedra and interstitial type clusters with the shape of dislocation loops. There are both types of clusters in phosphorous-containing alloys and SUS316. As shown in Fig. 1, defect cluster density at 493 K was not affected by phosphorus additions. Defect density of SUS316 at this temperature was almost the same as that of pure ternary alloy. In our previous HVEM study [1,2], on the other hand, interstitial type dislocation density of phosphorus-containing alloy at this temperature range (below 573 K) was almost 10 times higher than that of pure ternary. Nucleation period of interstitial type dislocation loops was prolonged by phosphorus and other minor solute additions. In the HVEM study, interstitial type dislocation loop density increased linearly with irradiation time in the early stage of irradiation. After this nucleation period, the loop density reached saturation. Therefore it is not likely that the defect cluster observed at 493 K in the present study was formed by agglomeration of free defects. The defect cluster formation due to cascade damage seems to be dominant.

At 613 K, on the other hand, large interstitial type dislocation loops and phosphides were observed. The authors have previously shown that phosphide, oriented along ⟨1 0 0⟩ directions, is not formed during thermal

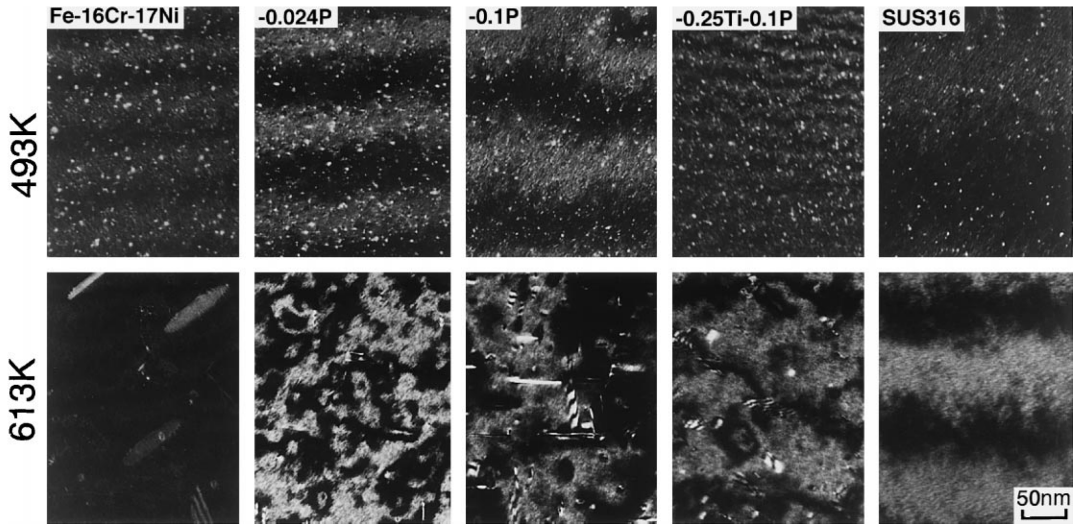

Fig. 1. Weak beam dark field images of microstructure formed by neutron irradiation after one reactor cycle (0.15 dpa). Upper and lower photos show the case of 493 and 613 K, respectively.

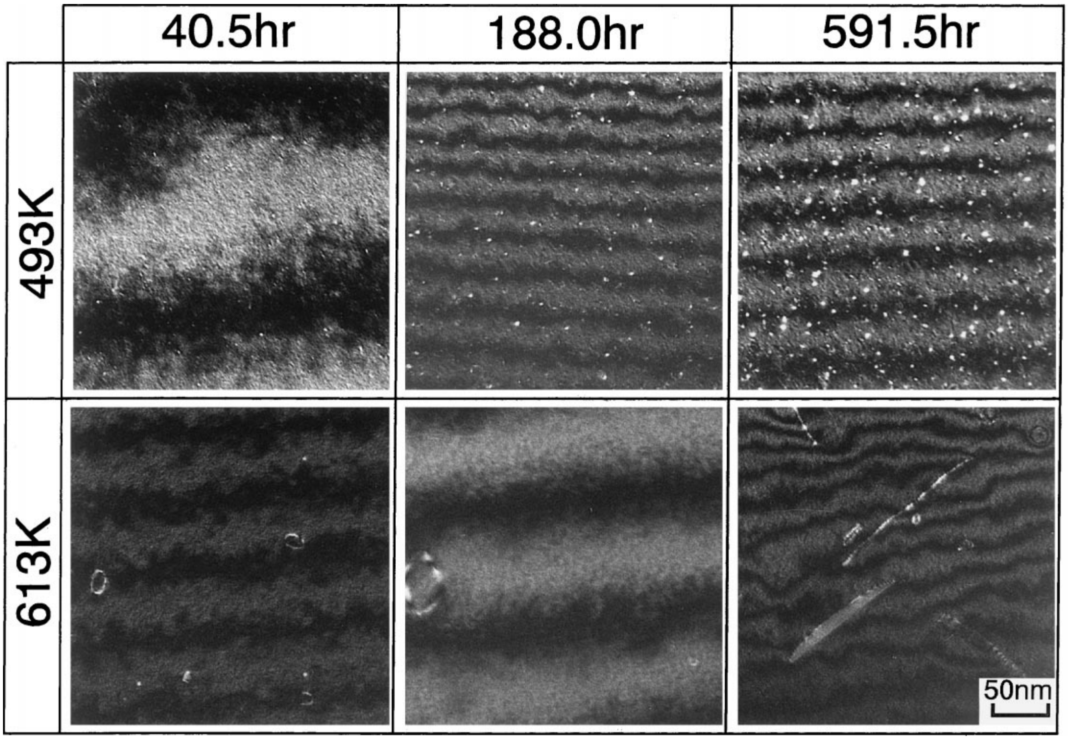

Fig. 2. Microstructural evolution of Fe–16Cr–17Ni during irradiation. Upper and lower photos show the cases of 493 and 613 K, respectively. One cycle irradiation (591.5 h) corresponds to 0.15 dpa for stainless steels.

aging. The formation of radiation-induced phosphides in these specimens are due to a long-range migration of interstitial-phosphorus complex and enrichment at defects sinks [5]. It is also well known that the phosphides are formed in the temperature range where void swelling becomes prominent.

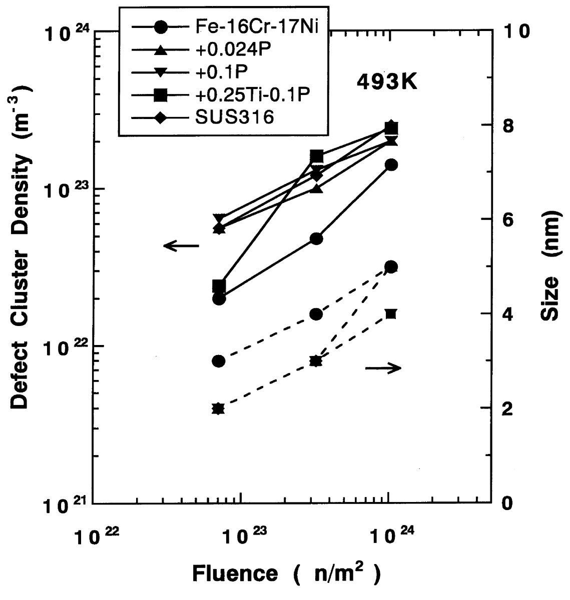

Fig. 3. Measured loop density and size of Fe–16Cr–17Ni, Fe–16Cr–17Ni–0.024P, Fe–16Cr–17Ni–0.1P, Fe–16Cr–17Ni–0.25Ti–0.1P and SUS316 at 493 K.

As discussed in Ref. [6], the main process of interstitial type loop nucleation in Fe–Cr–Ni austenitic alloys at elevated temperatures higher than 730 K, is the reaction of free interstitial. The role of direct nucleation in a cascade region is minor. In this case, the concentration of interstitial loops at saturation (C_L) in Fe–Cr–Ni alloys under irradiation at a constant rate is given as follows [1]:

$$C_L = 2^{1/3}(Z_{i,i}/Z_{L,i})^{2/3}(P/Z_{i,v}M_i)^{1/2}, \quad (1)$$

$$M_i = \nu_0 \exp(-E^i_m/kT), \quad (2)$$

where P and M_i are the defect production rate and the mobility of the interstitial, respectively, Z denotes the number density of the reaction site. For example, $Z_{L,i}$ means the number of reaction sites for dislocation and an interstitial. ν_0, k and T are frequency factor, the Boltzmann constant and temperature, respectively. Eq. (1) shows that the saturated loop density is proportional to $P^{1/2}$, in good agreement with the experimental results of several Fe–Cr–Ni alloys in a wide temperature range. Fig. 5 shows the saturated dislocation loop density of Fe–16Cr–17Ni alloy obtained in various irradiation facilities as a function of calculated

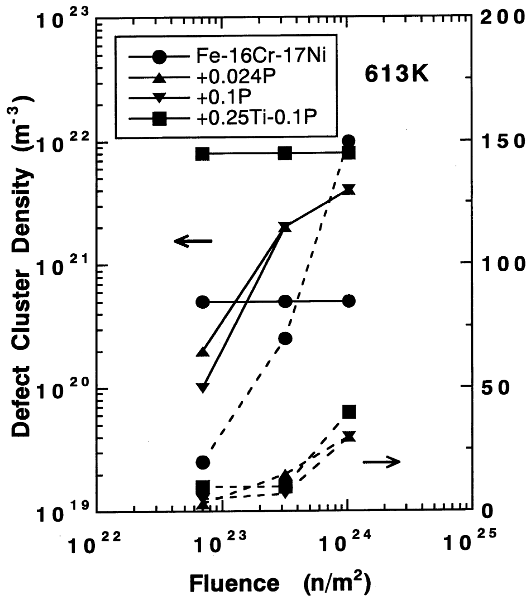

Fig. 4. Measured loop density and size of Fe–16Cr–17Ni and measured phosphide density and size of Fe–16Cr–17Ni–0.024P, Fe–16Cr–17Ni–0.1P and Fe–16Cr–17Ni–0.25Ti–0.1P at 613 K.

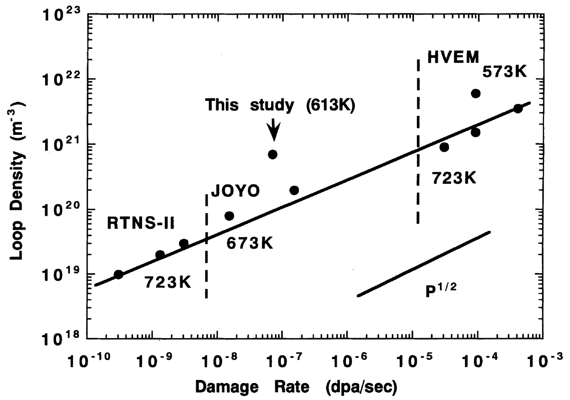

Fig. 5. Damage rate dependence of saturated dislocation loop density. Here, RTNS-II and JOYO are Rotating Target Neutron Sources at the Lawrence Livermore National Laboratory and the Japanese Fast Experimental Reactor at Power Reactor and Nuclear Fuel Development Corporation, respectively.

damage rate [6]. Here, RTNS-II and JOYO are Rotating Target Neutron Source at Lowrence Livermore National Laboratory and Japanese Fast Experimental Reactor at Power Reactor and Nuclear Fuel Development Corporation, respectively. In Fig. 5, measured loop density at 613 K obtained from the present study is also shown. Considering the difference of temperatures, the present study is in good agreement with the previous experimental results. This implies that the present irradiation condition of 613 K belongs to the case of minor cascade effects on loop nucleation.

Recent neutron irradiation studies showed that low-temperature irradiations during the start-up and shut-down procedures of reactors influenced microstructural evolution very much even though the dose is very low [7–9]. In this study, dislocation density of pure ternary alloy at 613 K was saturated even at the dose of 0.01 dpa. This result again showed that the low-dose irradiations during the start-up and shut-down procedures of a reactor are significant in the microstructural evolution of stainless steels.

5. Conclusions

The microstructural evolution of pure ternary, phosphorus-containing alloy and SUS316 during exact temperature and damage rate has been investigated using a new improved temperature controlled rig.

1. The defect cluster formation (loop and phosphide) at 493 and 613 K are controlled by the cascade damage and long-range migration of free point defects, respectively.
2. Dislocation density of pure ternary alloy at 613 K was saturated even at a dose of 0.01 dpa.
3. These results showed that the low-dose irradiations during the start-up and shut-down procedures of reactors is essential for the microstructural evolution of stainless steels.

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