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Influence of variable temperatures irradiation on microstructural evolution in phosphorus doped Fe–Cr–Ni alloys

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Abstract

To evaluate the influence of temperature change on microstructural evolution in phosphorus modified Fe–Cr–Ni alloys, irradiations at 473/923, 673/923, 773/923 K were performed on a Fe–16Cr–17Ni–0.1P alloy with 2.4 MeV Cu ions. In these experiments, specimen temperature was changed in a stepwise manner during the irradiation. In the case of 673/923 K-irradiation, the phosphide density after the second irradiation at 923 K was about 2 orders of magnitude higher than that in the constant temperature irradiation at 923 K. The influence of the first irradiation at lower temperature remained to a higher dose level of 75 dpa. High void swelling of about 12% was observed after varying temperature irradiation. In the constant temperature irradiation, on the other hand, void swelling was strongly suppressed. The influence of the first irradiation was relatively small in 473/923 K-irradiation. Defect clusters formed during the first irradiation at 473 K disappeared during temperature elevation up to 923 K, and the microstructural evolution during the second irradiation at 923 K was similar to that in the constant temperature irradiation.

1. Introduction

In austenitic stainless steels used for in-core materials in fast breeder reactors or proposed for fusion reactor structural materials, void swelling is one of the major problems. The major effort to reduce void swelling has focused on the addition of minor elements, and previous studies showed that phosphorus addition to austenitic stainless steels has a significant effect on suppressing void swelling [1–5].

The effects of phosphorus addition on void swelling can be categorized into those at lower temperature where precipitation is not prominent, and those at higher temperature where the microstructure is dominated by phosphide precipitates [6]. In the lower temperature range, enhanced interstitial loop formation due to phosphorus addition effectively suppresses void swelling. In the higher temperature range, formation of phosphides suppresses void swelling by acting as recombination sites and He trapping sites.

Recent fission neutron irradiation studies showed that temperature variation during irradiation strongly influences the microstructural evolution in various materials including Fe–Cr–Ni austenitic alloys [7–11]. Since temperature variation is also expected in fusion reactors, it is important to investigate the influence of temperature variation on microstructural evolution for realistic estimation of a material's performance in fission reactors. In this study, temperature variation effects on microstructural evolution and void swelling in phosphorus modified Fe–Cr–Ni alloy was studied by heavy ion irradiation.

2. Experimental procedure

The phosphorus modified model stainless steel (Fe–16Cr–17Ni–0.1P) was used in this study. The model

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alloy was melted from Johnson–Matthey high purity starting materials in a flowing hydrogen atmosphere. The specimens, after rolling and punching into TEM discs, were solution treated at 1323 K for 1.8 ks and air-cooled.

The specimens were irradiated with 2.4 MeV Cu^{2+} ions using a Tandem accelerator at the Research Institute for Applied Mechanics, Kyushu University. Irradiations were carried out at constant temperatures of 673, 773, and 923 K and with varying temperatures of 473/923, 673/923, and 773/923 K. In the varying temperature experiment, the first irradiation was made at 473, 673, and 773 K to 1 dpa followed by the second irradiation at 923 K to 75 dpa. After irradiation, the area of peak damage at a depth of around 450 nm was electropolished by a back-thinning method for TEM observation.

3. Results

3.1. Evolution of loops and precipitates

Figs. 1 and 2 show the microstructural evolution and dose dependence of phosphide density and size, respectively, under varying temperature irradiation when the second irradiation was made to 9 dpa. The influence of the first irradiation on microstructural evolution during the second irradiation was relatively small for 473/923 K-irradiation. A high density of defect clusters was formed in the first irradiation at 473 K, but these defect clusters were recovered during temperature elevation and the precipitate behavior of phosphides during the second irradiation was similar to that during constant temperature irradiation at 923 K. The microstructural evolution during the second irradiation was strongly affected by the microstructure formed during the first irradiation in the case of 673/923 and 773/923 K combinations. The high density of defect clusters observed during the first irradiation at 673 and 773 K remained after raising the specimen temperature to 923 K. For these two cases, formation of a high density of phosphides was observed during the second irradiation at 923 K. The precipitate behavior was very different from that of the irradiation at constant temperature of 923 K. The phosphide density formed during the second irradiation was 2 orders of magnitude higher for the 673/923 K-irradiation and was an order of magnitude higher for 773/923 K-irradiation than that for the constant temperature irradiation. The size of the phosphides for the two cases was about an order of magnitude smaller than that for the constant temperature irradiation.

3.2. Void evolution at high dose

Fig. 3 shows the microstructural evolution during constant temperature irradiation at 923 K for high dose

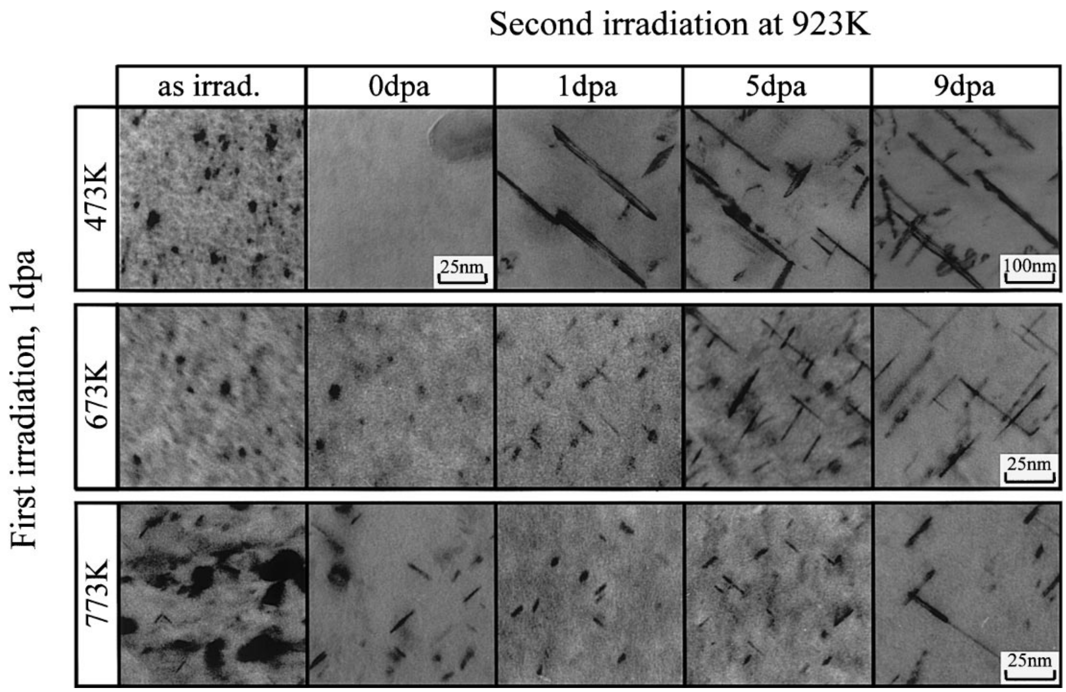

Fig. 1. Microstructural evolution during varying temperature irradiation. The first irradiation was made at 473, 673 and 773 K to 1 dpa and the second irradiation was made at 923 K to 9 dpa.

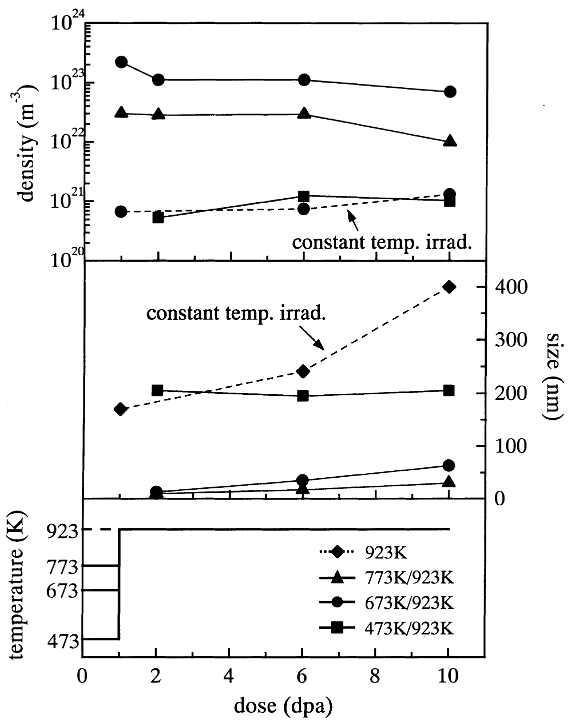

Fig. 2. Dose dependence of the phosphide density and size during varying temperature irradiation.

levels. Phosphides observed in the specimen at 10 dpa were also observed after 150 dpa with the void formation being strongly suppressed. Fig. 4 shows the microstructural evolution during varying temperature irradiation to high dose levels. Fig. 5 shows the dose dependence of phosphide density and void swelling. In the varying temperature irradiation, the first irradiation at lower temperature is influential up to the dose level of 75 dpa for 673/923 and 773/923 K-irradiations. In the case of 673/923 K-irradiation, the density of phosphides decreased rapidly with increasing dose level followed by the formation and growth of interstitial-type loops, and voids started to form after the second irradiation to 50 dpa. At 75 dpa, no phosphides were observed in the specimen and high void swelling of about 12% was observed. In the case of 773/923 K-irradiation, the density of phosphides also decreased with dose but in a less significant sense relative to that for the case of 673/923 K-irradiation. In this case, the phosphides still remained after the second irradiation to 75 dpa and no void formation was observed. In the case of 473/923 K-irradiation, on the other hand, no influence of the first irradiation was observed after the second irradiation to 75 dpa. In this case, void swelling was strongly suppressed in a manner similar to that for the case of constant temperature irradiation at 923 K.

4. Discussion

4.1. Influence of the first irradiation on the phosphide formation

The influence of the first irradiation was relatively small in the case of 473/923 K-irradiation compared to the cases of 673/923 and 773/923 K combinations. The

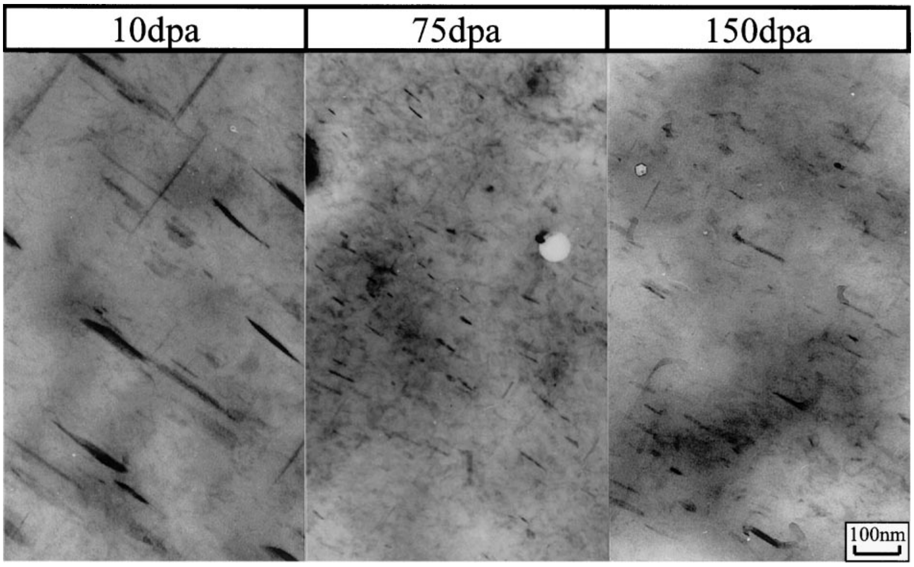

Fig. 3. Microstructural evolution during constant temperature irradiation at 923 K.

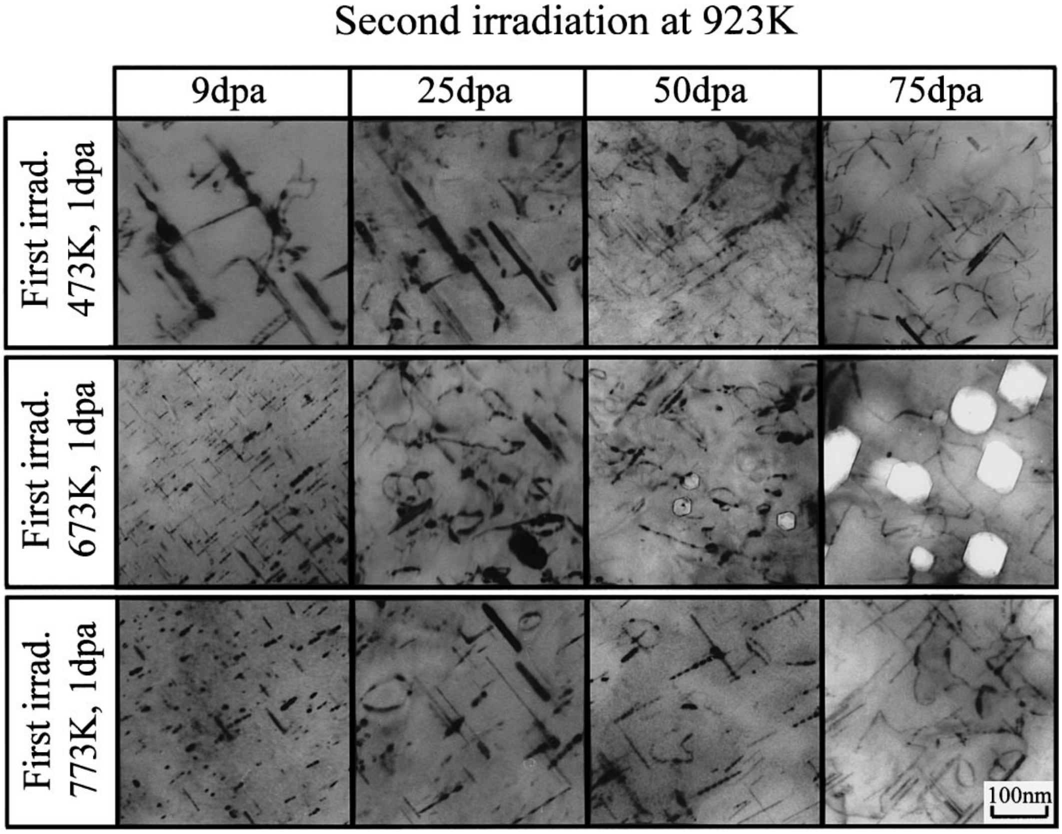

Fig. 4. Microstructural evolution during varying temperature irradiation at high dose. The second irradiation at 923 K was made to 75 dpa.

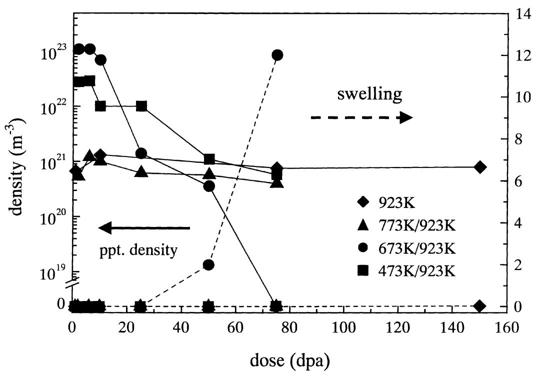

Fig. 5. Dose dependence of phosphide density and void swelling.

high density of interstitial-type dislocation loops observed at 473 K recovered during temperature elevation. Thermal annealing due to the temperature increase is a possible reason. Similar recovery of interstitial-type loops during varying temperature irradiation was reported in Ref. [12]. At 473 K, a high density of small size vacancy clusters would be formed due to the low mobility of vacancies, which will dissociate during the temperature elevation. Interstitial clusters formed at 473 K would shrink by absorbing excess vacancies induced by the temperature increase. Because of the recovery of the loops, no influence of the first irradiation would be observed on microstructural evolution during the second irradiation in the case of 473/923 K-irradiation. In 673/923 and 773/923 K-irradiation, on the other hand, the high density of small phosphides, which were oriented along ⟨1 0 0⟩ direction, were formed accompanied by the interstitial-type loops after the first irradiation at lower temperature. During the temperature rise, loops recovered as in the case of 473/923 K-irradiation, but small phosphides were stable and remained at a high density. The density of these phosphides was much higher than that for the constant temperature irradiation at 923 K. The density of phosphide precipitates during the second irradiation was strongly affected by the density of phosphides that remained after the temperature increase.

4.2. Influence of the first irradiation on void swelling behavior

It is known that the high stability of phosphides up to higher dose levels contributes to suppressing void swelling. The present ion irradiation study on phosphorus modified Fe–Cr–Ni alloys at a constant temperature of 923 K shows strong suppression of void swelling due to phosphide precipitates up to dose levels of 150 dpa, as shown in Fig. 3. But in the case of 673/923 K-irradiation, high void swelling of about 12% due to the reduction of the phosphide density was observed at a dose level of 75 dpa. This fact indicates that, under certain irradiation conditions, varying temperature irradiation may weaken the effect of phosphorous addition on suppressing void swelling.

The enhanced phosphide formation at the beginning of the second irradiation, which was observed for 673/923 and 773/923 K-irradiations, may reduce the phosphorus concentration in the matrix. In the case of 673/923 K-irradiation, growth of the phosphides almost ceased at the beginning of the second irradiation. The density decreased rapidly with increasing dose levels. Along with the dissolution of phosphides, formation and growth of dislocation loops were observed. It is known that a dislocation is a biased defect sink, which contributes to a supersaturation of vacancies in the matrix. Reduction in incubation period for void formation at 673/923 K may be explained as a result of the dissolution of phosphides and formation of dislocation loops. The prominent void swelling after the dissolution of phosphides in irradiation at constant temperature in a low phosphorus containing alloy (−0.024P) was also reported [13].

During irradiation at 773/923 K, on the other hand, no void formation was observed at 75 dpa. Insufficient dissolution of phosphides for enhancing void formation may be the cause. However, since the dislocation loop formation was also observed along with the decrease in phosphide density, reduction of the incubation period for void formation might be expected when the irradiation is continued further.

5. Conclusion

The influence of stepwise temperature change on microstructural evolution and on swelling in a phosphorus modified Fe–Cr–Ni alloy during heavy ion irradiation was investigated. The main results are summarized as follows:

1. The influence of the first irradiation at lower temperature was prominent in the 673/923 and 773/923 K-irradiation while no influence of the first irradiation at lower temperature was observed in the 473/923 K-irradiation.
2. The density of phosphides formed at the beginning of the second irradiation at higher temperature was two orders of magnitude higher for the 673/923 K-irradiation and one order of magnitude higher for the 773/923 K-irradiation than that for the constant temperature irradiation at 923 K.
3. The influence of the first irradiation at the lower temperature remained up to higher dose levels. Phosphide dissociation with increasing dose levels was observed and high void swelling of 12% was observed at 75 dpa in the 673/923 K-irradiation.

These results suggest that, under certain irradiation conditions, the first irradiation at lower temperature strongly affects the phosphide formation during the second irradiation at higher temperature. The change in the phosphide formation may result in a shortening of the incubation time for void formation.

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