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Effect of electron-beam irradiation temperature on irradiation damage of high Mn–Cr steel

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Abstract

The effect of electron-beam irradiation temperature on irradiation damage of a high Mn–Cr austenitic steel for structure material of nuclear and/or fusion reactors from the point of view of the reduced radio-activation was investigated by using the 1250 kV HVEM and an energy dispersed X-ray analyzer (EDX) in a 200 kV FE-TEM with beam diameter of about 0.5 nm. Void formation was not observed even at the elevated irradiation temperature. Dislocation loop growth was observed and the density and size of dislocation loop increased with irradiation dose. Irradiation-induced segregation of Cr and Mn at grain boundaries were observed by electron-beam irradiation condition. The amount of Mn segregation increased with irradiation temperature, however, the case of Cr was suppressed.

1. Introduction

Structure materials for nuclear and/or fusion reactors are quite different from ordinary structures since they are used under neutron radiation condition. SUS 304 and SUS 316 steels with controlled content of alloying elements used for nuclear reactor materials have good properties in un-radiated condition, but properties of these materials are reduced with the change of composition by segregation of solute atoms under long-term neutron radiation conditions [1–3].

According to the calculation of Noda et al. [4], neutron irradiated alloying elements Ni, Mo and Nb results in radioactive isotopes that decay over a long period to Fe, Cr and Mn, so to attenuate the radioactivity to levels that are no longer hazardous requires a long time, more than 100 years. Since SUS 304 and SUS 316 steels have 9–12%Ni, attenuation of radiation requires a very long time. Therefore, addition of reduced radioactive elements to the fusion reactors material was examined. Mn is proposed as a replacement for Ni due to its rapid decay of radioactivity to one tenth of Ni level.

Therefore, much attention has been paid to the high Mn–Cr austenitic steels because of its potential use for structure materials of nuclear and/or fusion reactors from the point of view of the reduced activation [5–9]. Among of them, reduced radio-activated 12%Cr–15%Mn austenitic steels with high content of nitrogen have good properties of high temperature strength and high temperature phase stability [7]. So, it would be important to investigate the irradiation damage of these materials at high temperature.

The aim of this study is to investigate the effect of electron-beam irradiation temperature on irradiation damage of reduced activation 12%Cr–15%Mn austenitic steel by using electron-beam irradiation techniques to simulate the reactor damage processes.

2. Experimental procedure

High Mn–Cr steel used has the following chemical composition: C:0.10, N:0.18, Si:<0.10, Mn:15.10,

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P:<0.003, S:0.005, Cr:11.85, Ni:0.84, W:2.15, V:0.46, Ti:0.10. Specimens for electron-beam irradiation were prepared from a high Mn–Cr austenitic steel which was hot-rolled and solution-treated at 1373 K for 1 h. Disk specimens with 3 mm diameter for TEM observation were punched out and electrochemically-jet polished.

The electron-beam irradiation was carried out with a damage rate of about 0.5×10^{-3} dpa/s at 573, 673 and 873 K up to doses of 5.4 dpa in a 1250 kV HVEM. The concentration of irradiation defects decrease rapidly near specimen surface compare to inner side of specimen, because specimen surfaces are act as sink sources of point defects during electron irradiation [10]. From this, specimen thickness of irradiation area was selected about 400 nm to avoid surface effect. Irradiation-induced segregation analyses were carried out by an energy dispersed X-ray analyzer (EDX) in a 200 kV FE-TEM with a beam diameter of about 0.5 nm.

3. Results and discussion

3.1. Microstructure changes by irradiation

Fig. 1 shows the microstructure change observed for the solution treated high Mn–Cr steel during electron irradiation at 573 K with HVEM. Fig. 1(a) shows the microstructure before irradiation, and Fig. 1(b), (c) and (d) show irradiated microstructure after up to dose of 1.8, 3.6 and 5.4 dpa, respectively. Voids were not formed even after irradiation dose of 5.4 dpa. In the case of electron irradiation at 873 K (not shown in here), voids were also not observed even after irradiation dose of 5.4 dpa despite the higher irradiation temperature. So it could be considered that irradiation damage resistance of high Mn–Cr steel is superior to that of SUS 304 steel [11].

Fig. 2 shows the dislocation structural change observed for the solution treated high Mn–Cr steel during electron irradiation at 673 K with HVEM. Dislocation loops of interstitial atoms type were formed in the early stage of irradiation as shown in Fig. 2(a), and their size and number density increased with irradiation time and then formed coarse and large size of dislocation loops as shown in Fig. 2(d).

Point defects with high mobility agglomerate and secondary defects such as dislocation loops are formed as seen in the figures. The dislocation loops grew with irradiation as shown in Fig. 2. Generally, interstitial dislocation loops are preferentially nucleated rather than that of vacancy and grow by preferentially absorbing the interstitial atoms [12].

The dislocations and voids are effective sink sites for point defects so that both the dislocation loop and void microstructure change with electron irradiation [13]. The long-range migration of the point defects can also result in irradiation-induced segregation that will be discussed later.

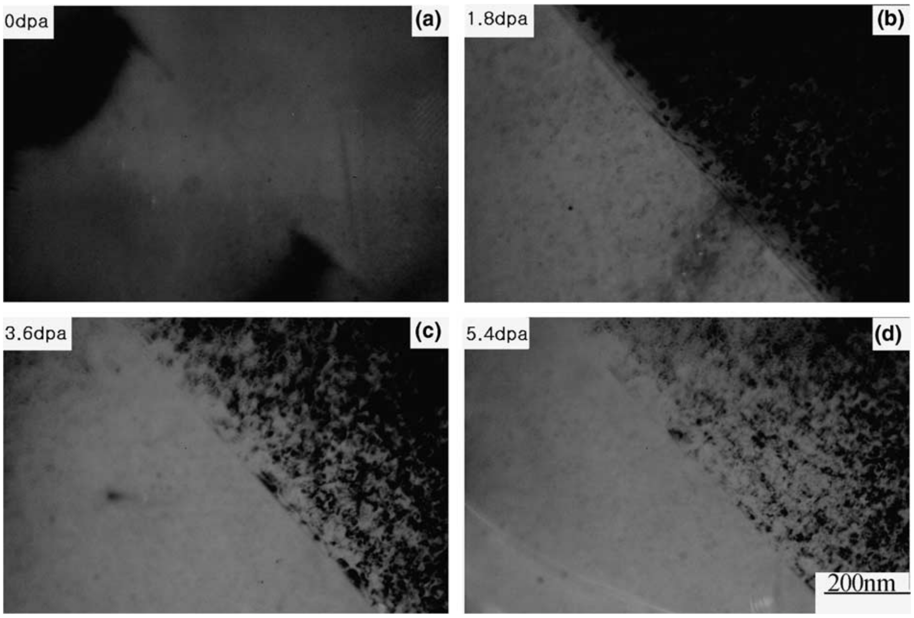

Fig. 1. Microstructure changes after electron-beam irradiation up to (a) 0 dpa, (b) 1.8 dpa, (c) 3.6 dpa and (d) 5.4 dpa at 573 K of solution treated high Mn–Cr steel.

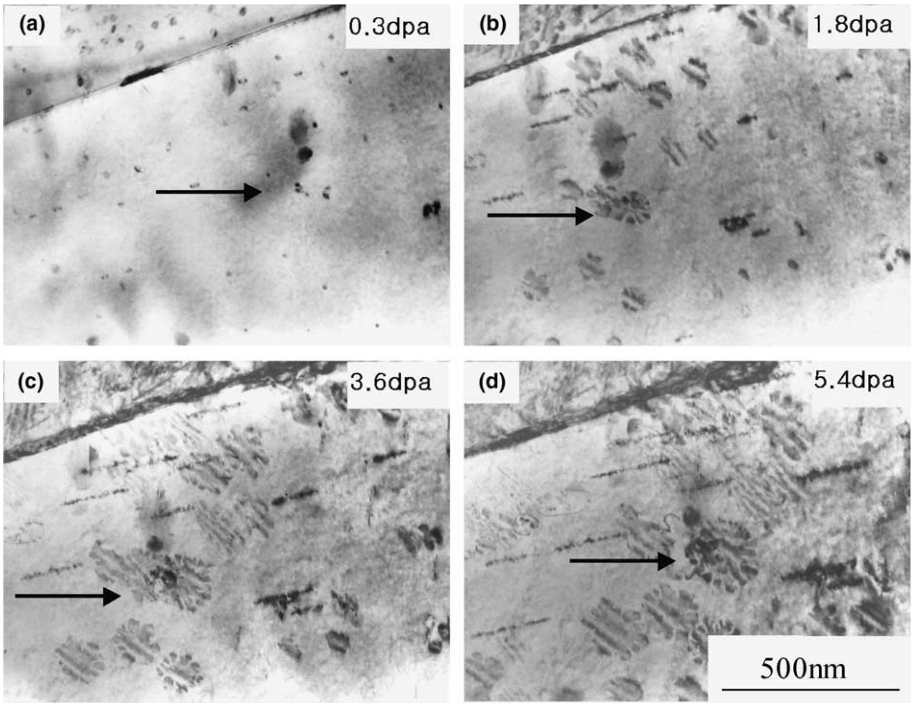

Fig. 2. Dislocation formation changes after electron-beam irradiation up to (a) 0.3 dpa, (b) 1.8 dpa, (c) 3.6 dpa and (d) 5.4 dpa at 673 K of solution treated high Mn–Cr steel.

3.2. Irradiation-induced grain boundary segregation

Fig. 3(a–c) show compositional concentration profiles of Cr and Mn near a grain boundary after electron irradiation up to dose of 5.4 dpa at 573, 673 and 873 K, respectively. Equilibrium segregation occurred in the process of reducing the surface free energy by solute atom diffusion to grain boundary or other surface. Under irradiation conditions, however, large amount of point defects and the secondary defects are introduced continuously as shown in Fig. 2. In this case, the diffusion of solute atoms takes place by interaction of solutes with the fluxes of point defects to defect sinks so that non-equilibrium segregation with enrichment or depletion of solute atoms occurred near sink sites such as grain boundaries and voids.

The phenomena of non-equilibrium segregation, such as concentration changes of Cr and Mn, were also observed at grain boundaries by electron irradiation. The enrichment of Cr and the depletion of Mn at grain boundaries were produced by electron irradiation at 573 K as shown in Fig. 3(a). In Fig. 3(b), however, little change of Cr concentration and the enrichment of Mn at grain boundaries was observed by electron irradiation at 673 K. In the case of an irradiation temperature of 873 K, Cr concentration was not changed and Mn concentration became more enriched than for the other irradiation conditions, as shown in Fig. 3(c).

That is, Cr and Mn concentrations near grain boundaries showed some differences with irradiation temperatures, as shown in Fig. 3. According to the report of Kato et al. [14], fine precipitates were formed at grain boundaries and within grains in the early irradiation stage, then the size and number of precipitates increased gradually with irradiation dose in high Mn steel electron-irradiated to a dose of 10 dpa at 723 K. And they also reported that these precipitates affect the grain boundary segregation. Therefore, it is considered that the changes of precipitation behavior affect the concentration distributions of near grain boundaries with each irradiation temperature of 573, 673 and 873 K.

The atomic radius of Cr is larger than that of Fe matrix so that it is more effective to relax the surrounding strain energy with Cr, namely Cr solute interacts preferentially with vacancies. In this case, Cr diffuses by exchange mechanism with vacancies, and then solute atoms flow in the opposite direction of vacancy flow [15]. That is, Cr depletes at grain boundary in the irradiated region while Cr concentrates in areas away from grain boundaries.

On the other hand, Mn is undersize atom compared to Fe atom, Mn atoms migrate in the same direction as

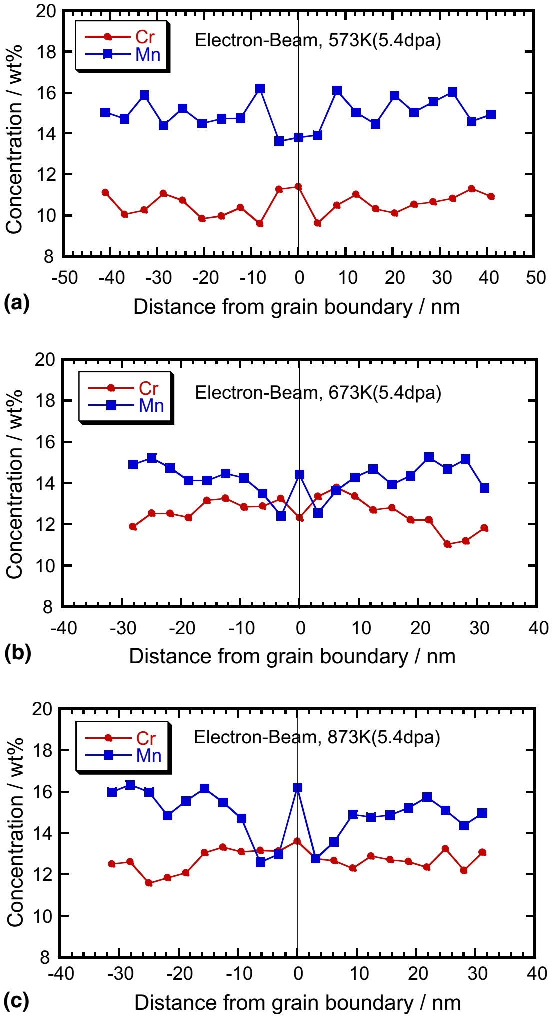

Fig. 3. Comparisons of concentration profile near a grain boundary after electron-beam irradiation up to 5.4 dpa at (a) 573 K, (b) 673 K and (c) 873 K in solution treated high Mn–Cr steel.

interstitial point defects. Mn is depleted in the irradiated region of matrix because Mn with the interstitial atoms migrates away from the irradiated region. That is, solute atom migration was occurred with point defect migration.

Fig. 4 shows the changes of irradiation-induced segregation near a grain boundary with electron-beam irradiation temperature by the trimming the results from Fig. 3. The change of grain boundary segregation (ΔCgb) was calculated from the difference between concentration at the grain boundary and concentration at the nearest position from the grain boundary. Cr concentration near a grain boundary shows about +1.8%Cr concentration difference (namely, enrichment

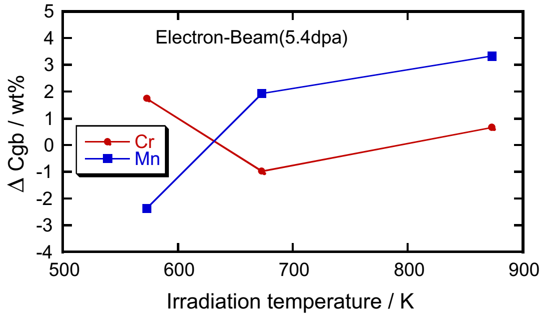

Fig. 4. Changes of irradiation induced segregation near a grain boundary with electron-beam irradiation temperature of solution treated high Mn–Cr steel.

of Cr) at irradiation temperature of 573 K, however, the minimum value of concentration difference shows at irradiation temperature of 673 K and concentration difference shows about +0.7%Cr at irradiation temperature of 873 K. From this, it could be understood that the enrichment of Cr at the grain boundary was reduced and then concentration difference near the grain boundary disappeared gradually with increasing irradiation temperature.

These tendencies are similar with the previous result [14] that radiation-induced solute redistribution in the vicinity of grain boundaries in Fe–10%Cr–(5–15)%Mn–3%Al steels were studied by means of electron irradiation up to 10 dpa at 723 K, then the change of grain boundary segregation showed almost zero value in the case of 15%Mn.

Mn concentration near a grain boundary shows about −2.4%Mn concentration difference (namely, depletion of Mn) at irradiation temperature of 573 K, however, the concentration difference increased rapidly with irradiation temperature and shows about +3.3%Mn (namely, enrichment of Mn) at irradiation temperature of 873 K. That is, the amount of Cr enrichment at grain boundary decreases and that of Mn enrichment at grain boundary increases considerably with irradiation temperature. It might be possible that the region of unstable austenite phase would be formed near grain boundary by decrease of the Mn content near grain boundary with increasing irradiation temperature.

4. Conclusions

Effect of electron-beam irradiation temperature on irradiation damage of high Mn–Cr steel was investigated and results obtained are as follows.

1. Void formation was not observed in the irradiation temperature ranges from 573 to 873 K.

2. The dislocation loop growth was observed and the density and size of dislocation loop increased with irradiation dose.
3. The amount of Mn segregation at grain boundary increased with irradiation temperature, however, the case of Cr was suppressed.

Acknowledgements

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