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HVEM irradiation damage in heat affected zone of welded SUS304 steel

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Abstract

The heat affected zone (HAZ) of a welded SUS304 steel has been irradiated in an 1250 kV high voltage electron microscope at 673 K and up to 5.4 dpa (displacements per atom) to study the effect of electron irradiation on microstructure. The dislocation loop density of initial irradiation state increased with electron irradiation dose. Void size, void number density and void swelling increased and then saturated gradually with irradiation dose. The depletion of Cr and the enrichment of Ni at the grain boundary were also recognized by EDS analysis in the HAZ of welded SUS304 steel.

1. Introduction

It is an important issue to extend the useable lifetime of reactor. To accomplish this extension, one of the required abilities is the repair and/or replacement of irradiated components of the reactor. Welding is one available technique for repair/replacement irradiated materials such as SUS304 and SUS304L stainless steels which are popular structural materials for nuclear reactor [1]. However, the heat affected zone (HAZ) which is formed by the welding process has different physical properties compared to the matrix. Although some work have been performed on the effect of irradiation on microstructural damage of such materials [2–11], the effect of neutron irradiation on the HAZ during operating reactor has not been clarified. The aim of this study is to investigate the effect of electron irradiation on microstructural damage of HAZ by using high voltage electron microscope (HVEM) in order to investigate possible effects of the neutron damage process of reactor.

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2. Experimental procedure

The chemical composition of SUS304 material used in this study was shown in Table 1. The specimens for electron irradiation were prepared from the HAZ of SUS304 stainless steels aged at 673 K for 1000 h after tungsten inert gas arc welding. Single electron irradiation was carried out at 673 K up to 5.4 dpa (displacements per atom) in a 1250 kV HVEM. Irradiation-induced segregation analyses were carried out by an energy dispersive X-ray analyzer in a 200 kV FE-TEM with beam diameter of about 0.5 nm.

3. Results and discussion

3.1. Microstructure changes by irradiation

Fig. 1 shows the dislocation structural change observed for the HAZ part of welded SUS304 steel during electron irradiation at 673 K with HVEM. Dislocation loops were formed in the early stage of irradiation as shown in Fig. 1(a), and their size and number density

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Table 1
Chemical composition of used material (wt%)

	C	P	S	Cr	Ni	Fe
SUS304	0.05	0.025	0.004	18.25	8.27	Balance

increased with irradiation time and then formed tangled dislocation network as shown in Fig. 1(d).

The concentration of point defects such as atomic vacancies and interstitial atoms change with irradiation dose. In this process, point defects with high mobility agglomerate and secondary defects such as dislocation loops are formed. The dislocation loops grew with irradiation as shown in Fig. 1. Generally, interstitial dislocation loops are preferentially nucleated rather than that of vacancy and grow by preferentially absorbing the interstitial atoms [12].

Fig. 2 shows microstructural development of void formed by agglomeration of vacancies introduced by electron irradiation. Small voids were formed in the early stage of irradiation as shown in Fig. 2(a). The number density and size of void increased gradually with irradiation dose as shown in Fig. 2(c) and (d). And, arrow marks in Fig. 2 show two voids coalescence into one larger void.

It was recognized that voids were clearly observable after about 0.6 dpa by the electron irradiation. The voids formed in the early stage of irradiation continued to grow and nucleation of new voids and their growth were observed with irradiation dose. The dislocations and voids are effective sink sites for point defects so that both the dislocation loop and void microstructure change with electron irradiation as observed in Figs. 1 and 2. The long range migration of the point defects could result in irradiation-induced segregation that will be discussed later at Section 3.4.

Fig. 3 shows the electron irradiated region of specimen and the diameter of irradiated region was about 5 μm.

3.2. Size distribution and change of number density of voids

Fig. 4 shows the change of void size and its distributions as a function of dpa in SUS304 stainless steel. Voids formed at early stage of irradiation were small as shown in Fig. 4(a). Void size increased gradually with irradiation dose and the void distribution shows a shift to coarser voids as shown in Fig. 4(c) and (d). This means that small void formed at earlier irradiation grew while new voids were nucleated continuously with irradiation dose.

Fig. 5(a) and (b) show the change of mean void size and void number density with irradiation, respectively.

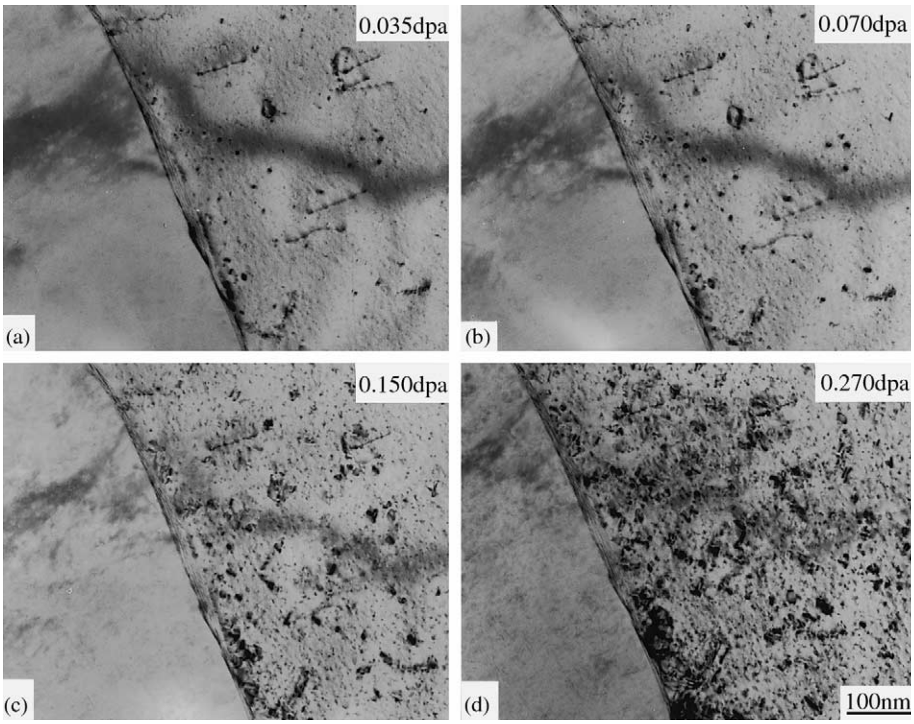

Fig. 1. Dislocation loop growth with electron irradiation up to (a) 0.035 dap, (b) 0.07 dap, (c) 0.15 dpa and (d) 0.27 dpa at 673 K in SUS304 steel.

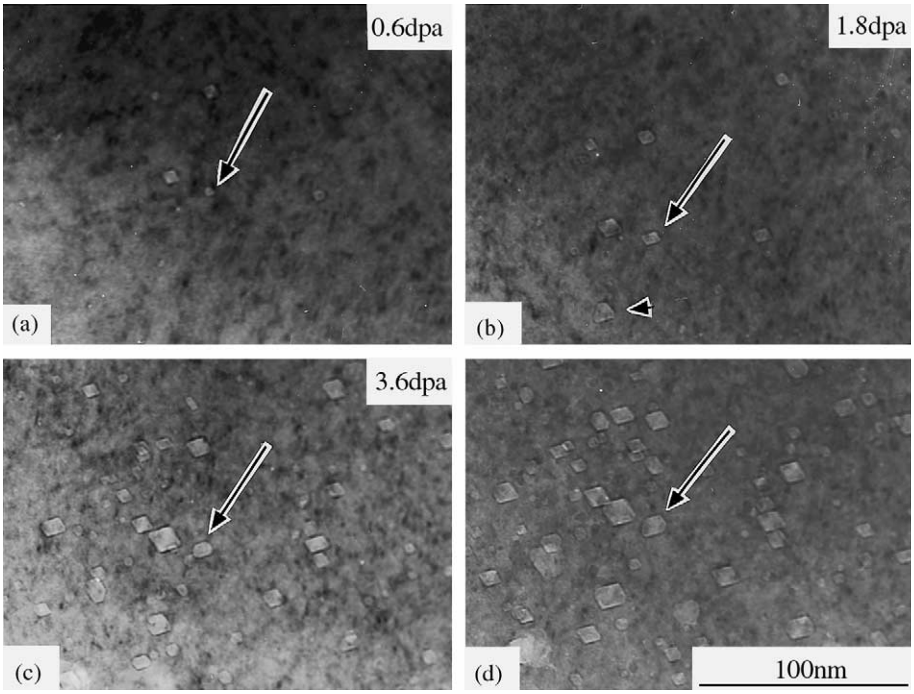

Fig. 2. Void formation changes after electron irradiation up to (a) 0.6 dpa, (b) 1.8 dpa, (c) 3.6 dpa and (d) 5.4 dpa at 673 K in SUS304 steel.

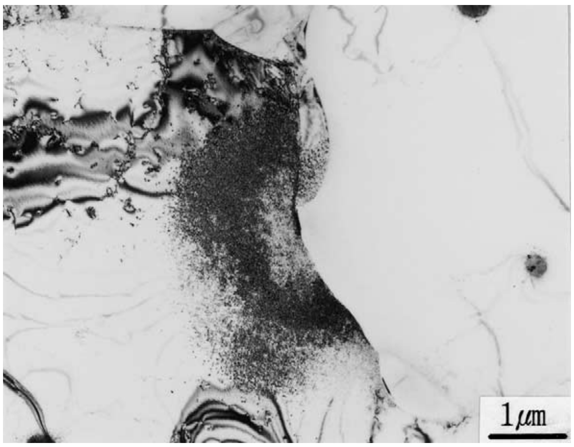

Fig. 3. Irradiated area after electron irradiation to 5.4 dpa at 673 K in SUS304 steel.

Mean size of void was smaller at initial state of irradiation but increased gradually due to growth and coalescence of some voids with irradiation, as shown in Fig. 5(a).

Void number density tends to approach a constant value with continued irradiation. This indicates that, although new voids are formed, void growth was continued by in part the coalescence of each void during irradiation, as shown in Figs. 2, 4(c) and (d). Decrease of void number density is mainly attributed to this coalescence process, even new voids were nucleated with irradiation.

3.3. Void swelling

Fig. 6 shows the change of void swelling with irradiation dose. The quantity of void swelling (V_s) was calculated from Eq. (1) by measuring the void diameter and the void number per unit volume in irradiated microstructure. Void was evaluated approximately as a spherical shape in this calculation.

$$V_s = \frac{\Delta V}{V} = \frac{\pi}{6}\frac{\sum d_|^3}{V} \tag{1}$$

where, $d_|$ means the void diameter and V, the volume of the measured area, that is $V = xyt$ (xy means measured area and t means specimen thickness). The specimen thickness was about 400 nm which was estimated by contamination method.

It has been recognized that the swelling rate increased rapidly in the early irradiation stage, then the swelling rate decreased gradually with irradiation dose as shown

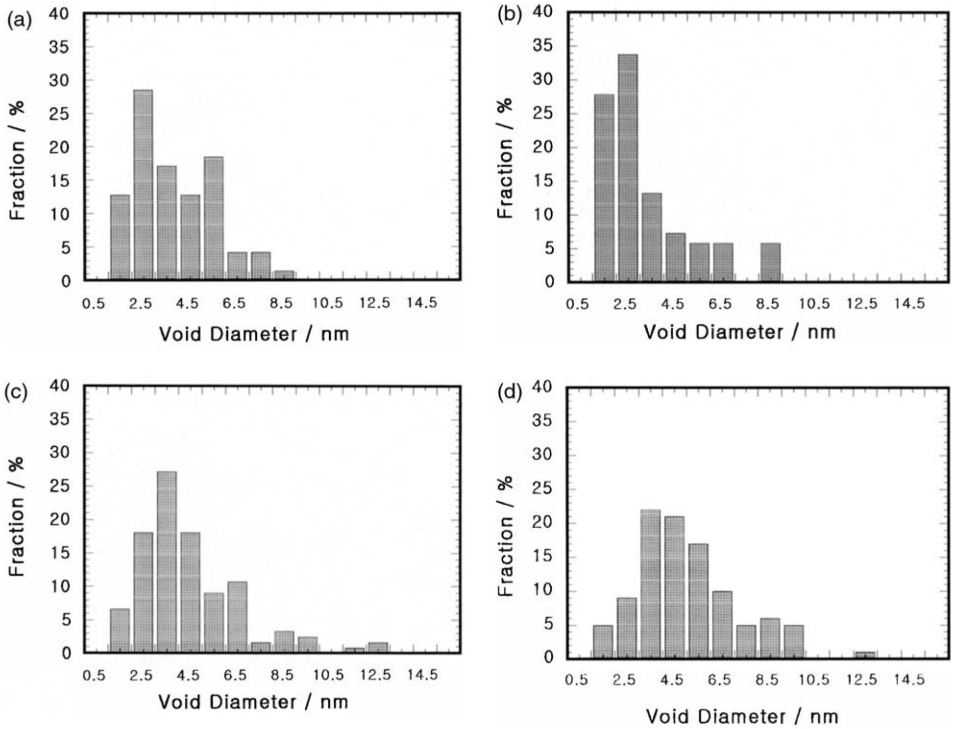

Fig. 4. Void size distributions of electron irradiation up to (a) 0.6 dpa, (b) 1.8 dpa, (c) 3.6 dpa and (d) 5.4 dpa at 673 K in SUS304 steel. Total numbers of voids in each figure are (a) 70, (b) 68, (c) 121 and (d) 135.

in Fig. 6. It is considered that this is reflecting the decrease of void number density due to absorbing or coalescence of voids during void growth with irradiation dose. This result coincides well with above result [13].

3.4. Irradiation-induced grain boundary segregation

Fig. 7 shows compositional concentration profile near a grain boundary after electron irradiation at 673 K. Equilibrium segregation occurred in the process of reducing the surface free energy by solute atom diffusion to grain boundary or other surface. Under irradiation condition, however, large amount of point defects and the secondary defects are introduced continuously as shown in Figs. 1 and 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 boundary and void etc.

The phenomenon of non-equilibrium segregation such as the enrichment of Ni and the depletion of Cr at grain boundary has been also reported by electron irradiation for SUS316 stainless steel [14]. 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 vacancy, then solute atoms flow to opposite direction of vacancy flow. That is, Cr depletes at grain boundary in the irradiated region while Cr concentration area away from grain boundary is increased.

On the other hand, Ni is undersize atom compared to Fe atom, Ni atoms migrate to the same direction with interstitial point defect. Ni is depleted in the irradiated region of matrix because Ni with the interstitial atoms migration toward the outside of irradiated region. That is, solute atom migration was occurred with point defect migration.

Ni enrichment at grain boundary due to Ni migration with interstitial atom coincides with previous results [2–4]. It is suggested that the irradiation-induced segregation phenomena takes place not only in matrix but also the HAZ part where quenched voids would be formed by welding the SUS304 steel.

4. Conclusions

Effect of electron irradiation on damaged microstructure of the HAZ in welded SUS304 steel was investigated and results obtained are as follows:

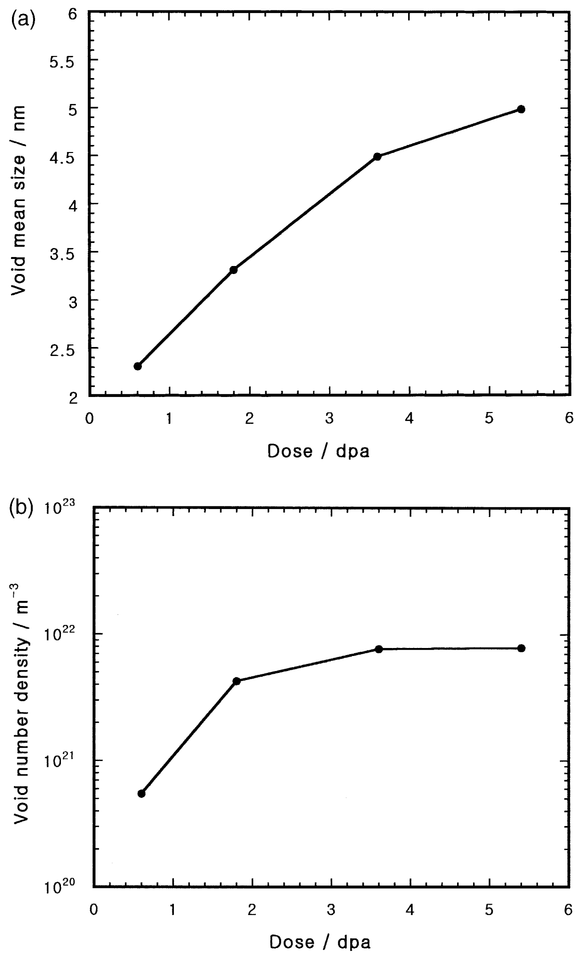

Fig. 5. Changes of (a) void mean diameter and (b) void number density with electron irradiation up to 5.4 dpa at 673 K in SUS304 steel.

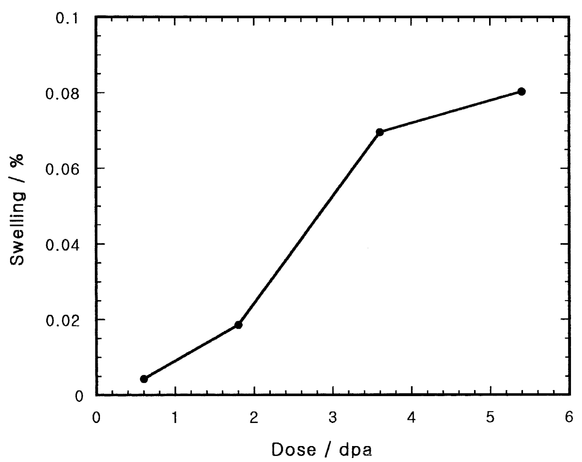

Fig. 6. Changes of void swelling with electron irradiation up to 5.4 dpa at 673 K in SUS304 steel.

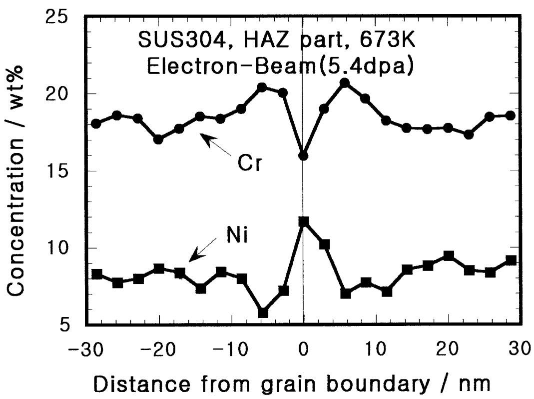

Fig. 7. Concentration profile near a grain boundary after electron irradiation up to 5.4 dpa at 673 K in SUS304 steel.

1. dislocation loop density of the initial irradiation state was increased with electron irradiation dose;
2. void size, void number density and void swelling ratio were increased and then tended to saturate gradually with increased electron dose;
3. irradiation-induced segregation was identified with the depletion of Cr and the enrichment of Ni at a grain boundary in the HAZ of welded SUS304 steel.

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