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Observation and Modeling of Radiation-Induced Solute Segregation Behavior of Grain Boundaries in Stainless Steel 316

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1. Introduction

Irradiation generates various kinds of disorders in crystals, and some point defects such as vacancies and self-interstitial atoms could survive from the recombination process in the early stage of irradiation. The freely migrating defects could move and re-incorporated into the crystal structure at various defect sinks such as grain boundaries or dislocations. These defect fluxes could induce the flux of the atoms in the crystals, and the enrichment and the depletion of the species could occur because of the preferential interaction between the species and point defect fluxes. These phenomena are known as radiation-induced segregation (RIS) [1].

Stainless steel 316 (SS316) is a main material for reactor internals. The neutron irradiation in the reactor core causes RIS of SS316; the depletion of Cr and the enrichment of Ni, Si, and P at grain boundaries. The depletion of Cr may cause an irradiation assisted stress corrosion cracking (IASCC), and can be a potential problem for the integrity of the nuclear power plant.

In this work, The RIS behavior of random boundaries and Σ3 twin boundary were observed. Further, we implemented the RIS modeling, and compared the calculation results with the experimental results.

2. Methods and Results

2.1 Experiments

The test material was a solution annealed plate of SS 316. The composition is listed in table I. Most important solutes are Cr and Ni. For the irradiation test, we made a small disk specimen like a TEM sample, 3 mm in diameter and 100 μm in thickness. The irradiation was carried out using a heavy ion machine. The ion source was Fe^{4+} with the energy of 8 MeV. The temperature of the specimen was 400°C, and the irradiation time was up to 4 hr. The total fluence was about 10 dpa, and the dpa of the sample was estimated by TRIM code. After irradiation, the orientation of the grain boundaries were identified by EBSD, and a small region was cut by FIB technique. The maximum irradiation depth of the samples was about 2 μm. We observed the region of 0.5 μm depth (~2 dpa) and the region of 1.0 μm depth (4 dpa). The depletion and enrichment of solute atoms were measured using TEM/EDS.

Table I: Composition of SS 316 (wt%)

Cr	Ni	Mo	Mn
17.1	11.1	2.1	1.3
Si	P	C	S
0.6	0.04	0.061	0.001

2.2 Modeling methodology

Fe-Cr-Ni ternary system was used for the modeling of RIS in SS316. We extend the ternary model implemented by Perks *et al*. [2] to our system which is containing twins. The model was based on the inverse Kirkendall effect, in other words, vacancy mechanism. Differential rate equations for point defects and solutes are described below.

$$\frac{\partial C_k}{\partial t} = -\nabla J_k$$

$$J_k = -D_k \alpha \nabla C_k + d_{kv} C_k \nabla C_v - d_{ki} C_k \nabla C_i$$

$$\frac{\partial C_v}{\partial t} = -\nabla J_v + \eta G_{dpa} - K_{vi}(D_v + D_i) C_v C_i - S_v D_v (C_v - C_v^{eq})$$

$$\frac{\partial C_i}{\partial t} = -\nabla J_i + \eta G_{dpa} - K_{vi}(D_v + D_i) C_v C_i - S_i D_i (C_i - C_i^{eq})$$

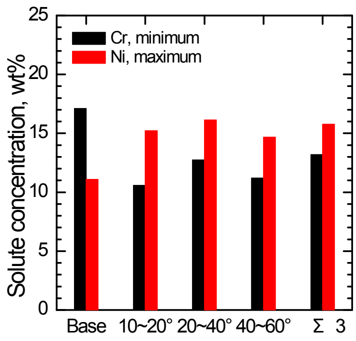
The detailed equations are described elsewhere [2]. The most important term is d_{kv} which represents diffusivity correlation factor of atom-defect relationship. As the factor increase, atoms move easily with vacancies.

To consider grain boundary effect, grain boundary sink strength (S^{GB}) was introduced. The boundary conditions of the system were fixed as deep boundary conditions which represented an infinite large grain. Additionally, there were one or two grain boundaries with S^{GB} at the center region of the grain. The calculated composition was convoluted with X-ray generation profile having the standard deviation of 1 nm [3].

2.3 RIS at twin boundaries

Fig. 1 shows the effect of misorientation between two grains on RIS behavior. The black bars represent the depleted Cr concentrations and the red bars represent the enriched Ni concentrations. In 2 dpa result, all specimens show similar RIS, even the special boundary, Σ3 twin boundary shows rather high RIS behavior. The tendency of the 4 dpa result is similar to the 2 dpa result except that Ni enrichment is clearly increased with increasing the irradiation dose. It was reported that Σ3 twin boundary showed a suppressed RIS behavior [4]. To elucidate the RIS behavior we observed high resolution TEM of Σ3 twin boundaries.

(a)



(b)

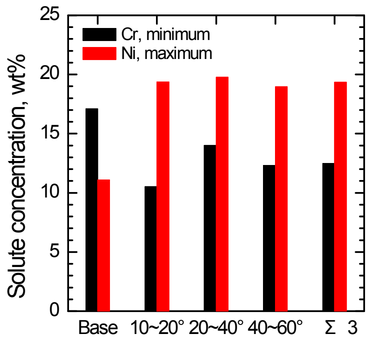


Fig. 1. Solute concentration changes of Cr and Ni with various grain boundary conditions.

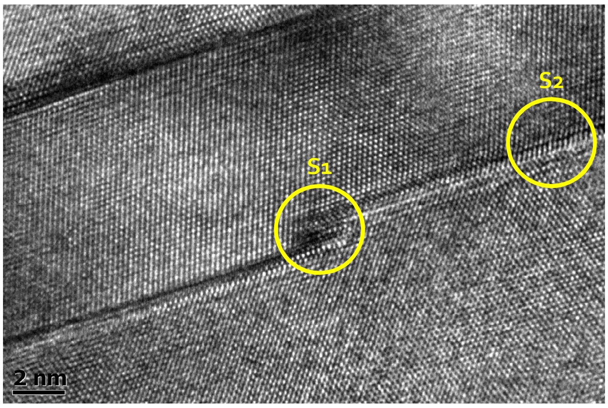


Fig. 2. HRTEM image of Σ3 twin boundary with the dose of 4 dpa.

Fig. 2 shows the HRTEM image of the Σ3 twin boundaries with the dose of 4 dpa. The twin relationship was clearly shown by the atom configuration in the image. As shown in figure, there is a ledge in the lower boundary, and TEM/EDS revealed that the S1 point shows extremely high RIS. The Cr composition was 9.4 wt% and Ni was 24.5 wt%. The ledge shows higher RIS behavior than random boundaries. On the other hands, point S2, coherent plane region, the depletion and enrichment were decreased than random boundaries. Hence, it is believed that the coherency between boundaries is very important factor for RIS.

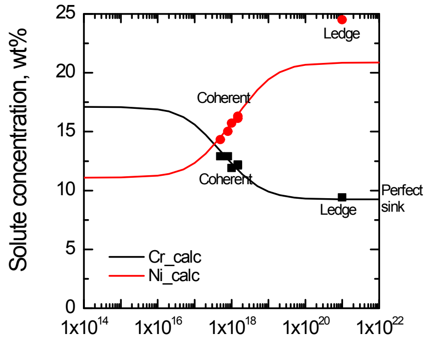


Fig. 3. Modeling prediction (lines) of RIS *vs*. grain boundary sink strength with the dose of 4 dpa at 400°C.

Fig. 3 represents the relationship between the S^{GB} and depletion/enrichment of solute. The measured points were superimposed on the calculation result line. The ledge of Σ3 acts as a perfect sink; however, the coherent region is expected to have a moderate S^{GB}. These values of S^{GB} can be used as a parameter for modeling of RIS in the multiple twin boundaries.

3. Conclusions

The ion-irradiation experiments were carried out to observe the RIS behavior in SS316, the coherency of Σ3 affected the RIS behavior drastically. A theoretical evaluation of RIS in SS316 was implemented, and the estimation was compared to the measured values. These results were sensitive to model parameters; however the calculated amount of depletion/enrichment shows a fair agreement with the measured one with the uncertainties of the parameters. It is essential to develop an elaborate model in the future.

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