Evaluation of irradiation-induced point defect migration energy during neutron irradiation in modified 316 stainless steel

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Abstract. The widths of void denuded zones (VDZs) which were formed near random grain boundaries by neutron irradiation were analyzed in order to perform quantitative evaluations for the irradiation-induced point defect behavior in the modified 316 stainless steel (PNC316) having been developed by JAEA. Namely, the temperature dependence of VDZ width was investigated and vacancy migration energy of the PNC316 steel was estimated from the VDZ width analysis for the neutron-irradiated specimens. The obtained value of vacancy migration energy was estimated as 1.46 eV, which was consistent with that from the exiting method using electron in-situ examination. This indicates that VDZ analysis could be effective method to evaluate especially vacancy migration energy during irradiation, and this would be realized from not in-situ observation but post-irradiation examination in the case of neutron irradiation.

Key Words: Austenitic stainless steel, Void denuded zone, Grain boundary, Neutron irradiation

1. Introduction

For the development of nuclear core materials, especially fuel cladding tube, in sodium-cooling type fast breeder reactor, void swelling suppression is one of the most important issues to keep the dimensional stability in reactor. A large number of theoretical and experimental investigations on void swelling behavior have been carried out [1-5], and void swelling directly depends on the diffusion of point defects induced by neutron irradiation as well as the strength of point defect sinks such as dislocations and precipitates. Evaluations of the point defects diffusion in metals during neutron irradiation have been qualitatively done through various researches [6-7], however the quantitative estimation is hardly performed due to the difficulty of in-situ experiments during neutron irradiation. Instead of that, the indirect estimations from the temperature dependence measurements of dislocation loop densities and growth rates using electron in-situ observation are often carried out [8], but the irradiation correlation between electron and neutron irradiations, such as the differences of irradiation dose rate and damage morphology, should be discussed with accuracy. The quantitative evaluation of point defect migration will help us to theoretically predict the void swelling behavior such as the incubation period and swelling rate with accuracy. Furthermore, this evaluation will give the useful information for the development of nuclear core materials with high void swelling resistance through the comparison of each point defect mobility with other steels having different additives or initial Ni contents.

Therefore, in this study, the evaluation of point defect diffusion, especially vacancy migration, during neutron irradiation was tried. In detail, from neutron-irradiated microstructures, vacancy migration energy was estimated using the knowledge that the widths of void denuded zones (VDZs) formed near random grain boundaries (GBs) depend on temperatures [9,10]. This estimation was performed for the modified 316 stainless steel (PNC316 steel) used to the fast breeder reactor Monju or JOYO, and vacancy migration behavior during neutron irradiation in the PNC316 steel was discussed.

2. Experimental procedure

The test material was the PNC316 steel which was the modified 316 stainless steel with 20 % cold working and additives such as P, Ti, B, and Si to improve the void swelling resistance. The chemical composition is given in TABLE 1. The fuel assembly composed of mainly the PNC316 steel, which was driver fuel, was irradiated in the experimental fast reactor JOYO. The PNC316 specimens were cut from this assembly, and foils of 3 mm in diameter were prepared. Neutron irradiation conditions of their foils are collated in TABLE 2, which were irradiated at temperatures from 722 K to 821 K and doses of 74.5–87.5 dpa (displacement per atom). After electro-polishing for the foils, the analysis of VDZ widths was performed by transmission electron microscope (TEM) observations focusing the void distributions near the random GBs, and the widths of VDZ were measured. Here, the VDZ width was determined as the average distance between the GB position and voids closest to the GB. Based on the theoretical equation for VDZ width [9], vacancy migration energy during neutron irradiation was estimated from the Arrhenius plots of the obtained VDZ widths and the reciprocal temperatures. This value was compared with the energy estimated from the existing method; the temperature dependence evaluation of dislocation loop growth rates using electron in-situ observation. Electron irradiation examination was carried out for the solution-annealed PNC316 steel using a 1 MV high-voltage electron microscope (HVEM) in Hokkaido University. All electron-irradiated areas had an enough foil thickness greater than 300 nm, to minimize the effect of the surface sink.

TABLE 1: CHEMICAL COMPSITION OF THE PNC316 STEEL SPECIMENS

(wt %)

C	Si	Mn	P	Fe	Cr	Ni	B	N	Mo	Ti	V	Nb+Ta
0.041	0.76	1.71	0.025	bal.	16.50	13.45	0.0030	0.005	2.34	0.070	<0.01	0.046

*Final heat treatment: 1313 K × 2 min.

*Cold working: 20 %

TABLE 2: IRRADIATION CONDITIONS OF THE PNC316 STEEL SPECIMENS

Specimen No.	Temperature (K)	Dose (dpa)
Specimen 1	722	79.5
Specimen 2	782	85.0
Specimen 3	787	87.5
Specimen 4	788	80.0
Specimen 5	821	74.5

3. Result and Discussion

3.1. Void distribution after neutron irradiation

Figures 1 show typical void distributions observed near random GBs with high misorientation angles in the PNC316 steel, which were neutron-irradiated to 74.5–87.5 dpa at 722 K to 821 K. In the microstructures in the neutron-irradiated PNC316 steel specimens, there were not only voids but also much precipitates and complex dislocation structures. The application of cold working is well known to increase point defect sinks such as dislocations and prolong the incubation period of void swelling. On the other hand, the addition of minor elements plays a role of enhancing point defect sinks that correspond the interface between the matrix and precipitates formed by the additives. In addition, it causes a suppression of point defect migration by the interaction between solute atoms and additives, so that the incubation period is prolonged and void growth rate is reduced due to suppressing the mean free path and the aggregation of interstitials or vacancies.

Garner [11] reported that the numbers of dislocations introduced by cold working were reduced during neutron irradiation to higher doses, and voids would be nucleated only after decreasing the dislocation density. Therefore, it is considered that the void distribution in the PNC316 steel could only be observed after weakening the dislocation effect (which extends the incubation period of void swelling). Namely, FIG. 1 indicates the microstructure after relatively high dose irradiation and their void distributions were formed at the steady state for void nucleation and growth.

Specimen 1	Specimen 2	Specimen 3	Specimen 4	Specimen 5
722 K	782 K	787 K	788 K	821 K

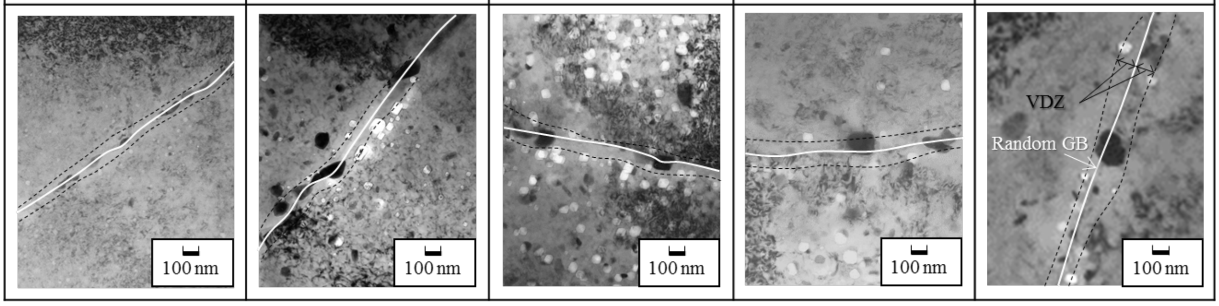

FIG. 1. TEM images; void distributions near random grain boundaries in the neutron-irradiated PNC316 steel specimens.

3.2.VDZ width near random grain boundary

As indicated in FIG. 1, VDZ formations in all neutron irradiation conditions of this study were confirmed. The white solid and black dotted lines in FIG. 1 indicate the positions of random GB and VDZ, respectively. The detail mechanism of VDZ formation has been reported in elsewhere [9, 12-15]. It follows that the concentration of excess vacancies introduced during irradiation is lowered to the critical concentration level to form voids near a random GB which acts as a neutral sink because excess irradiation-induced vacancies near the GB could flow toward the GB and be absorbed at the GB. Namely, the excess vacancy concentration is decreased near a random GB, and voids are denuded there. Furthermore, it is known that VDZ width depend on the temperature and displacement rate but not irradiation dose [9].

In this study, different VDZ widths should be caused by only temperature change because the displacement rates in all neutron irradiation conditions, these were about 10^{-6} dpa/s, are almost same. In TABLE 3, the average VDZ widths measured in this study are listed. For all VDZ widths, it was confirmed that the standard deviation was not large. And, it was found that VDZ widths increased with increase of irradiation temperature. This indicates that vacancy mobility and mean free path were enhanced as increasing temperature and vacancy flow distance toward a random GB was extended. Moreover, VDZ widths in the PNC316 steel in this study were 50 - 90 nm at 749 K to 821 K. By contrast, VDZ width in the model alloy Fe-15Cr-15Ni [15] was about 141 nm at 749 K, and the width of PNC316 steel was clearly lower than that in the Fe-15Cr-15Ni. This indicates that vacancy flow toward a random GB was reduced in the PNC316 steel with additives, which would be mainly attributed to large substitutional elements such as Nb and Ti because these oversized elements would interact preferentially with vacancies to reduce vacancy mobility [15, 16].

TABLE 3: VDZ WIDTHS OF THE NEUTRON-IRRADIATED PNC316 STEEL

Specimen No.	VDZ width	Standard deviation
Specimen 1	47.7 nm	± 10.9 nm
Specimen 2	76.3 nm	± 12.0 nm
Specimen 3	77.2 nm	± 13.0 nm
Specimen 4	83.2 nm	± 18.2 nm
Specimen 5	95.2 nm	± 13.9 nm

3.3.Vacancy migration energy analysis

Konobeev et al. [9] have reported on the theoretically-analyzed relationship between VDZ width and vacancy diffusivity under the condition in which the concentrations of excess point defects were beyond the thermal equilibrium concentrations. And, it was shown that VDZ width is a function of vacancy diffusivity. Therefore, in this study, an analysis of the relationship between VDZ width and vacancy diffusivity (or migration energy) for the PNC316 steel was conducted based on the theoretical model proposed by Konobeev et al. In this model, the relationship between VDZ width (W_d) and vacancy diffusivity (D_v) (or migration energy (E_v)) under irradiation can be expressed as follows:

$$W_d = [D_v/4K\mu]^{1/4} \qquad (1).$$

Here, $D_v = D_0 \exp(-E_v/k_BT)$; k_B is the Boltzmann constant, T is the absolute temperature, E_v is the vacancy migration energy, D_0 is a constant coefficient, K is the dose rate, and μ is the recombination parameter.

Based on the equation (1), the activation energy for the formation of VDZ in the PNC316 steel was estimated as 0.36 eV from the Arrhenius plots of the obtained VDZ widths and the reciprocal temperatures, which is indicated in FIG. 2.

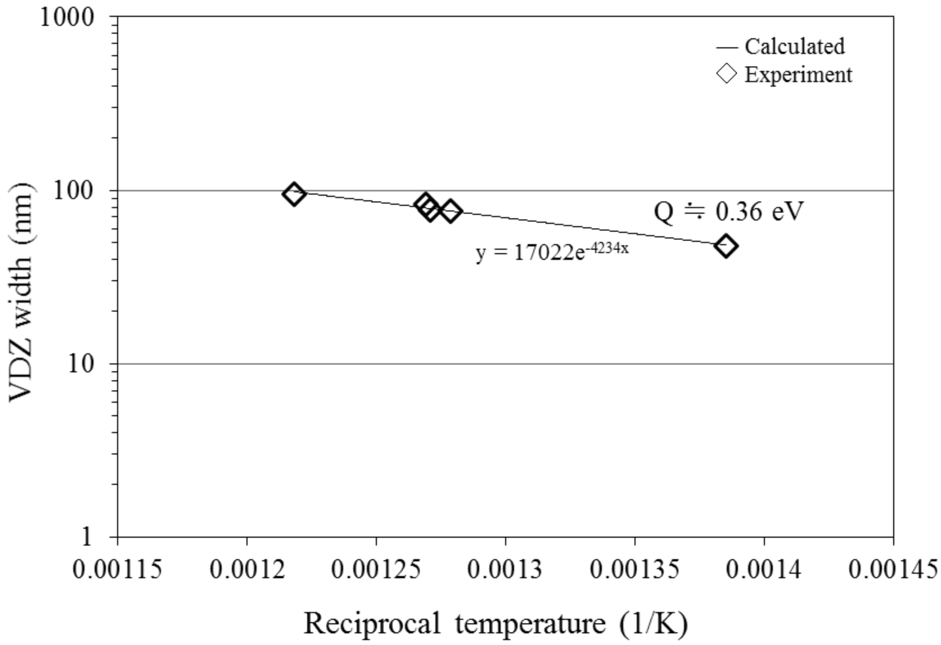

FIG. 2. Relationship between the VDZ width and reciprocal temperature.

On the other hand, in order to investigate the actual vacancy migration energy, the growth rates of dislocation loops were investigated from electron in-situ irradiation experiments. Vacancy migration energy in the PNC316 steel was estimated as 1.37 eV (see FIG. 3) from the growth rate evaluation of some dislocation loops; this is an existing method to estimate vacancy migration energy [8].

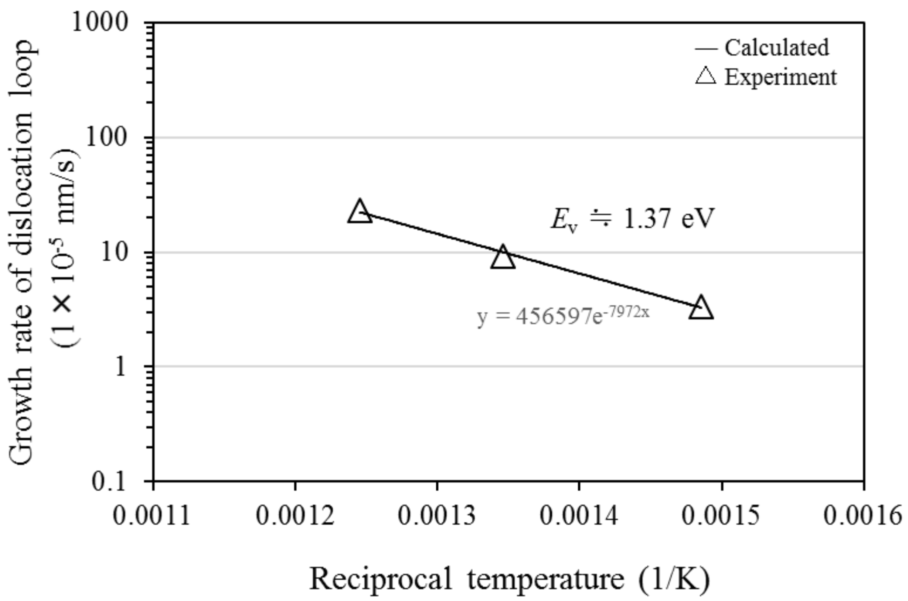

FIG. 3. Relationship between the growth rate of dislocation loop and reciprocal temperature.

According to the equation (1), VDZ width depends on the vacancy diffusivity or vacancy migration energy to the one forth power. Thus, comparing the activation energy (0.36 eV) for

VDZ formation with vacancy migration energy (1.37 eV), it was confirmed that the former was about one forth times of the latter. Therefore, it is considered that the temperature dependence of VDZ widths in the PNC316 steel should be undoubtedly followed by the equation (1).

In addition, the equation (1) is applied when the process of point defect annihilation is not sink dominant but recombination dominant. As the PNC316 steel is the alloy with high sink strength as mentioned above, it is suggested that the process of the microstructure development corresponds to sink dominant case at relatively low dose before void swelling occurs and the nucleation of voids is suppressed. But, after void swelling starts (voids start to nucleate and grow constantly), the process of the microstructure development would be changed to recombination dominant case due to the effect of weakened sink strengths.

Next, as it was found that the temperature dependence of VDZ width in the PNC316 steel could be enough governed by the equation (1), the vacancy migration energy during neutron irradiation in PNC316 steel was also estimated using the equation (1). The estimated value is 1.46 eV, which corresponds to the four times of the activation energy. From this result, it was found that the vacancy migration energy obtained by VDZ analysis could be the almost same as that (1.37 eV) estimated by the existing method (the growth rate evaluation of dislocation loops). And, this value is larger than 1.21 eV, which was obtained from dislocation loop growth measurements under electron irradiation for 316L stainless steel containing oversized Ti [16-17]. Moreover, as vacancy migration energy in SUS316L steel (commercial steel) is about 1.00 eV [16], the value of PNC316 steel implies that the vacancy mobility could be low as a result of interaction of vacancies with minor alloying elements.

Consequently, it was found that the evaluation of especially vacancy migration during neutron irradiation could be possible from VDZ width analysis for the PNC316 steel (actual steel). It could be carried out from neutron-irradiated examination (post-irradiation examination), not in-situ observation. Clarifying the vacancy migration energy for the PNC316 steel in this study would be useful to enhance the theoretical prediction accuracy for void swelling behavior. Furthermore, it is expected that this evaluation (VDZ width analysis) could be the effective method for alloy development to optimize effects of additional elements and main solute atom contents as well as evaluations of point defect accumulation behavior in the neutron-irradiated microstructures.

4. Summary

In this study, vacancy migration energy relating to void swelling behavior was estimated in the PNC316 stainless steel from VDZ width analysis by TEM observations using the neutron-irradiated specimens. The obtained knowledge is summarized as follows.

- It was confirmed that VDZ width had the temperature dependence, which indicates that vacancy mobility and mean free path were increased with increasing irradiation temperature.
- VDZ widths obtained in this study were 50 - 90 nm at 749 K to 821 K. These values were lower than that of the Fe-Cr-Ni model alloy. This is caused by the interaction between additives and solute atoms and the point defect mobility in the PNC316 steel would be suppressed.
- The activation energy for VDZ formation in the PNC316 steel was estimated as 0.36 eV, and it was about one forth times of vacancy migration energy (1.37 eV) which was estimated from the existing method (the growth rate evaluation of dislocation

loops). This was content with the theoretical equation for VDZ formation mechanism.

- Vacancy migration energy estimated from VDZ width analysis was 1.46 eV, which is the almost same as that (1.37 eV) estimated by the growth rate evaluation of dislocation loops. This could be possible to directly evaluate vacancy migration energy relating to void swelling from post irradiation examination.

Appendix 1:

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