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Simulating the influence of radiation temperature variations on microstructural evolution

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Abstract

The influence of temperature variations on microstructural evolution in austenitic stainless steels is discussed in order to help interpret the response of materials in the HFIR-RB-13J temperature variation experiment. A kinetic microstructural evolution model developed for irradiated austenitic stainless steels was modified to provide a fully dynamic calculation of point defect and point defect cluster concentrations. Using the modified model, microstructural evolution was predicted for simulated HFIR-RB-13J temperature variation experiments and variations in material parameters were evaluated. The results indicate that repeated temperature excursion to 573 from 773 K always results in increased dislocation loop density and reduced cumulative defect flux within the calculated material parameter range, while excursions to 473 from 623 K may increase or decrease them depending on material parameters. The primary influence of temperature variation could be explained by accumulation and release of matrix defects at the temperatures of interest. The applicability of the current model must be further studied by careful investigation of the results from the HFIR-13J and related experiments.

1. Introduction

It has been pointed out that temperature variations during neutron irradiation experiments may cause significant modification of resultant microstructures in irradiated materials [1]. Since most of the conventional 'nominally isothermal' neutron irradiation experiments were subjected to significant temperature and neutron flux variations during reactor start-up and shut-down sequences, the influence of such unsteady conditions on irradiation effects need to be assessed in order to properly understand the physical mechanisms of the irradiation effects as well as the temperature dependent macroscopic effects. In addition, typical operation cycle of the fusion power devices will presumably be much shorter than that of the fission reactors. Therefore, to predict material behavior in fusion reactors, it is necessary to develop an understanding of the effects of variations in operating conditions.

In order to investigate the effects of temperature variations, an irradiation experiment in the High Flux Isotope Reactor (HFIR) RB-13J at Oak Ridge National Laboratory (ORNL) was proposed and carried out as one of the major tasks in the Japan–US collaborative JUPITER program for fusion materials research [2,3]. Prior to the HFIR experiment, a variety of temperature-controlled irradiations were performed in the Japan Materials Testing Reactor (JMTR) using precision temperature control irradiation capsules. One of the JMTR experiments included a subset of the HFIR-RB-13J material matrix [4]. The affect of temperature variations was also investigated using charged particle irradiation experiments [5]. The objective of this work is to support the interpretation of the potentially complex experimental results to be obtained from the HFIR and JMTR irradiations, through a theoretical evaluation of the influence of temperature variations on the microstructural evolution. Austenitic stainless steel was selected as the material to be studied because its irradiation response is most broadly understood and modeled so far.

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2. Model description

The computational model used in this work is based on a comprehensive rate theory model of microstructural evolution in irradiated austenitic stainless steel, originally developed by Stoller and Odette [6,7], which has successfully been simulating the microstructural and swelling behavior or Type 316 stainless steels irradiated in fast reactors. The model was also extensively used for materials response evaluation in the fusion blanket conditions [8]. It was then modified, mainly to include the time evolution of vacancy clusters, both point defect reaction-produced and cascade-produced, and helium-vacancy complexes [9–11]. The modified model calculated the point defect and cluster concentration fully dynamically, instead of using the pseudo-steady-state solution in the original one. The configuration of vacancy clusters was assumed to be stacking fault tetrahedron. The configuration of cascade-produced vacancy clusters influences the temperature dependence of freely migrating defect flux in an intermediate temperature range [11].

This modification was successful in simulating the free defect suppression at lower temperatures but also showed that the evolution of cascade-production of interstitial clusters must be included in order to reproduce the realistic temperature dependence of dislocation loop evolution. Since the temperature dependence of loop evolution is essential in this work, in-cascade interstitial cluster production was included, following Stoller [12] and Gan et al. [13].

Molecular dynamics computer simulations suggest that a significant fraction (~60%) of the surviving interstitials cluster by the end of cascade-cooling phase [14]. On the other hand, previous rate theory work applied an effective clustered interstitial fraction ($\mathrm{CDF_i}$) of about 10^{-5} [13]. This discrepancy can be explained by the hypothesis that the most of the cascade-produced interstitial clusters are immediately removed from the matrix by the interaction with local strain gradient [15]. Models of interstitial cluster motion and dislocation dynamics need to be advanced and proved for practical engineering alloys in order to include a physics-based treatment of this mechanism [16–18]. In this work, the effective $\mathrm{CDF_i}$ was assumed to be a function of dislocation density in a conventional rate theory model in the following way:

$$\mathrm{CDF_i} = \mathrm{CDF_i^0} \times (1 - \rho_\mathrm{d}\pi r_\mathrm{c}^2), \tag{1}$$

where ρ_d is the dislocation density and r_c is the minimum distance between the dislocation lines and stable interstitial clusters. This treatment implies that cascade interstitial clusters produced within a r_c of the nearest dislocation line will automatically be removed into the dislocation as a stream of isolated interstitials. The value of r_c was initially set to 10 nm, which is close to half of the mean dislocation spacing experimentally observed in mixed microstructures of dislocation networks and small interstitial loops in stainless steels after low to medium temperature irradiation [19]. $\mathrm{CDF_i^0}$ corresponds to the effective $\mathrm{CDF_i}$ in low dislocation density conditions and is used as a fitting parameter.

Other parameters used in this work are basically the same as those in [11]. Fig. 1 shows the influence of r_c on the maximum interstitial loop number density ($N_\mathrm{l}^\mathrm{max}$). The experimental data compiled in [19] are plotted together. The $\mathrm{CDF_i^0}$ was kept at 5×10^{-6}, since changing it results in a significant $N_\mathrm{l}^\mathrm{max}$ variance in the high temperature regime. The r_c determines the saturated N_l at low to intermediate temperatures and is fixed to 10 nm hereafter.

The 'capture radius' model employed in this study works only when the value of $\mathrm{CDF_i^0}$ is unrealistically small compared to the MD values. The potential problems of this treatment may be associated with treating the motion of interstitial clusters as identical with the random diffusion of isolated single interstitials. This could lead to the inappropriate build-up rate of dislocation loops during the early stages of irradiation and thereby imposes the need to carefully evaluate the impact of model assumptions when comparing to the experimental data. This treatment is a simple expedient to permit the existing model to be used in the evaluation of the temperature change experiments, more theoretical development is plainly required to capture the details implied by some of the MD results [14,16–18,20].

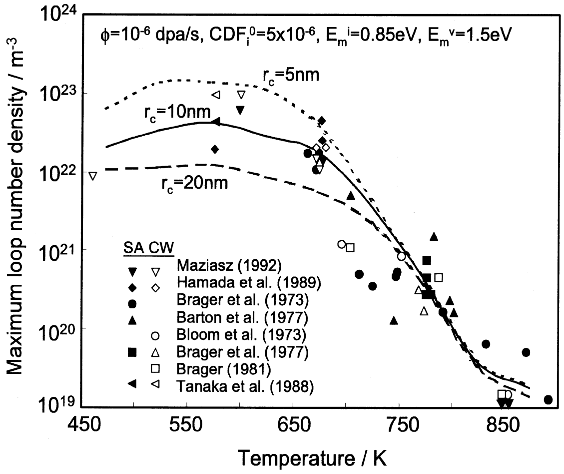

Fig. 1. The influence of dislocation capture radius for SIA-clusters on maximum Frank loop number density calculated for the case of $\mathrm{CDF_i^0} = 6 \times 10^{-6}$. The experimental data points are from the compilation in [19].

3. Results and discussion

3.1. Simulating HFIR-13J experiments

The neutron irradiation in the HFIR-RB-13J temperature variation experiment consists of four different conditions; constant temperature at 773 K (referred '773 K' hereafter), 573/773 K variations ('573/773 K'), constant at 623 K ('623 K') and 473/623 K variations ('473/623 K'). The total dose in each condition is about 5 dpa for stainless steel. The variation capsules were subjected to 10 cycles of temperature change; each cycle consists of 0.05 dpa irradiation at the lower temperature (T_1) followed by 0.45 dpa irradiation at the higher temperature (T_2).

The calculated time evolution of the interstitial loop number density and network dislocation density for the HFIR-13J cases are shown in Figs. 2(a) and (b). The initial dislocation density was set to $3 \times 10^{13}\ \mathrm{m}^{-2}$, for the typical solution-annealed condition in stainless steels. In the 573/773 K case, a high density of loops is rapidly produced during the T_1 periods. The excess vacancies accumulated at T_1 then remove the substantial loop fractions right after the temperature changes to T_2, and the N_l gradually approaches the steady-state value during the rest of the T_2 periods. The network dislocation density will be slightly increased in the temperature variation case, due to the increased source term from the loop reactions. The effect of temperature variations on the dislocation structures may not be clear after the irradiation of 10 cycles, though the increased loop production may be detected after the first cycle. Observable cavity production in stainless steels will not occur by 5 dpa. Also, the mechanical property will not significantly be influenced by the temperature variations.

Fig. 3(a) shows the time evolution of interstitial and vacancy fluxes during the first three cycles for the 773 and 573/773 K cases. During the T_1 periods, the interstitial flux is always higher than the vacancy flux. However, the absolute differential flux in the T_1 periods is much smaller than that in the T_2 periods. The cumulative net vacancy flux (Φ_v) and the cumulative differential defect flux (Φ_d), defined as

$$\Phi_v \equiv \int (D_v C_v - D_i C_i)\, dt, \tag{2}$$

$$\Phi_d \equiv \int |D_v C_v - D_i C_i|\, dt, \tag{3}$$

are plotted in Fig. 4(a) against the elapsed time. Φ_v and Φ_d are introduced as parameters that correlate positively with swelling and irradiation creep, respectively. The temperature variation will reduce Φ_v by the increased

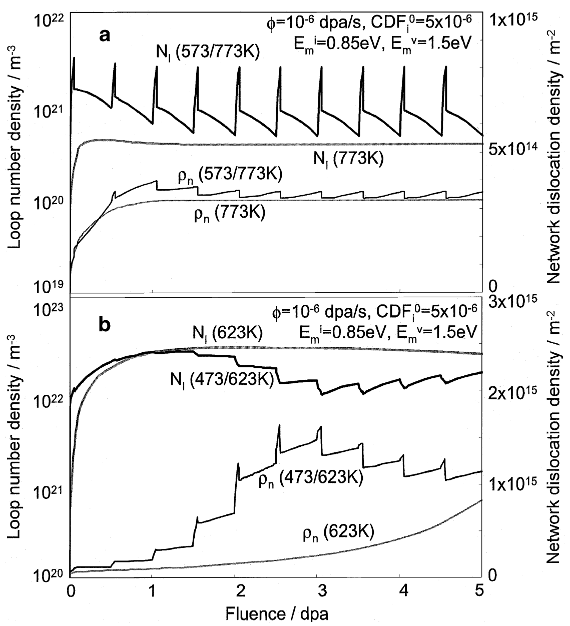

Fig. 2. Calculated influences of temperature variations in HFIR-13J experiment on the time evolution of Frank loop number density and network dislocation density in (a) 773 and 573/773 K cases and (b) 623 and 473/623 K cases.

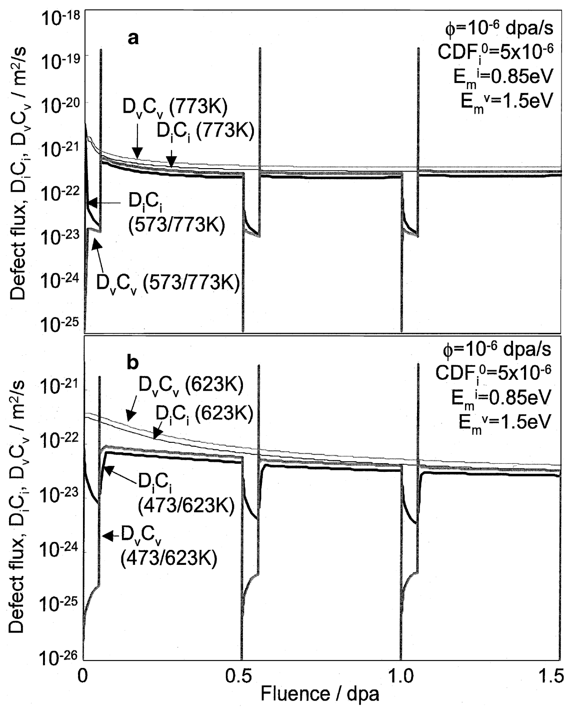

Fig. 3. Calculated time evolution of interstitial and vacancy fluxes during the first three cycles of HFIR-13J experiment in (a) 773 and 573/773 K cases and (b) 623 and 473/623 K cases.

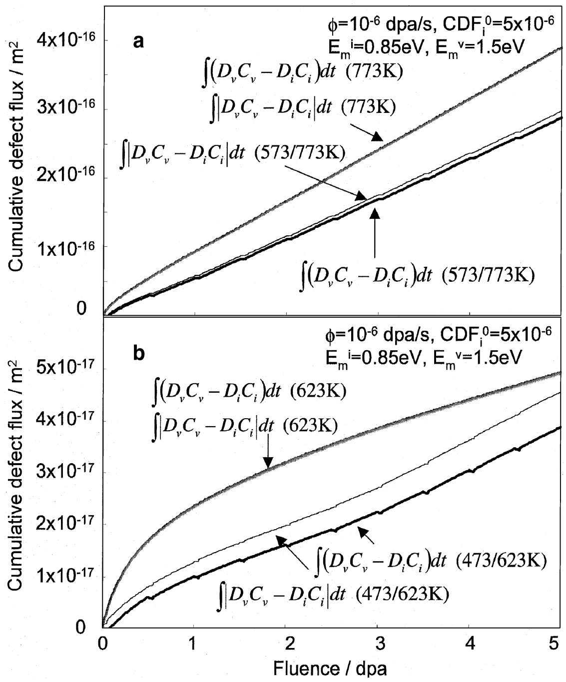

Fig. 4. Comparison of cumulative net vacancy flux and differential defect flux between constant and varied temperature conditions in (a) 773 and 573/773 K cases and (b) 623 and 473/623 K cases.

dislocation sink strength and Φ_d slightly more due to the inverse defect flux during the T_1 periods.

The calculated time evolution of dislocation microstructure in the 623 and 473/623 K cases presented in Fig. 2(b) suggests a somewhat enhanced influence of temperature variation at the lower temperatures. In the 623 K case, dislocation loops are produced initially at high rates and N_l then tends to saturate by about 1 dpa. The steady but slow growth of the loops increases network dislocation density slowly. Since N_l has become saturated after about 1 dpa, net interstitial flux during the T_1 periods, shown in Fig. 3(b), does not effectively contribute to the loop production anymore but enhances the network dislocation production through the loop growth and reactions. The temperature variation may thus alter the dislocation structure in the initial irradiation stages; however, the microstructure will come close to that of the constant temperature cases in the end of irradiation. Although the current dislocation evolution model in the mean field approach may not be quite applicable to such a low temperature condition as the 473/623 K case, we can at least conclude that the inverse defect flux during the T_1 periods acts in different ways between the unsaturated and saturated loop conditions. As shown in Fig. 4(b), the consequences of temperature variation on Φ_v and Φ_d will be similar to those in the 573/773 K case.

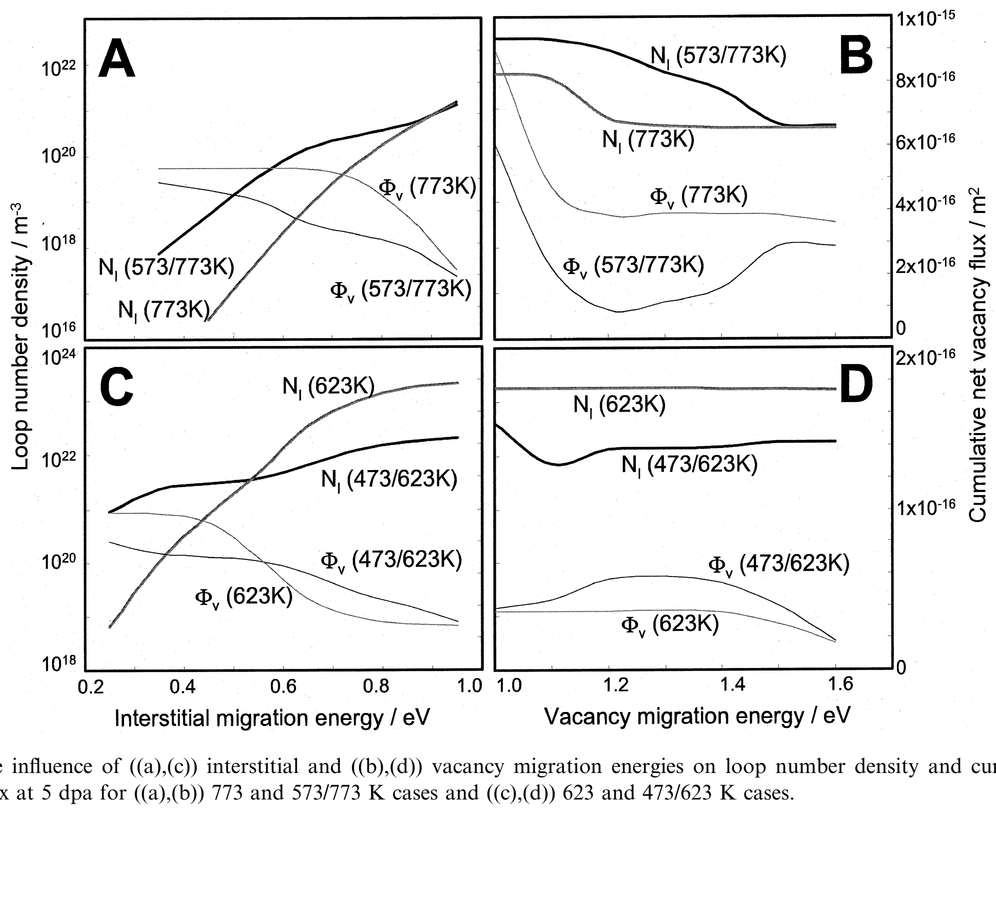

Fig. 5. The influence of ((a),(c)) interstitial and ((b),(d)) vacancy migration energies on loop number density and cumulative net vacancy flux at 5 dpa for ((a),(b)) 773 and 573/773 K cases and ((c),(d)) 623 and 473/623 K cases.

3.2. Influence of material parameters

As observed in the previous section, microstructural modification by the temperature variation at the fluence level of 5 dpa is mostly controlled by the evolution of dislocation loops. Therefore, the influence of interstitial and vacancy migration energies (E_m^i and E_m^v, respectively), material parameters that strongly affect the loop evolution, were studied to evaluate their effect on the degree of microstructural modification by temperature variation in the HFIR-13J cases. The range of $E_m^{i,v}$ values selected encompass those expected for the model austenitic alloys included in the HFIR-13J experiment.

Figs. 5(a)–(d) compare values of N_l and Φ_v obtained at 5 dpa as functions of E_m^i and E_m^v for the temperature variation and constant temperature cases. In the comparison of the 773 and the 573/773 K cases, differences in N_l reach about two orders with small E_m^i as shown in Fig. 5(a). This is because N_l at 773 K is drastically reduced as E_m^i decreases, while in the 573/773 K case, the substantial fraction of the 573 K irradiation-produced loops survive the subsequent 773 K irradiation. The reduced E_m^v always increases N_l through decreasing the recombination fraction of interstitials. The network dislocation density at the end of irradiation was not strongly affected by the temperature variation in any combination of migration energies. The temperature variation reduced Φ_v in all the $E_m^{i,v}$ cases, generally by enhancing the dislocation sink strength. The swelling-creep compliance should not significantly be affected by the temperature variation, since $(\Phi_d - \Phi_v)/\Phi_v$ did not exceed 6% in any case.

In the 473/623 K case, temperature variation increased N_l for smaller values of E_m^i, for the same reason as the 573/773 K case, and decreased N_l for larger E_m^i, as shown in Fig. 5(c). In the latter cases, the accelerated network dislocation evolution by enhanced loop growth during the 473 K periods suppresses the loop density. Therefore, with temperature variation, N_l is less sensitive to E_m^i as compared to the constant temperature cases. The Φ_v are again mostly determined by the dislocation sink strength and follow the inverse trend of N_l. The microstructures developed in both the 623 and the 473/623 K cases are relatively insensitive to E_m^v, and $(\Phi_d - \Phi_v)/\Phi_v$ was smaller than 10% in any case. The change in the swelling-creep compliance becomes significant in cases where: (1) the T_1 period is substantially longer, (2) temperature variation cycles are very short or (3) a similar point defect transient is induced by pulsed irradiation rather than by temperature variation.

4. Conclusion

Microstructural evolution in austenitic stainless steels during the HFIR-RB-13J temperature variation experiment was calculated using a fully dynamic rate theory model. A dislocation density-dependent CDF_i model was introduced so that the calculated dislocation loop evolution might fit the experimental data in a broad temperature range.

Repeated temperature excursion to 573 from 773 K always resulted in increased dislocation loop density and reduced cumulative defect flux within the calculated material parameter range. Repeated temperature excursions to 473 from 623 K may increase or decrease the loop density and cumulative defect flux depending on material parameters. The influence of temperature variation could mostly be explained by accumulation and release of matrix defects at the temperatures of interest.

The applicability of the model employed must be further evaluated by careful investigation of the results from the HFIR-13J and related experiments. Additional model development is required to account for some of the details of cascade damage production observed in molecular dynamics cascade simulations.

Acknowledgements

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