Interaction of Vacancies with Solute Atoms in Electron-Irradiated Fe-Cr Alloys

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1. Introduction

The swelling resistance of ferritic/martensitic (F/M) steels at an elevated temperature has increased the interest in their use as structural materials for nuclear fusion power reactors. Particularly, F/M steels with high Cr contents are candidates for first-wall and breeding-blanket materials in fusion reactor systems. It is known that the addition of Cr to pure Fe reduces radiation-induced swelling significantly [1]. The present work was aimed at qualitatively investigating the role of point defects during the microstructural evolution of Fe-Cr model alloys. For Fe-Cu alloys, it is probable that these vacancy-Cu complexes provide the nucleation sites for full-fledged Cu precipitates. It would be of interest to establish whether low swelling of Fe-Cr alloys under irradiation is related to the interactions between vacancies and Cr atoms. For this purpose, we irradiated Fe-Cr alloys with different Cr contents by high-energy electrons. Then, a positron annihilation (PA) measurement was made in order to verify the production of point defects and to investigate the resultant atomic configuration in the vicinity of vacancies. In parallel, we performed a Monte Carlo computer simulation to estimate the atomic arrangements of the evolved microstructure in the electron-irradiated Fe-Cr alloys. This work elucidates the effects of vacancies on the swelling behavior of Fe-Cr alloys by combining experimentation results and a computer simulation.

2. Experimental

Binary Fe-Cr alloys with Cr contents of 5, 9 and 15 wt% were prepared. These samples were irradiated with 2 MeV-electrons at the JAEA accelerator at a controlled temperature of < 50^{o}C. The total electron dose amounts to 1×10^{18} e/cm^2, which corresponds to ~7.5×10^{-5} dpa. It is likely that most of the point defects produced by electron irradiation may exist in the form of isolated Frenkel pair. Based on this assumption, the calculated concentration of vacancy was ~6.5×10^{18} /cm^3 for the Fe-9Cr alloy. The PA lifetime and coincidence Doppler Broadening (CDB) measurements for five samples (unirradiated Fe, irradiated Fe, Fe-5Cr, Fe-9Cr, Fe-15Cr) were performed at room temperature. The PA lifetime data was analyzed by using the PALSFIT program. The CDB spectra were obtained with two high-purity Ge-detectors in order to plot the ratio curve, which enabled us to examine the elemental information from the vicinity of the open-volume defects such as vacancies and voids. The overall energy resolution of the CBD system was 0.9 keV.

3. Computational Methods

The point defects produced during the early stage of irradiation play a role in the development of microstructure through the interactions with surrounding atoms. For F/M steels containing Cr atoms, it might be important to understand the behavior of solute atoms and point defects under irradiation. Metropolis Monte Carlo (MMC) method provides a convenient tool to predict an atomic configuration of microstructure by means of energy minimization techniques. We developed an MMC computer code to predict the minimum-energy configuration of the atomic structure for Fe-Cr alloys as a result of electron irradiation. The key factor that determines the atomic configuration of a structure is the interatomic potential. In this study, we employed a two-band model of the Fe-Cr system for the potential developed by Olsson [2]. The focal point in the MMC simulation is the role of the vacancies produced by electron irradiation in the formation of atomic clusters. We investigated the atomic configuration of the system through the MMC simulation by changing the initial Cr contents.

4. Results and Discussion

The results for the positron annihilation lifetime data of each sample are listed in Table 1. The average positron lifetimes of the irradiated metals are about 120 ps, whereas the lifetime of the unirradiated Fe is 106 ps, corresponding to the lifetime of well-annealed pure Fe. This is clear evidence that open-volume defects exist in the irradiated samples. For the detailed analysis, the spectra for the irradiated samples were decomposed into two components of τ_1 and τ_2. The τ_1 component corresponds to the annihilation of free positrons non-localized in the lattice. A relatively long lifetime component τ_2 reveals that the positrons are annihilated at

Table 1 Positron annihilation lifetimes and intensities for five samples (unirradiated pure Fe / irradiated pure Fe, Fe-5Cr, Fe-9Cr, Fe-15Cr)

Sample	Average PA lifetime, τ_{avg} (ps)	PA lifetime (ps)		Intensity (%)	
		τ_1	τ_2	I_1	I_2
pure Fe	106.0				
irr. pure Fe	120.0	99.5	169.8	78.6	21.4
irr. Fe-5Cr	120.6	97.6	178.2	76.1	23.9
irr. Fe-9Cr	121.0	104.6	217.5	92.1	7.9
irr. Fe-15Cr	120.4	95.6	166.4	68.8	31.2

vacancies which are formed due to irradiation. The τ_2 components, listed in Table 1, clearly indicate that a certain amount of vacancies are distributed in the irradiated samples. Fig. 1 shows the CDB ratio spectra for the irradiated samples normalized to the momentum distribution of well-annealed pure Fe. The enhancement in the low-momentum region ($< 7 \times 10^{-3}$ m_oc, where c is the speed of light and m_o is the electron rest mass) represents that positron are trapped in open-volume defects such as vacancies. A broad peak for all the samples could be clearly seen in the low-momentum region. It is also observed that there is no significant difference in the high-momentum region of the CDB spectra between the irradiated pure Fe and Fe-Cr alloys. It is expected that the atomic configurations of pure Fe near the vacancies are similar to those of the Fe-Cr alloys. That is, vacancies do not form a cluster with the neighboring Cr atoms but exist in an isolated form.

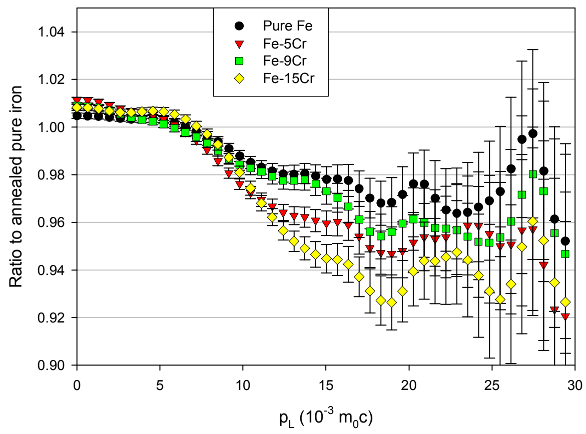

Fig. 1. Ratio curves of CBD spectra for electron-irradiated samples: pure Fe, Fe-5Cr, Fe-9Cr, Fe-15Cr, normalized to the momentum distribution of well-annealed pure Fe.

The spatial Cr distributions, which were initially random, in Fe-5Cr, Fe-9Cr, and Fe-15Cr were examined after the computer simulation. Fig. 2 shows one example of Cr-atom distribution of the Fe-9Cr alloy before and after the MMC calculations. Although it is difficult to see the changes in Cr-atom distribution, the tendency to from the Cr agglomeration can be seen to some extent. Regardless of the Cr contents, we did not observe Cr-vacancy complexes but a small-sized Cr clusters in the block from the visual inspection. It appears that the Cr atoms tend to gather together without any vacancy. Although the structure of a Cr cluster is not a complete precipitate, this cluster takes a transitional form which lies between the molecules and the bulk matter. The tendency to resist the formation of a Cr-vacancy complex is of significance in that binary Fe-Cr alloys reveal low swelling under irradiation.

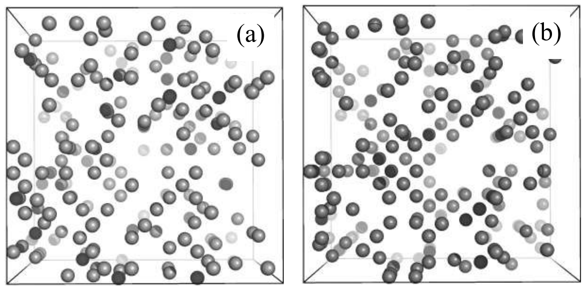

Fig. 2. Spatial distribution of Cr atoms for the Fe-9Cr alloy (a) before the simulation, and (b) after the simulation.

5. Conclusions

The present study demonstrates that the coupling of a PA measurement and a computer simulation helps us understand the microstructural evolution of the binary Fe-Cr alloys. The measured data for the CDB spectra of the electron-irradiated samples revealed that vacancies existed in an isolated form without clustering with Cr atoms. Such a finding was supported by the MMC simulations. We did not observe an evolution of a Cr-vacancy complex but only clustering of Cr atoms. This result strongly suggests that the swelling resistance of F/M steels under irradiation is related to the interactions between Cr atoms and vacancies.

REFERENCES

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