Journal of Nuclear Materials 386–388 (2009) 165–168

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Effects of radiation-induced defects on microstructural evolution of Fe–Cr model alloys

J. Kwon [a,*], T. Toyama [b], Y.-M. Kim [a], W. Kim [a], J.-H. Hong [a]

[a] Korea Atomic Energy Research Institute, Yuseong, Daejeon, Republic of Korea
[b] Institute for Materials Research, Tohoku Univ., Oarai, Ibaraki, Japan

A B S T R A C T

We investigate the role of radiation-induced defects in the microstructural evolution of Fe–Cr model alloys with two approaches; a positron annihilation measurement and computer simulations. We prepared Fe–Cr model alloys with different Cr contents which were irradiated with 2 MeV-electrons at the JAEA accelerator at Takasaki in Japan. The production of vacancy-type defects was verified by positron annihilation lifetime spectroscopy. The average positron lifetimes of the irradiated samples were increased by about 14% when compared to the unirradiated one. The ratio curves, derived from a Doppler broadening measurement, did not reveal a prominent change between the irradiated pure Fe and Fe–Cr alloys, which implies that there was no formation of a vacancy-Cr complex. We performed computer simulations by using the Metropolis Monte Carlo method to predict a possible microstructure composed of Cr and vacancies. As expected, the simulation also revealed that no vacancy-Cr complexes were formed. Both results support a small amount of radiation-induced swelling in the Fe–Cr alloys.

1. Introduction

The swelling resistance of ferritic/martensitic (F/M) steels at elevated temperatures 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 [1,2]. For binary Fe–Cr alloys irradiated to 30 dpa at temperatures in the range of 380–460 °C, their swelling is suppressed considerably by the addition of a small amount (~1%) of Cr, but this reduction in swelling tends to become saturated at 5% Cr. Further addition of Cr (>5%) increases swelling again [1]. Such a complex behavior has not been completely accounted for as yet. Low swelling in Fe–Cr alloys was, however, explained by the presence of dislocation loops characterized by a different bias to interstitials and a different mobility [3].

This work was aimed at qualitatively investigating the role of point defects during the microstructural evolution of Fe–Cr model alloys. For Fe–Cu alloys irradiated by neutrons, irradiation-induced vacancies are stabilized by forming vacancy-Cu complexes, which was revealed by coincidence Doppler broadening (CDB) measurement of positron annihilation radiation [4,5]. 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. Positron annihilation techniques, including PA lifetime spectroscopy and CDB spectroscopy, were applied for the measurement. 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 computer simulations.

2. Experimental

Binary Fe–Cr alloys with Cr contents of 5, 9 and 15 wt% were prepared by vacuum induction melting using electrolytic Fe and Cr stock. The alloys were heat treated at a rate of 5 °C/s to a recrystallization temperature for 3–5 h and then water quenched. The samples were irradiated with 2 MeV-electrons at the JAEA accelerator at a controlled temperature of <50 °C. The total electron dose amounts to 1×10^{18} e/cm^2, which corresponds to ~7.5×10^{-5} dpa. It is unlikely that 2 MeV-electrons lead to displacement cascades in metals because of the production of low energy recoil atoms. Most of the point defects produced by electron irradiation may exist in the form of isolated Frenkel pair. Based on this assumption, the

* Corresponding author.
E-mail address: jhkwon@kaeri.re.kr (J. Kwon).

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doi:10.1016/j.jnucmat.2008.12.079

calculated concentration of vacancy was $\sim 6.5 \times 10^{18}/cm^3$ for the Fe–9Cr alloy [6].

The PA measurements for five samples (unirradiated Fe, irradiated Fe, Fe–5Cr, Fe–9Cr, Fe–15Cr) were performed at room temperature. We employed a ^{22}Na β^+-source of about 1 MBq and collected about 2×10^6 counts for 12 h. The time resolution of the system was 180 ps in full width at half maximum. The PA lifetime data was analyzed by using the PALSFIT program [7] by subtracting

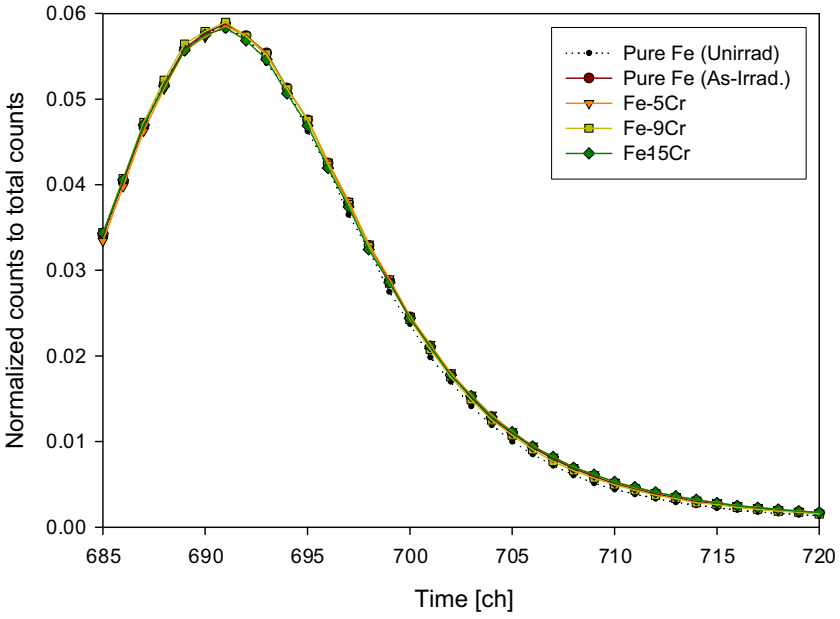

Fig. 1. Experimental positron annihilation lifetime spectra for five samples (unirradiated pure Fe/irradiated pure Fe, Fe–5Cr, Fe–9Cr, Fe–15Cr).

Table 1
Positron annihilation lifetimes and their 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

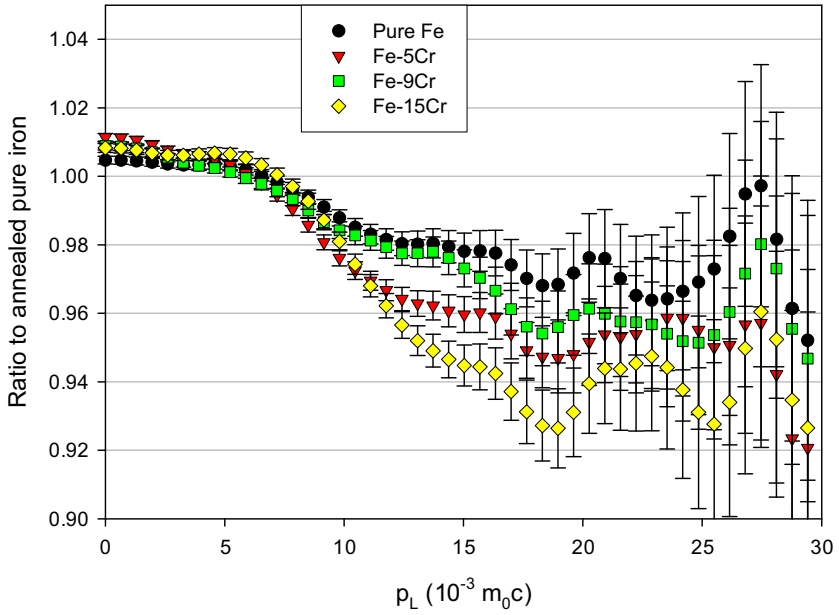

Fig. 2. Ratio curves of CBD spectra for electron-irradiated samples: pure Fe, Fe–5Cr, Fe–9Cr, Fe–15Cr, normalized to the momentum distribution of well-annealed pure Fe.

the source components and the background. The CDB spectra were measured 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.

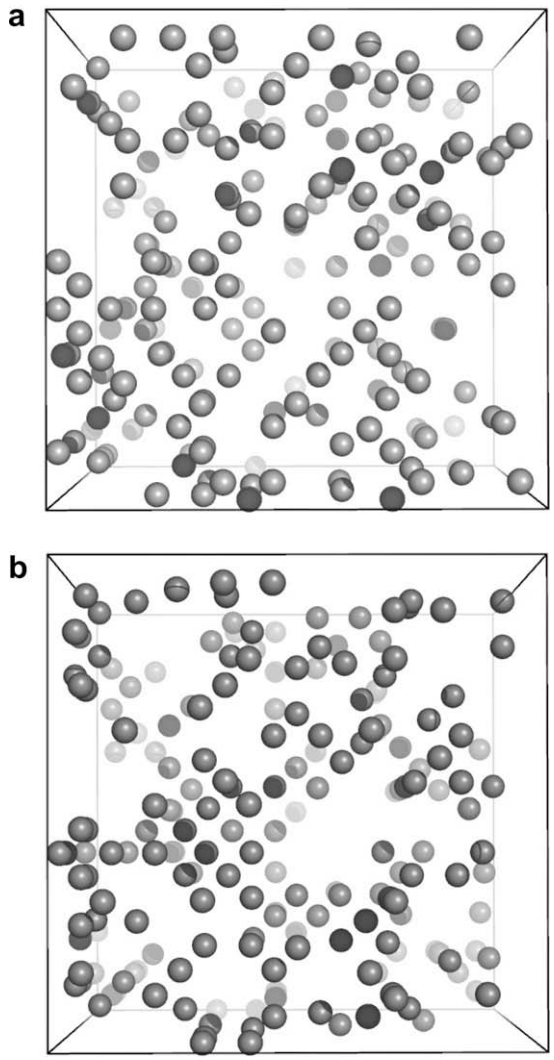

Fig. 3. Spatial distribution of Cr atoms for the Fe–9Cr alloy (a) before the simulation and (b) after the simulation. The dimension of a cube is $20a_o \times 20a_o \times 20a_o$ (a_o: lattice constant). The grey spheres represent Cr.

3. Results and discussion

3.1. Positron annihilation measurement

The positron lifetime spectra for five samples are shown in Fig. 1. We can see a small difference in the spectra between the unirradiated Fe (dotted line) and irradiated metals. 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. The results for the positron annihilation lifetime data of each sample are listed in Table 1. For the detailed analysis, the spectra for the irradiated samples were decomposed into two components, 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 vacancies which are formed due to irradiation. The lifetimes in the vacancy, which were experimentally determined [8], are 170 ps for Fe and 180 ps for Cr. The τ_2 components, listed in Table 1, clearly indicate that a certain amount of vacancies are distributed in the irradiated samples. No difference in the τ_2 component was, however, found for the initial Cr contents.

Fig. 2 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 ($<7 \times 10^{-3}$ $m_o c$, 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 induced by irradiation. 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. This result is consistent with the PA lifetime spectra. Moreover, 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 clusters with the neighboring Cr atoms but exist in isolated form.

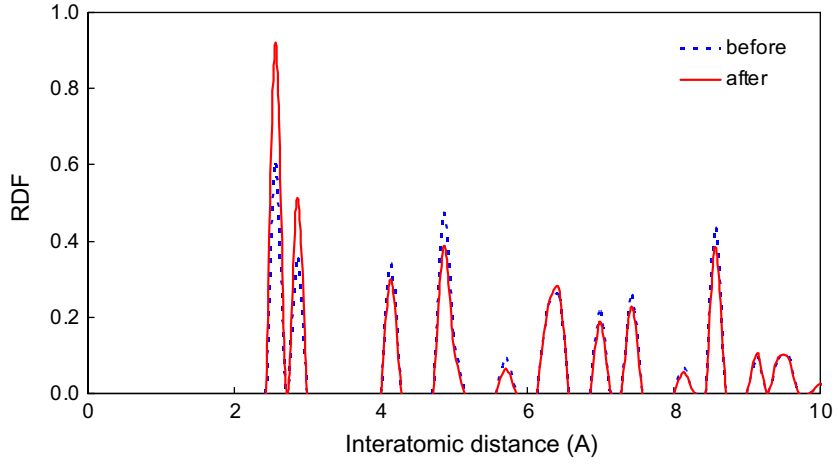

Fig. 4. Cr–Cr radial distribution function for the Fe–9Cr alloy obtained from the MMC simulations. It is assumed that initially the Cr atoms are distributed randomly.

3.2. Monte Carlo simulations

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. The Metropolis Monte Carlo (MMC) method provides a convenient tool to predict the atomic configuration of the microstructure by means of energy minimization techniques. The details of the MMC method are described well in Ref. [9]. 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 et al. [10]. The focal point in the MMC simulation is the role of the vacancies produced by electron irradiation in the formation of atomic clusters. We investigate the atomic configuration of the system through the MMC simulation by changing the initial Cr contents. The size of the simulation block is $20 \times 20 \times 20$ and the system temperature is set to be 50 °C. According to the irradiation condition described in Section 2, two vacancies can be created at most. Based on this fact, we only included two isolated vacancies in the MMC calculation. Each simulation for the three Fe–Cr model alloys was interrupted after 200 000 steps.

The spatial Cr distributions, which were initially random, in Fe–5Cr, Fe–9Cr, and Fe–15Cr were examined after the simulation. Fig. 3 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 form 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. These results are in agreement with the *ab initio* calculations of Olsson et al. [11], where the interactions of Cr atoms and vacancies are explained by the binding energy. Fig. 4 shows the Cr–Cr radial distribution function for the Fe–9Cr alloy, which represents the distribution of Cr atoms that are packed with each other in a system. We observed a big increase in the first and second peaks after the MMC simulation as compared to before the simulation, which represent clustering of Cr atoms. Similar behaviors were also found for the Fe–5Cr and Fe–15Cr alloys.

4. Conclusions

The present study demonstrates that the coupling of PA measurements and computer simulations 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 suggests that the swelling resistance of F/M steels under irradiation is related to the absence of interactions between Cr atoms and vacancies.

Acknowledgement

The authors would like to thank the Korean Ministry of Science and Technology for the promotion and sponsorship of the research work under 'National mid- and long-term atomic energy R&D program'.

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