Formation of Cu precipitates in a high-energy-particle-irradiated and thermally aged Fe-0.6%Cu alloy

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Abstract. The stability of Cu precipitates in thermally aged Fe-Cu alloys after high-energy particle irradiation was investigated using a mono-energetic positron beam apparatus. The S-parameters did not change with increasing incident positron energy above 15 keV, which means positron annihilation at the surface does not influence the Doppler broadening (DB) spectra. In a non-aged sample, the S-parameter increased, while the W-parameter did not change upon ion irradiation at above 10 keV. By contrast, in the thermally aged sample, the S-parameter increased, while the W-parameter decreased upon ion irradiation. The DB spectra of thermally aged and non-aged samples have nealy the same shape after high-energy particle irradiation. In both thermally aged and non-aged samples, almost all positrons annihilate with electrons in Cu, and vacancy clusters covered with Cu atoms are formed upon neutron irradiation. This agrees with the results of previous studies. However, the defect growth process may be different in aged and non-aged samples.

1. Introduction

Cu precipitates form in Fe-Cu alloys during thermal aging at high temperatures, as well as upon irradiation with high-energy particles [1,2], since Cu atoms are almost insoluble in α-Fe. Cu precipitates obstruct dislocation motion during deformation, which increases the hardness and decreases the ductility of the alloy, thus inducing embrittlement in Fe-based alloys containing Cu impurities, such as old commercial reactor pressure vessel (RPV) steels. Besides Cu precipitates, defect clusters, such as interstitial clusters and vacancy clusters, are also formed during irradiation, which further increase the hardness and decrease the ductility of RPV steels.

Positron annihilation spectroscopy can detect vacancy clusters and even single vacancies. Doppler broadening (DB) of positron annihilation radiation is a nondestructive technique for testing defect clusters. In the dominant decay mode of a thermal positron and electron, two gamma rays are emitted. In the laboratory frame, the energies of the two photons emitted by the annihilation of a positron and electron are different. This difference in photon energy is proportional to the longitudinal component of the electron-positron momentum in the direction of gamma emission. Through measurements of the photon energies, information about the momentum distribution of core electrons can be obtained. Thus, DB measurements can provide useful information about the distribution of elements around the annihilation site [3−6]. Positron annihilation spectroscopy using slow-positron beam techniques is very effective for investigating the near-surface microstructure of ion-irradiated materials [7].

It is well known that Cu atoms aggregate by a vacancy mechanism even in thermally aged Fe-Cu alloys. However, details of the process of precipitate formation in thermally aged Fe-Cu alloys and irradiated Fe-Cu alloys remain unclear. In the present study, samples were irradiated with ions and neutrons, and the stability of Cu precipitates in thermally aged Fe-Cu alloys was investigated.

2. Experimental

Fe-0.6 wt% Cu was prepared from Fe (99.99%, purity) and Cu (99.9%, purity) in vacuum using a high-frequency induction furnace. Two types of heat treatments were performed before irradiation. In one case, the samples were aged at 1223K for 0.5 h in a vacuum, and quenched in water. In the other case, specimens were aged at 773 K for 100 h in vacuum after being quenched in water, to obtain small Cu precipitates. Samples consisted of 10×10×0.1 mm sheets. Fe ion irradiation was performed at the Quantum Science and Engineering Center, Kyoto University, using a tandem Pelletron accelerator (model: 6SDH-2). The ion energy was 2.5 MeV, and the irradiation temperature was 573 K. The irradiation dose at damage peak area was 1 dpa. Figure 1 shows simulated depth distributions of damage in Fe irradiated by 2.5 MeV Fe ions. In order to investigate the irradiation depth dependence of the microstructural evolution of ion-irradiated samples, the DB measurements were performed using a mono-energetic positron beam apparatus at the Institute of High Energy Physics, Chinese Academy of Sciences. The incident positron energy was 0.18-20 keV. Figure 2 plots the depth profile of the positron stopping probability for energies of 5, 10, 15, and 20 keV. Both the irradiated and opposite side were measured to investigate the effect of irradiation. The parameters *S* and *W* are defined as the ratio of the low-momentum ($|P_L| < 3\times10^{-3}mc$) and high-momentum ($10\times10^{-3}\ mc < |P_L| < 27\times10^{-3}\ mc$) areas in the DB spectrum to the total area, respectively. Fission neutron irradiation was performed at Research Reactor Institute, Kyoto University. The irradiation dose was 4.3×10^{-3} dpa (2.0×10^{18} n/cm^2), and the irradiation temperature was 573 K. Coincidence Doppler broadening (CDB) measurements and positron annihilation lifetime measurements were carried out at room temperature using a Na-22 source for neutron-irradiated samples.

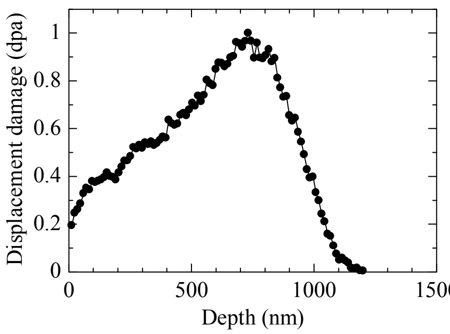

Figure 1. Depth distribution of damage in Fe irradiated by 2.5 MeV Fe ions.

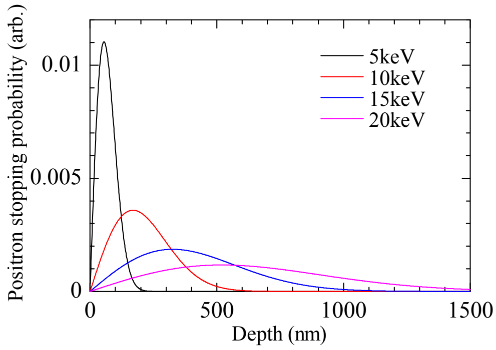

Figure 2. Depth profile of positron stopping probability for energies of 5, 10, 15, and 20 keV.

3. Results and discussion

Figures 3 and 4 show the dependence of the S- and W-parameter on incident positron energy, respectively. Figure 5 shows S–W plots at all incident positron energies. 1#, 2# and 3# denote the quenched sample irradiated with Fe ions, the sample irradiated with Fe ions after aging at 773 K and the sample aged at 773 K (non-irradiated), respectively. "B" denotes measurements made on the non-irradiated side. The S-parameter decreased and the W-parameter increased with increasing incident positron energy. S-parameters did not change with increasing incident positron energy above 15 keV. Positron annihilation at the surface did not affect the DB spectra above 15 keV. Comparison of 1# and 1#B reveals the saturation value of the S-parameter increased upon Fe ion irradiation while that of the

W-parameter did not change. This means that irradiation defects and Cu precipitates were formed during ion irradiation. If Cu precipitates had not formed during irradiation, the increase in the S-parameter due to the formation of irradiation defects would have led to a decrease in the W-parameter. Comparing 2# with 2#B shows that for the aged samples, the saturation value of the S-parameter increased while that of the W-parameter decreased upon Fe ion irradiation. The increase in the S-parameter indicates the formation of irradiation defects. As mentioned in the next paragraph, Cu precipitates can grow by irradiation even in an aged sample. Some of the irradiation defects promote the growth of aging-induced Cu precipitates, while the rest contribute to the formation of new Cu precipitates. The fact that the W-parameter is the same for both irradiated-quenched and aged samples (1# and 2#) indicates that positrons favour annihilation in irradiation-induced newly formed Cu precipitates over those formed by aging. Thus, if only vacancy clusters are formed and Cu precipitates do not grow during irradiation, the W-parameter would decrease more. In S-W plots, we can see the same trend in 2#B and 3#, and 1# and 2#. On the non-irradiated side of ion-irradiated samples without aging (1#B), Cu precipitates are not formed. Thus, these samples exhibit the lowest W-parameter. The same heat treatment is applied to 2#B and 3#.

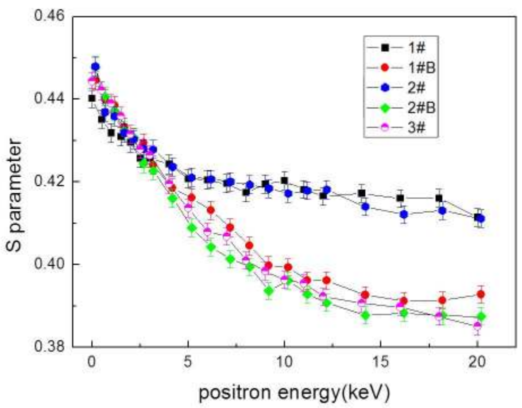

Figure 3. Dependence of S parameter on incident positron energy.

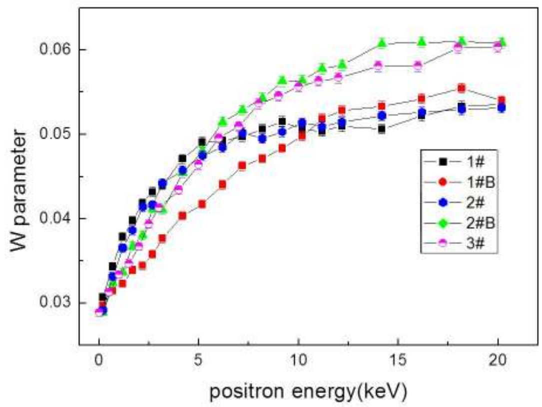

Figure 4. Dependence of W parameter on incident positron energy.

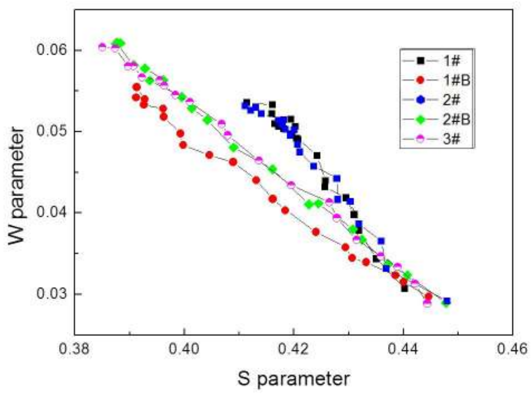

Figure 5. . S–W plots in all incident positron energies.

Figures 6 and 7 plot the CDB ratio curve of neutron-irradiated Fe-Cu alloys with respect to pure Fe and Cu, respectively. The CDB ratio curve of the non-aged Fe-Cu alloy peaks in the high-momentum region after neutron irradiation. As seen in figure 7, the CDB ratio curve of the Fe-Cu alloy aged at 773 K for 100 h has a small valley in the high-momentum region (15×10^{-3} mc < $|P_L|$ < 30×10^{-3} mc). Thus, in the aged sample, some of the positrons annihilate with electrons in Fe [3]. But, after neutron irradiation, the CDB ratio curves of the irradiated-quenched and aged Fe-Cu alloys are almost flat in the high-momentum region. Cu precipitates grow as a result of neutron irradiation and almost all positrons annihilate with electrons in Cu. Vacancy clusters covered with Cu atoms are formed by neutron irradiation not only in quenched sample, but also the aged sample. This result agrees with references 4 and 6. It is expected that they are also formed under Fe ion irradiation, and more positrons annihilate in them than in Cu precipitates formed by aging, because the latter do not contain vacancy-type defects. The size and density of Cu precipitates cannot be determined by DB measurements alone; transmission electron microscopy and other characterization techniques are necessary. Table 1 shows positron annihilation lifetimes of neutron-irradiated quenched and aged Fe-Cu alloys. The vacancy cluster size and density are slightly larger in the aged sample than in the quenched sample. In the aged sample, the number of Cu atoms that do not form Cu precipitates is smaller than in the quenched sample before irradiation. This may influence the growth of Cu precipitates. The defect growth process may be different for the aged and non-aged samples. Different irradiation doses are required to investigate these possibilities.

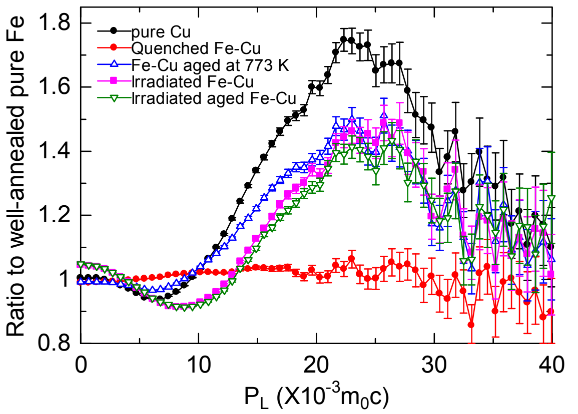

Figure 6. Ratio curves of CDB spectra of pure Cu, quenched Fe-Cu alloy, Fe-Cu alloy aged at 773 K, irradiated quenched Fe-Cu alloy and irradiated aged Fe-Cu alloy with respect to that of pure Fe.

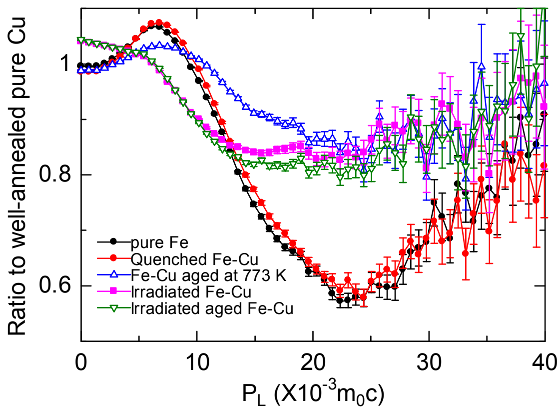

Figure 7. Ratio curves of CDB spectra of pure Fe, quenched Fe-Cu alloy, Fe-Cu alloy aged at 773 K, irradiated quenched Fe-Cu alloy and irradiated aged Fe-Cu alloy with respect to that of pure Cu.

Table 1. Positron annihilation lifetimes of neutron-irradiated Fe-Cu alloys.

	Mean lifetime (ps)	Short lifetime (ps)	Long lifetime (ps)	Long lifetime intensity (%)
Quenched	172 ± 1	129 ± 1	463 ± 5	13 ± 1
Thermally aged	186 ± 1	126 ± 1	479 ± 4	17 ± 1

4. Summary and future plan

The stability of Cu precipitates in thermally aged Fe-Cu alloys was investigated. DB spectra of thermally aged and non-aged samples are almost the same after both ion and neutron irradiation. Almost all positrons annihilate with electrons in Cu, and vacancy clusters covered with Cu atoms are formed upon neutron irradiation in both aged and non-aged samples. Determining the exact size and density of Cu precipitates requires detailed observation of the defect structure by transmission electron microscopy, three-dimensional atom probes, etc. Further experiments using lower irradiation doses and shorter aging times will be conducted in the near future.

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