Cu Precipitates in Fe Ion Irradiated Fe-Cu Alloys Studied Using Positron Techniques

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Abstract. In this paper, we summarized our recent experimental results on Fe-Cu model alloys irradiated by Fe ion. Two kinds of Fe-Cu alloys with 0.3%Cu and 0.6%Cu were prepared and irradiated by 2.5 MeV Fe ion at 573 K. Irradiation dose is 0.1 dpa and 1.2 dpa for each type alloy respectively. Positron annihilation techniques of slow positron beam were used to investigate the irradiation induced defects. Results show that the S parameters are higher in the specimens with high irradiation dose, but the intensity of Cu peaks of CDB is lower. It indicates that the precipitation of Cu atoms formed easily as lower irradiation dose.

1. Introduction

The formation of Cu precipitates plays a key factor in the reactor pressure vessel (RPV) steels as irradiation induced by neutrons [1, 2]. Extensively studies focused to the formation and migration of the Cu precipitates at past decades, because the precipitation of Cu atoms can increase the hardness and decrease the ductility, especially it can leads to embrittlement in irradiation induced RPV steels [3-5]. As one of the menaces to the safety of nuclear power reactor operating, to understand the formation mechanism of irradiation induced defects and Cu precipitates in RPV steels is one of the important research subjects [6-9].

As the complicated irradiation effects in commercial RPV steels, such phenomena of radiation induced segregation and precipitation is important as the migration of Cu atoms excess point defect fluxes and super saturation during the displacement and energy deposition of implanted particles. It is well know that substantial recombination and defect clustering can be produced by neutron irradiation in metals as high energy recoils of fast neutrons within dense cascades [10, 11]. Recent work has suggested that remarkable precipitates formed during post-radiation procedure with temperature affection [3-5]. One assumed viewpoint is that embryos of Cu precipitates can also be nucleated and contribute to the hardening directly [11, 12]. Compared to the neutron irradiation effects in RPV steels, high-energy heavy particle can produce similar displacement damage within acceptable experiment research. Fe ion produced by accelerator was usually performed as the implanted particles, which can produce complex defects without impurity elements.

The mechanism of the formation of defects and nucleating and clustering of Cu atoms are still unclear under irradiation performed, and the evolution of defects with radiation dose and Cu contents still require further investigation, especially in the nucleation stage of the Cu precipitates [4]. But it is little know experimentally because the difficulty of observation method, such as transmission electron microscopy and atom probe tomography [1, 13, 14]. Positron annihilation techniques (PAT) have

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been used to investigate tiny Cu precipitates, defects and their complexes in Fe-Cu alloys [5], especially the positrons can be implanted to the interesting depth with appropriate energy using slow positron beam. Previously, we have published several papers related to Cu precipitates in Fe-Cu alloys. In the present study, PAT of slow positron beam were carried out to elucidate the effects of radiation dose and Cu concentration on defects and Cu precipitates formation in Fe ions irradiated Fe-Cu alloys.

2. Experimental

2.1. ***Materials*****:** Two series of Fe-Cu alloys with 0.3 and 0.6 mass% Cu were smelted from Fe (99.99% purity) and Cu (99.9% purity) in vacuum using a high-frequency induction furnace. Specimens, with a thickness of 0.2 mm and 8 mm square sheet, were irradiated with Fe ions after quenched in water from 1173 K. Fe ions with 2.5 MeV were implanted using a tandem Pelletron accelerator (model: 6SDH-2) of Quantum Science and Engineering Center, Kyoto University. Irradiation dose performed to 0.1 dpa and 1.2 dpa for each series at 573 K, respectively.

2.2. ***Positron annihilation techniques:*** PAT experiments were performed at slow positron beam facility of institute of high energy physics (IHEP). Slow positrons are generated by a 50 mCi (2005) ^{22}Na radiation source. During the measurement of positron annihilation Doppler broadening spectroscopy (DBS), beam energy was changed from 0.18 keV to 25.0 keV. In this present study, two parameters were introduced to DBS to characterize the information of specimens, which are named S and W parameters respectively. S parameter was defined as the ratio of the counts in the centre area near the 511 keV (510.2-511.8 keV) of γ-rays to the total counts of spectrum (503.34-518.66 keV). And W parameter was defined as the ratio of the counts in the summed areas of 514.83–518.66 keV and 503.34–507.17 keV to the total counts of spectrum.

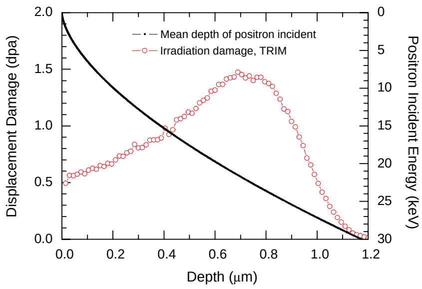

Figure 1. Depth distribution of damage calculated using the TRIM code for the Fe-Cu alloy irradiated with Fe ions to 1.2 x 10^{17} Fe/m^2.

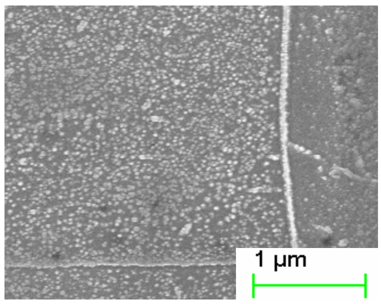

Figure 2. SEM image of Fe-0.3%Cu alloy with a irradiation dose as 0.1 dpa induced by 2.5 MeV Fe ions at 573 K.

Compared to the DBS, two HPGe detectors were used to detect the γ-ray pairs emitted by annihilation of positron and electron in coincident Doppler broadening (CDB) techniques. The information of elements around the positron trapping position can be described, because the momentum of atom-bound electron can be measured in CDB if it was annihilated [16, 17]. The damage peak of Fe ions in Fe-Cu alloys was about 0.7 μm from the irradiation surface, according to the simulation using the TRIM code. In order to investigate the precipitation of Cu atoms, CDB measurements with a mono-energetic positron beam were performed to the irradiation damage area in this present study. The implanted mean depth (nm) of positrons may be estimated by empirical equation as $R=40\times E^{1.6}/\rho$, using parameters of positron energy E with keV and materials density ρ with g/cm^3 [18-20]. The results of TRIM code simulation and the mean depth of positron incident are shown as Figure 1.

3. Results and discussion

3.1. ***SEM results:***The surface microstructure of Fe-Cu alloy was observed by SEM (Hitachi S-4700). Figure 2 showed the result of the Fe-0.3%Cu alloy with a radiation dose as 0.1 dpa induced by Fe ions at 573 K. It shows that some crystalline structure formed with many tiny clusters on the surface, especially aggregated at the grain boundary. The concentration of Cu distributed on these tiny clusters and the grain boundary is high than mean distribution, which confirmed by the energy dispersive spectrometer (EDS) of SEM. Some paper has also reported that Cu atoms migrated and aggregated at the grain boundaries with the diffusion of vacancies defects [21].

3.2. ***PAT results:*** Figure 3 showed the S parameters with positron energy changed from 0.18 keV to 20 keV. One un-irradiated Fe-Cu specimen was also measured as the reference specimens. According to the description in Figure 1, the positrons were implanted near 0.7 μm from surface and almost closing to the area of Fe ions deposition. Results show that the S parameters near to 20 keV are dramatically increased after irradiated, but there is no clearly difference affected by Cu content and irradiation dose. Because of closing to the Fe ions deposition area, the defects may be saturated to trapping positrons to annihilate. At the range of positron energy changed from 2 keV to 15 keV, the S parameters of specimens with 1.2 dpa is larger than those with 0.1 dpa. It indicated that more defects were produced at the area of ions track with high irradiation dose. The affection to S parameters contributed by Cu concentration can also be negligible. And the S parameters lower than 2 keV may be contributed by the surface defects, as shown in Figure 2.

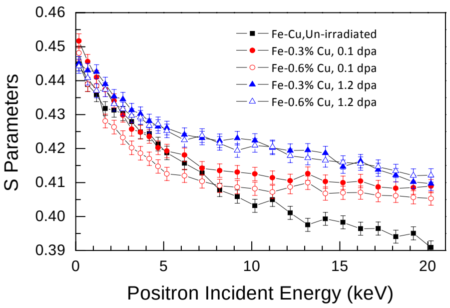

Figure 3. S parameters with positron energy for Fe-Cu alloys with different Cu content and irradiation dose.

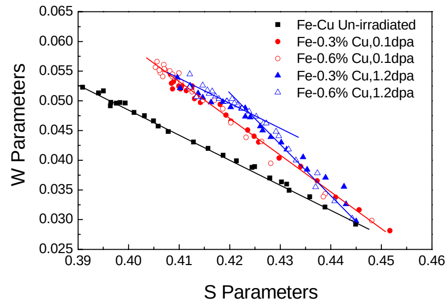

Figure 4. S-W curves for Fe-Cu alloys with Cu content and irradiation dose. The straight line was drawn only as guide eye.

As the W parameter was usually used to describe the information of core electrons annihilated with positrons, the slopes of S-W plots can present the annihilation mechanism of positrons after trapped [17]. The S-W plots of specimens are shown in Figure 4. It shows that the slopes changed clearly in irradiated alloys compare to un-irradiated one. And an inflection potion appeared clearly in the specimens with 1.2 dpa, which is corresponding to the 2~15 keV of positron energy range. It indicates that the mechanism of positron annihilation was changed in 1.2 dpa specimens. It may be contributed more by positrons annihilation trapping to defects. That can also be explained by the change of S parameters affected by irradiation dose, as shown in Figure 3.

According to the results calculated by TRIM, CDB experiments were performed as the positron energy with 13 keV, which related to the depth near 0.3 μm of Fe-Cu alloys from irradiation surface. The results with ratio to pure Fe are shown in Figure 5. It shows that a typical peak was observed near to 24 (10^{-3} m_0c) in irradiated Fe-Cu alloys, same momentum with the peak of pure Cu. The difference at low momentum related to the formation of defects induced by irradiation [22], as shown in small graph. Comparing with the results of S parameters and S-W plots, more detailed information can be investigated using CDB experiments, especially related to high momentum as a range with 13~30 (10^{-3} m_0c). In which, the intensity of Cu peaks are higher with 0.1 dpa dose than 1.2 dpa. It means that most of the positrons annihilated with electrons of Cu at 0.1 dpa. It can be attributed to that the Cu precipitates can formed easily within the small defects induced with 0.1 dpa, or that the increasing of counts related to free electrons affect to the decreasing of Cu peaks in 1.2 dpa specimens. The other reasonable explain is that many tiny Cu precipitates with vacancies type defects were produced in 0.1 dpa specimens, and positrons can be trapped easily by these vacancies type defects [15]. But in 1.2 dpa specimens, those tiny Cu precipitates and vacancies are formed to clusters and complex clusters, which lead to the increasing of S parameters and decreasing of Cu peaks [4, 23]. The change of Cu peaks with Cu content in 0.1 dpa specimens is few, but is increased with Cu content increasing in 1.2 dpa specimens. This can also attribute to the clustering of vacancies induced by high irradiation dose, and easier to form complex clusters in lower Cu content specimens [3, 24].

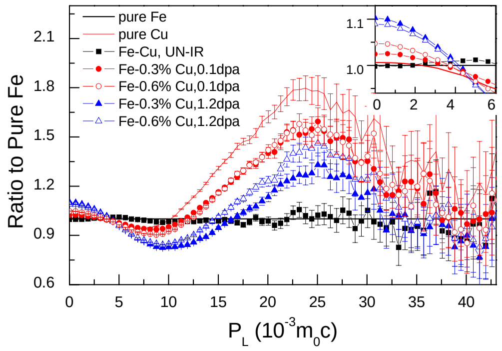

Figure 5. CDB results as the positron energy with 13 keV, including pure Cu, un-irradiated Fe-Cu alloy, Fe-0.3%Cu alloys with 0.1 dpa and 1.2 dpa irradiation dose, and Fe-0.6%Cu alloys with 0.1 dpa and 1.2 dpa irradiation dose, respectively. Small graph is the detailed information as the momentum lower than 6 (10^{-3} m_0c).

4. Conclusion

The PAT were performed to study the formation of defects and precipitation of Cu atoms, especially the CDB based on the slow positron beam were used to investigate the Cu precipitates formed with complex defects induced by Fe ions irradiation. The defects formed in Fe-Cu alloys at the ions track area and deposition area, and more defects produced as the irradiation dose increased. Precipitation of Cu atoms is affected by the irradiation dose and the Cu content in Fe-Cu alloys. Further experiments will be performed to investigate the migration of defects and tiny Cu precipitates with post-annealed time and temperature.

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