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Positron Annihilation Study of Electron-Irradiated Fe-Cu Alloys

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It is well known that the recovery of defects in iron alloys is affected by impurity atoms. Especially, the embrittlement process after high energy neutron irradiation in low alloy ferritic steels could be strongly influenced by small amount of impurity copper atoms. Fundamental process of this effect was studied by positron lifetime measurements with isochronal annealing in three series of Fe-Cu alloys of copper concentrations 0.02wt.%, 0.22wt.% and 0.05wt.% after 28MeV electron irradiation at 77K. The maximum value of positron lifetime τ_2 was about 280psec in Fe-0.02wt.% Cu, and about 190psec in Fe-0.22wt.% Cu, which was much smaller than that of pure iron (450psec). This showed that the vacancy cluster (microvoid) formation was disturbed as the concentration of copper atoms increased, and the vacancy migration temperature in Fe-Cu alloys was about 280K, which is higher than that of in pure iron[1) 2)]. It was also observed that the positron lifetime certainly decreased in higher temperature region in all alloys, which differs from other positron lifetime measurement data of various binary iron alloys, i.e., obtained by Corbel et al[2)3)].

1. Introduction

This study is mainly aimed at understanding the fundamental process of the embrittlement of pressure vessel steels under the high energy neutron irradiation environments. Although, it is well known impurity copper atoms enhance the important radiation embrittlement in low alloy ferritic steels, but its process has not been clarified yet[4)]. Therefore, it is important to know how the copper atoms effect the migration and aggregation of radiation-induced point defects. For this purpose, it was attempted by means of positron annihilation lifetime measurement to make clear the correlation between the vacancies and copper atoms in this study.

At present, Fe-Cu alloys have been investigated after 28MeV (6×10^{18} electrons/cm^2) electron irradiation 77K. Electron irradiation induces Frenkel-pairs (vacancy and interstitial pairs) into specimens. And positron annihilation technique yields the information on the vacancy type defects at a microscopic scale beyond the resolution limit of the electron microscopy. In the present paper, we will describe the influence of copper atoms on vacancy-clustering.

2. Experimental

All samples were prepared from pure metals, such as pure iron ATOMIRON 4N (Showa Denko Co.) and pure copper (Johnson Matthey Chem. Ltd.), by floating zone leveling method

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under hydrogen atmosphere. Since alloys were prepared in high-purity hydrogen gas, total amount of impurities were reduced to a very low level. The samples for each alloys were cut into two pieces of size 0.25mm × 8mm × 8mm for positron annihilation measurements. After above procedure, all specimens were chemically polished in H_2O_2 and HF bath (H_2O_2 : HF = 20 : 1). All specimens were heat treated for 1h at 750℃ in vacuum and then quenched in a water at 15℃, this treatment certified solid solution of copper atom in ferritematrix. And then, they were chemically polished again. Electron irradiations were carried out in flowing liquid nitrogen (77K) by using 28MeV KURRI-LINAC to the final dose of about 6×10^{18} electrons/cm^2. The positron annihilation lifetime measurements were performed in a crystat at 100K. As a positron source we used ^{22}NaCl of about 10μCi. And it was sandwiched between two identical specimens. During the isochronal annealing treatments, the specimens were heat treated for 20min/20K at each step in all temperature range. The measured lifetime spectra were analyzed by means of "RESOLUTION" program (the least square fitting program produced in Risø.). Applying two-state-trapping-model, all lifetime spectra could be fitted with sufficient precision.

3. Results and Discussion

Fig. 1 shows the result of positron lifetime measurement in Fe-0.22wt.%Cu alloy before irradiation, obtained in the annealing range from 300K to 900K. In whole temperature range, the value of lifetime τ is about 112psec which is almost the same value as that in pure iron before irradiation. This means that positron annihilation almost happens at the ferrite-matrix. On the other hand, the positron trapping at sites of copper-cluster has been

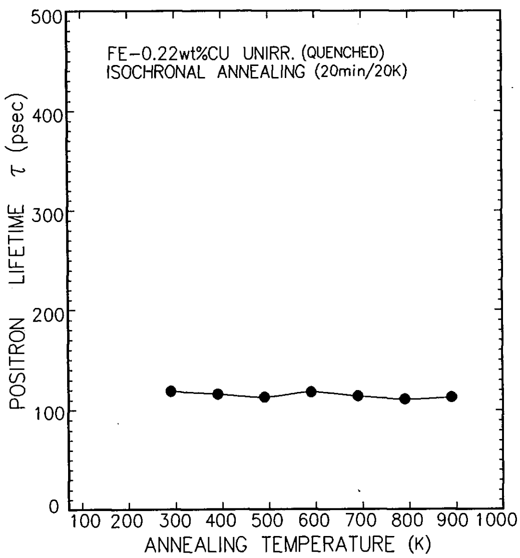

Fig. 1 Isochronal annealing result for Fe-0.22wt.%Cu before irradiation obtained by positron lifetime measurement.

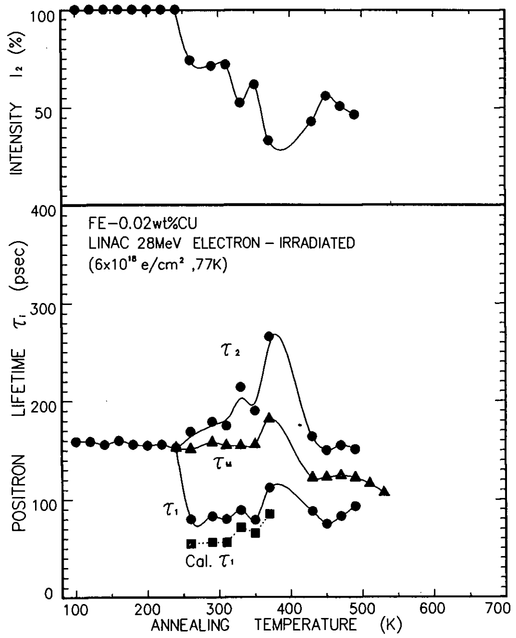

Fig. 2 Isochronal annealing result for Fe-0.02wt.%Cu irradiated by electrons obtained by positron lifetime measurement.

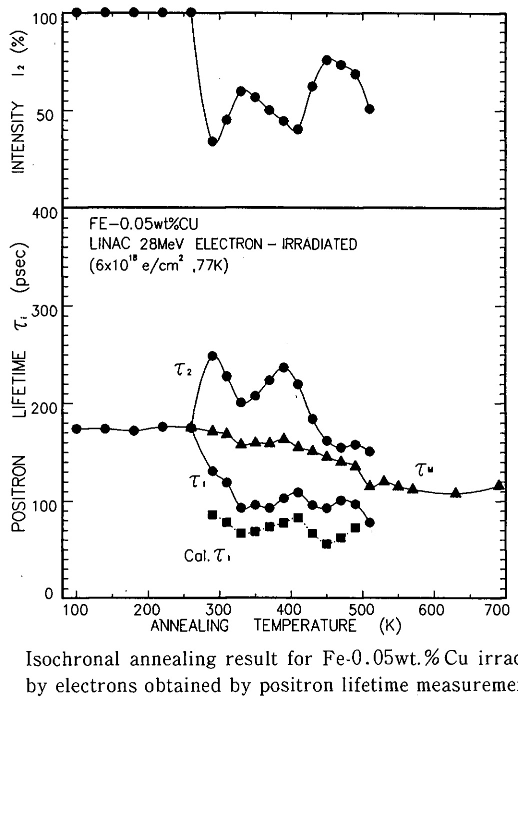

Fig. 3 Isochronal annealing result for Fe-0.05wt.%Cu irradiated by electrons obtained by positron lifetime measurement.

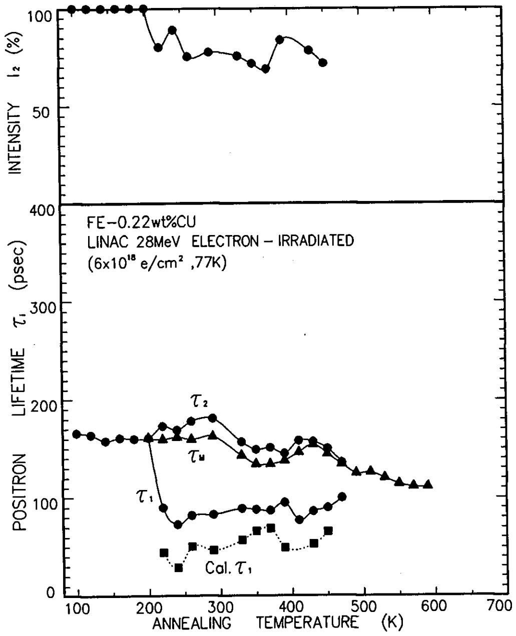

Fig. 4　Isochronal annealing result for Fe-0.22wt.%Cu irradiated by electrons obtained by positron lifetime measurement.

reported in higher concentration alloy (Fe-0.8wt.%Cu)[5]. This suggests the copper atoms in our specimen could not form precipitates without the radiation-induced defects.

In **Fig. 2, 3** and **4** isochronal annealing results for Fe-0.02wt.%Cu, Fe-0.05wt.%Cu and Fe-0.22.wt%Cu are shown, respectively. Vacancy concentration is considered to be about 100ppm which is high enough to trap all positrons. In the low temperature range, the value of positron annihilation lifetime is about 170psec which corresponds to the value at a single vacancy site. The appearance of longer positron lifetime component, associated with vacancy migration and agglomeration which is called stage III, was observed at about 280K for all of these cases. This temperature is different from that reported by Corbel et. al[6]. At this temperature, two component decomposition of the lifetime spectrum became possible, which suggests that the interaction between the vacancy and copper atom is strong enough and vacancies are trapped by copper atoms until this temperature. This behaviour is considered to be favorite to the formation of copper-rich complexes because the vacancy-copper interaction prevents the annihilation of vacancies at self-interstitial atom or sink sites. The copper-vacancy clusters are stabilized by its strong binding. Vacancy clustering behaviour in the isochronal annealing process is dependent on the concentration of copper atoms. It is observed that long lifetime τ_2 decreased in the temperature range, 300 − 350K and above 400K. The former decrease suggests vacancies (V_1) were emitted from copper-vacancy complexes ($Cu_1 - V_n$: $n = 1 \sim 4$) and began to migrate at 300K which assisted the formation of copper precipitates, namely, different copper-vacancy complexes (Cu_n − V: $n \geq 2$) were formed, which have shorter positron lifetime because number of

Table 1 Atomic size factor calculated by King et. al. $=(1/\Omega)(d\Omega/dc)=(3/a)(da/dc)$. ($\Omega$: atomic volume, c : solute concentration, a : lattice parameter)

Atomic size factor in pure iron. calculated by King et. al.	
Alloy	Size factor (%)
Fe − Au	44.16
− Sb	36.40
− Mo	27.51
− Cu	17.53
− V	10.51
− Mn	4.81
− Ni	4.65
− Cr	4.36
− Co	1.54
− Si	−7.88

vacancies is less, or free volume could be decreased due to larger atomic size of copper stoms mentioned below. The strong interaction between vacancy and copper atom is considered to be due to the difference of atomic size factor calculated by King et. al[7] in **Table 1**. Actually the maximum value of positron lifetime τ_2 is 280psec at most. According to the calculation performed by Puska and Nieminen[8], microvoids of positron lifetime 280 spec consist of about four vacancies, and this seems to decease as copper content increases, for example, τ_2 in Fe-0.22wt.% Cu is about 180psec. On the other hand, long lifetime τ_2 decreased again with increasing relative intensity I_2 above 400K. This could be considered to be the decomposition of microvoids due to recombination with interstitial atoms at this temperature and positrons might be trapped by copper-rich zones, which gives shorter lifetime. After this, positron lifetime decreased to the bulk value, which means copper atoms were dissolved into ferrite-matrix again. More investigations will be needed to make clear the detailed mechanism.

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