Effect of Cu content on the defect evolution in Fe-Cu alloys investigated by PALS

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Abstract. The effect of Cu content on the evolution of defects in Fe-x%Cu alloys (x= 0.15, 0.3, and 0.6) were investigated using Positron Annihilation Lifetime Spectroscopy (PALS). The vacancy-type and Cu-vacancy complexes defects were respectively produced by quenching from 1173 K and cold-rolled deformation followed by isochronal annealing. The PALS results with isochronal annealing showed that the temperature of defects recovery increased in deformed Fe-Cu alloys with Cu content. The increment of Cu content also restrained the migration of vacancies in as-quenched Fe-Cu alloys.

1. Introduction

Fe-Cu model alloy has been always regarded as one of the most suitable systems for studying the Cu precipitates and the evolution of micro defects in the reactor pressure vessel (RPV) steels [1,2]. Extensive studies have showed that the increment of the ductile-to-brittle transition temperature (DBTT) was due to the tiny Cu precipitates in RPV steels [1,3,4]. Since the influence of small amount Cu element on DBTT of steel used for RPV steels is a serious problem, the content of the Cu impurity must be limited strictly in commercial reactor structural materials. It is well known that Cu atoms have a very low solubility in Fe-base alloys and the Cu precipitates formed easily in Cu-bearing steels during deformation, irradiation and aging. Formation of Cu precipitates, evolution of defects and the interaction between Cu precipitates and defects have been widely investigated [5,6]. Formation of Cu-vacancy clusters and the deformation-induced vacancies enhanced the diffusion of Cu atoms were also investigated using positron annihilation spectroscopy in rolling deformed Fe-Cu alloys with isochronal treatment [7]. In addition, Nagai et al. investigated the interaction of irradiation-induced vacancies and Cu aggregations in binary Fe-Cu alloys, they found that irradiation could lead to a vacancy-solute complex which would speed up the precipitation [8]. Although the Fe-Cu binary alloys have been extensively studied, the effect of Cu content on the evolution of defects along with the interaction between Cu atoms and vacancies in quenched and deformed Fe-Cu alloys are still poorly understood. However, the mechanism of the interaction of Cu atoms with defects plays a critical role in the properties of Fe-Cu alloys.

It is difficult to probe experimentally using conventional detection means such as Transmission Electron Microscopy (TEM) as many of these defects are on atomic scale. Positron annihilation techniques (PAT) have been used to investigate tiny Cu precipitates, defects and their complexes in

Fe-Cu alloys [9]. Importantly, PALS has been widely applied as one of the few techniques to detect the evolution of vacancy-type defects sensitively and selectively in metals [10-12].

In order to clarify the effect of Cu content on the evolution of defects and the interaction between Cu atoms and vacancies, PALS was performed on three sorts of Fe-x%Cu alloys (x= 0.15, 0.3, and 0.6) treated with different ways. Deformation and quenching were used to produce defects in Fe-x%Cu alloys. Isochronal annealing was a common method to study the evolution and recovery of defects.

2. Experiments

As specimens, Fe-x%Cu alloys (x= 0.15, 0.3, and 0.6) were arc melted from Fe (99.99 % purity) and Cu (99.999 % purity) in vacuum using a high-frequency induction furnace. After melting, solution treatment of the samples consisted of heating at 1073 K for 24 h, and then well-annealed at 1173 K for 0.5 h in vacuum, followed by quenching into ice water to introduce vacancies [13]. The other series of specimens were cold-rolling deformed to 30% in order to induce the complex defects after quenching. After introduction of defects by quenching and deformation, the quenching specimens were isochronally annealed for 30 min in a vacuum of 10^{-5} Pa from 373 K to 673 K; and the deformed specimens was annealed from 373 K to 973 K. PALS analysis was carried out to characterize the defects in Fe-Cu alloys using a fast-slow coincident ORTEC system with a time resolution of 200 ps (full width at half maximum). The ^{22}Na positron source was placed between the two pieces of samples, and then the "sample-source-sample sandwich" was placed between the two BaF_2 detectors to acquire the lifetime spectra. Each spectrum was accumulated to a total of 2×10^6 counts, which aimed to reduce the statistical error in the calculation of lifetimes. The positron lifetime spectra were analyzed by the LT-9 software [7]. Positron annihilation lifetime and intensity could reflect the defect size and density information.

3. Results and Discussion

The results of isochronal annealing for as-quenched Fe-x%Cu alloys with 30% deformation obtained by PALS are shown in Figure 1. τ_m is the mean lifetime which reflects the total amount of vacancy-type defects in alloys. The long lifetime τ_2 corresponds to the size of vacancy clusters [14]. I_2 is the intensity of τ_2 which correlate the defect density. The short lifetime τ_1 may be contributed by positron lifetime of interstitial atoms and other defects, such as dislocation or bulk material [15]. It is seen that the value of τ_m in Fe-x%Cu alloys after 30% deformation were about 150 ps, and the mean lifetime of a perfect lattice in Fe was about 106 ps [16]. The τ_m is distinctly longer than the lifetime of Fe bulk, which indicated that a significant amount of defects were induced by deformation. The long lifetime τ_2 remained almost constant value after isochronal annealing at the temperature region 373 K-523 K, while the intensity of τ_2 increased gradually. The reason may be that a small amount of deformation-induced dislocations could trap vacancies, which could be released gradually with the increment of annealing temperature and finally formed to vacancy clusters or annihilated at the vacancy sinks, as the increasing of long lifetime (τ_2) intensity (I_2). As shown in Figure 1, the long lifetime τ_2 for Fe-x%Cu alloys all increased to around 200 ps after isochronal annealing at 573 K, suggesting the vacancies aggregated and formed the vacancy clusters. It was worth noting that the annealing behavior of the long lifetime in Fe-0.15%Cu was slightly different from that in Fe-0.3%Cu and Fe-0.6%Cu from 523 K to 623 K, the long lifetime in Fe-0.15%Cu increased to around 223 ps and remained stable, while the τ_2 of Fe-0.3%Cu and Fe-0.6%Cu decreased to around 160 ps subsequently. It indicated that the behavior of defects from 523 K to 623 K related to Cu content, namely, the increment of Cu content may promote the occupation of vacancies by Cu atoms and benefit for the formation of Cu-vacancy complexes further. Thus the increase of Cu content may enhance the separation of vacancy clusters in Fe-Cu alloys. On the other hand, when the mean lifetime decreased to the lifetime of a perfect lattice in pure Fe (~106 ps), this marked the complete recovery of defects [16]. The results of the mean lifetime in Figure 1 show that the perfect recovery temperature was 923 K in Fe-x%Cu (x= 0.15, 0.3) and 973 K in Fe-0.6%Cu. Cu-vacancy complexes had gradually transformed to Cu precipitates around 823 K after annealing and Cu precipitates may be formed on dislocations in matrix. The recovery process of the precipitates is strongly affected by the Cu content, while the recovery temperature corresponds to the solubility limit. As a result, it could be concluded that Fe-0.6%Cu had a higher recovery temperature compared to Fe-x%Cu (x=0.15, 0.3) because of the solubility limit, namely Cu.

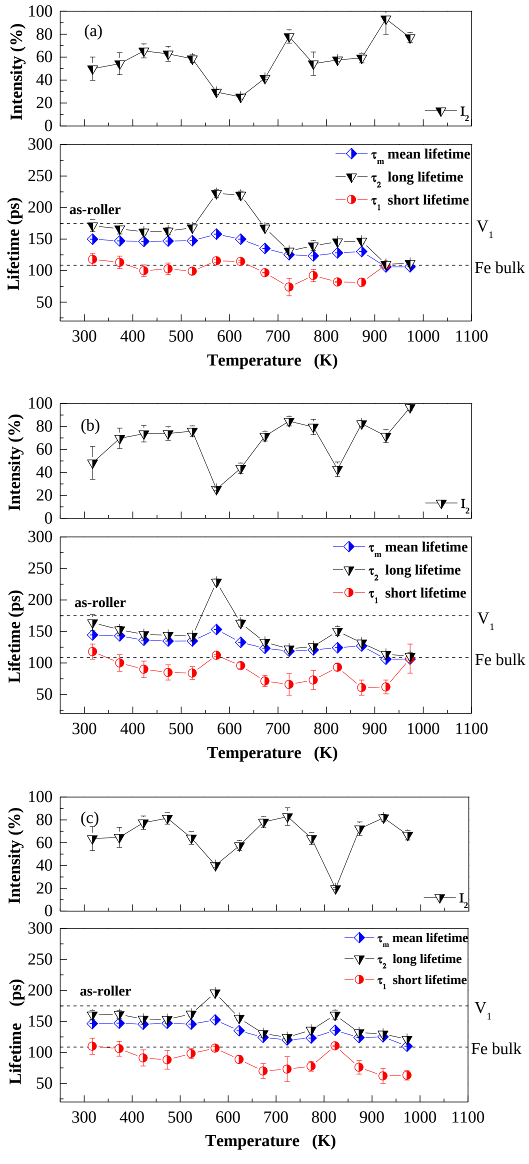

Figure 1. PALS for quenched Fe-x%Cu alloys with 30% deformation. (a) Fe-0.15%Cu (b) Fe-0.3%Cu (c) Fe-0.6%Cu. The calculated values for bulk and V_1 in pure Fe are shown by horizontal dashed lines.

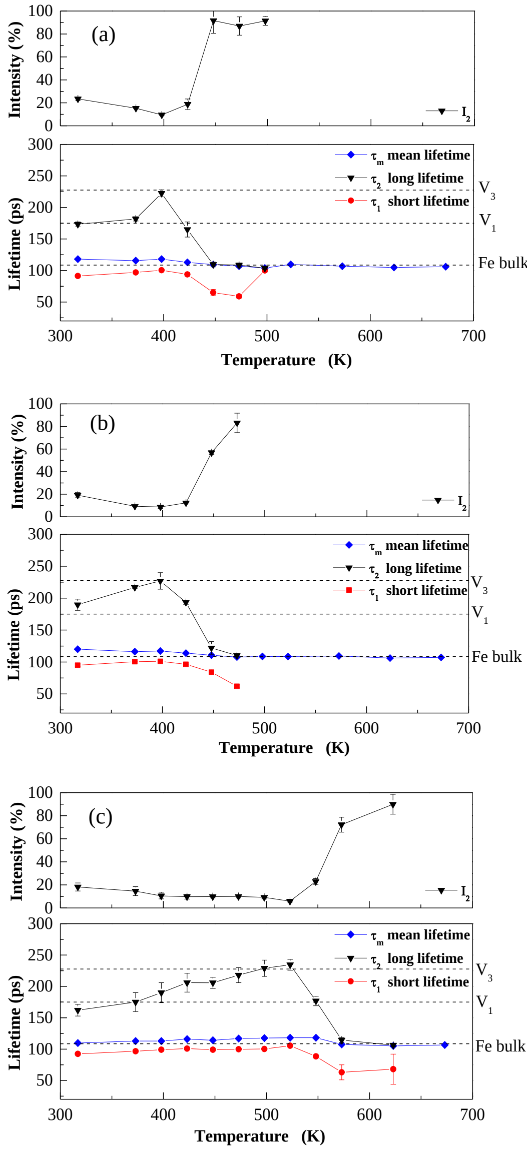

Figure 2. PALS for only quenched Fe-x%Cu alloys. (a) Fe-0.15%Cu (b) Fe-0.3%Cu (c) Fe-0.6%Cu. The calculated values for bulk, V_1 and V_3 in pure Fe are shown by horizontal dashed lines

In order to further investigate the influence of Cu content on the evolution of vacancy-type defects in Fe-Cu alloys and rule out the effect of deformation-induced Cu-vacancy complexes on the behaviour of vacancy-type defects, the only quenched specimens were investigated by PALS. As shown in Figure 2, the complete recovery temperature of Fe-0.15%Cu, Fe-0.3%Cu and Fe-0.6%Cu after isochronal annealing is 448 K, 473 K and 623 K, respectively. This phenomenon also demonstrated that the complete recovery temperature of defects increased with the Cu content because of the solubility limit. The complete temperatures for deformed Fe-x%Cu alloy in Figure 1 were distinctly larger than those for quenched samples in Figure 2. Therefore, Cu-vacancy complexes induced by deformation pushed the complete recovery temperature higher indirectly. As shown in Figure 2, the long lifetime in Fe-0.15%Cu and Fe-0.3%Cu increased to around 227 ps at 398 K after annealing, which meant single vacancies migrated and formed clusters. However, the corresponding temperature in Fe-0.6%Cu was 523 K. This also demonstrated that Cu content had an effect on the migration of vacancies in as-quenched Fe-Cu alloys. To be precise, Cu atoms may restrain the migration of vacancies.

4. Conclusion

The effect of Cu content in Fe-x%Cu (x= 0.15, 0.3, and 0.6) on the evolution of defects were investigated systematically by PALS. In summary, according to the results of PALS, the variation of the temperature where three sorts of specimens completely recovered revealed that the complete recovery temperature presented a significant increase with the increase of Cu content. It could be assumed that Cu precipitates transformed from Cu-vacancy complexes induced by deformation and subsequent isochronal annealing pushed the complete recovery temperature higher because of the solubility limit. In addition, Cu content had an effect on the migration of vacancies in as-quenched Fe-Cu alloys. To be precise, Cu atoms may restrain the migration of vacancies.

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