

Application of Positron Annihilation Spectroscopy to Study the Relationship Between Microstructure and Metallurgical Property of a Commercial Pure Copper

M.S. Abd El Keriem

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The microstructure variations of cold-rolled (80% reduction in thickness) commercial pure copper (99.5%) after isochronal annealing between room temperature (RT) and 1223 K were studied by using positron annihilation lifetime (PAL) microhardness measurements and optical and scanning electron microscopy (SEM) observations. The results indicate that the behavior of the short lifetime (τ_1) with the annealing temperature is nearly similar to the behavior of the microhardness measurements. The measured values of τ_1 were found to be higher than the bulk lifetime value at all temperatures except at the temperature interval 823-923 K. This could be attributed to recrystallization and grain growth in the copper. However, the trend of the intensity (I_2) and the trapping rate (K) with annealing temperature are the same. The existence of two peaks for both I_2 and K are probably due to the occurrence of two recrystallization stages.

Keywords commercial copper, grain growth, positron annihilation, recrystallization

1. Introduction

In some applications, nondestructive techniques are required to inspect different types of defects. The use of positron annihilation spectroscopy (PAS) for the study of lattice defects in solids is based on two facts. First, the positrons are antiparticles to electrons so that the positron may annihilate the electron. Second, when the positrons are injected into the material, they may be trapped by the defects be annihilated there.[1-3] The positron annihilation characteristic depends on the type of defect at which the positron is trapped. This may give a way of differentiating between different types of defects.[4]

The positron lifetime spectroscopy is also a very useful tool with which to study the various diffusion-limited processes in metals and alloys (e.g., recovery and recrystallization, precipitation, thermal aging, etc). It also gives accurate information about the atomic density, the degree of disordering, and the mean grain size of lattice alloys.[2-5]

Lynn et al.[6] gave the first systematic data for the dependence of the positron lifetime on the mean grain size for polycrystalline copper. The data were interpreted in terms of e^+ trapping in grain boundaries by Leighly.[7] More detailed measurements of e^+ annihilation on fine-grained Zn-Al alloys as a function of the mean grain size[8,9] were analyzed by means of the rate equation of the standard two-state trapping model.[10,11] Seeger[12] gave an exact treatment of a model for both the e^+ diffusion inside the grains and the trapping-detrapping reactions at the grain boundaries. Recently, the recovery stages in aluminum and Al-1 wt.% Mn were studied using PAS.[13]

M.S. Abd El Keriem, Physics Department, Faculty of Science, King Khalid University, Abha-9004, Saudi Arabia. Contact e-mail: magdysalah20011@yahoo.com.

The purpose of this investigation was to study the microstructure variations of commercial copper (99.5%) during isochronal annealing in the temperature interval from room temperature (RT) up to 1223 K by using positron annihilation lifetime (PAL) as a nondestructive technique, microhardness measurements, and optical and scanning electron microscopy (SEM) observations.

2. Experimental Procedure

Commercial (99.5%) copper samples were cold-rolled (~80% reduction of thickness). A series of the above samples were annealed at temperatures from 423 K up to 1223 K in steps of 50 K for 1 h each. The cooling of the samples to RT was performed slowly in the oven to avoid quenching effects. The positron lifetime measurements were carried out at RT for the as-received samples and the annealed sample. Positron lifetime spectra were recorded using a plastic fast-fast lifetime spectrometer with a time resolution of 200 picoseconds (ps) for ^{60}Co. Spectra were recorded at a rate of 550 counts per second (cps) at 20 microcuries (μCi). Usually, ^{22}Na, in the form of NaCl deposited on Kapton foil, is used as the positron source. The positron source is sandwiched between two similar samples that are each about 1 mm thick. The two samples included in the positron source are put between the two detectors of the positron lifetime set-up. With the emission of a positron, the source emits a γ-quantum, a photon that is recorded on one detector. A γ- ray (1.024 MeV) is emitted if an electron and its antiparticle, the positron, meet and annihilate, and one of the annihilation photons is recorded on another detector. The time difference between the two signals from the two detectors is, consequently, the positron lifetime. With a suitable electronics set-up, this time difference is transformed into what is called the lifetime spectra. A positron can annihilate through different processes, each of which gives rise to a certain mean lifetime (τ). The lifetime spectrum shows a curve,

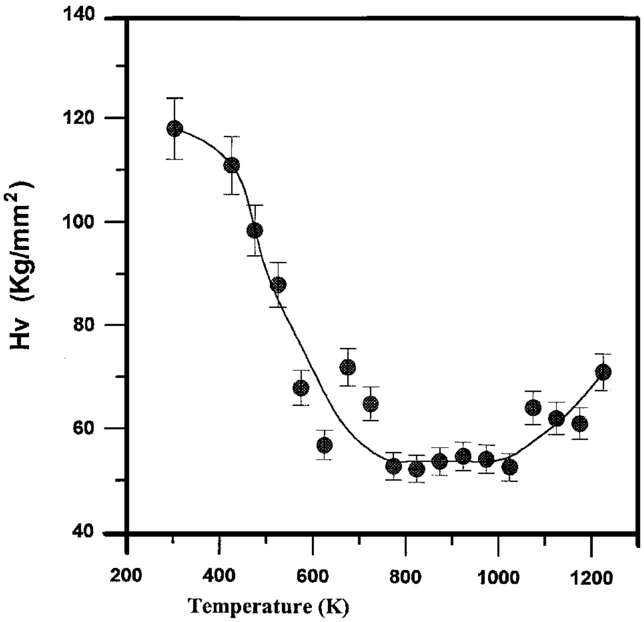

Fig. 1 The effect of annealing temperature on the microhardness measurements of cold-rolled commercially pure copper (99.5%)

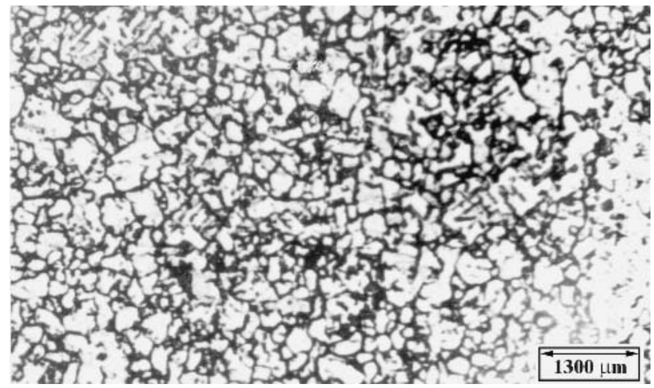

Fig. 2 Microstructure of pure copper (99.5%) annealed at 573 K

which is a decaying exponential, containing many components due to several annihilation processes. The area under one component divided by the total area of the spectrum is called the relative intensity (I) of the component. The lifetime spectra were analyzed into three components using the computer program PATFIT-88 (Risø National Laboratory, Roskild, Denmark). The spectra were corrected for a 3% contribution from the source, and the variance of fit ranges from 0.94-1.25. The positron lifetime technique is discussed in a number of articles (e.g., Ref. 14 and 15).

The Vickers microhardness measurements for the test samples were determined after polishing by using a Tukkon Vickers hardness tester with an applied load of 50 g. More than 10 readings were taken for each sample, and the standard deviations were calculated.

For optical microscopy and SEM observations, the samples were etched after polishing in a solution consisting of 10 g of ferric chloride, 10 ml of hydrochloric acid, and 100 ml of water.

3. Results and Discussion

The effect of isochronal annealing on the Vickers microhardness measurements of the prepared samples is shown in Fig. 1. A rapid decrease in the microhardness is seen as the annealing temperature is increased up to ~723 K. A plateau at the intermediate temperature range (773-1023 K) follows this. An increase of microhardness is observed at higher temperatures above 1023 K. The rapid decrease of microhardness can be attributed to a recovery and partial recrystallization. The lower values of microhardness and stability are due to primary recrystallization and grain growth. The increase of hardness observed at higher temperatures may be due to certain rearrangements of grain boundaries after the primary recrystallization and grain growth occurs.

Optical and SEM were employed to investigate the microstructures. Figure 2 indicates that the sample annealed at 573 K/h was not completely recrystallized. Grain growth and the formation of subgrains are shown in Fig. 3 for the sample annealed at 723 K/h. An increase in the subgrains is observed when the annealing temperature is raised. This can be seen in Fig. 4 for the sample annealed at 773 K. However, such subgrain growth deteriorates at higher temperatures. This is clearly depicted in Fig. 5 for the sample annealed at 873 K. The formation of islands can be observed at much higher temperatures. Figure 6 shows such a trend at annealing temperatures of 1073 K and 1173 K, for example. A similar behavior was reported recently[16] on much purer copper samples.

The relationship between isochronal annealing and the positron annihilation short lifetime (τ_1) and the average lifetime ($\bar{\tau}$) is shown in Fig. 7. It appears that the behavior of τ_1 with temperature is nearly similar to the behavior of microhardness, with the exception that the variations in τ_1 are sharper and clearer. Unlike hardness determinations, the lifetime technique is completely nondestructive and, therefore, can be used in many applications for which hardness measurements are unacceptable. The strong dependence of the bulk lifetime on temperature was observed earlier for copper.[17]

The change of $\bar{\tau}$ with temperature is completely different from that of τ_1. An initial increase of $\bar{\tau}$ is observed up to 573 K. This initial increase, indeed, is followed by a sharp decrease to show a minimum at about 823 K. Finally, a plateau is observed at a much higher temperature, which indicates stability in the trapping signal. The saturated trapping signal was observed before for βCu-Zn in thermal equilibrium experiments at a temperature of about 620 K, indicating a high vacancy concentration (~10^{-4}) at this temperature.[18]

Figure 8 shows the dependence of the intensity I_2% of positrons on the annealing temperature. A comparison between Fig. 8 and $\bar{\tau}$ in Fig. 7 distinguishes four different regions. The first region is characterized by an increase of $\bar{\tau}$ that is associated with an increase of I_2 from ~23.5--~38.5% in the temperature range from RT to 573 K. This region could be attributed to a partial recrystallization of the system. The second region, which extends between 573 K and 823 K shows a decrease of $\bar{\tau}$ accompanied by a decrease of I_2 from 38.5--~26%, which could be attributed to a complete recrystallization. The optical

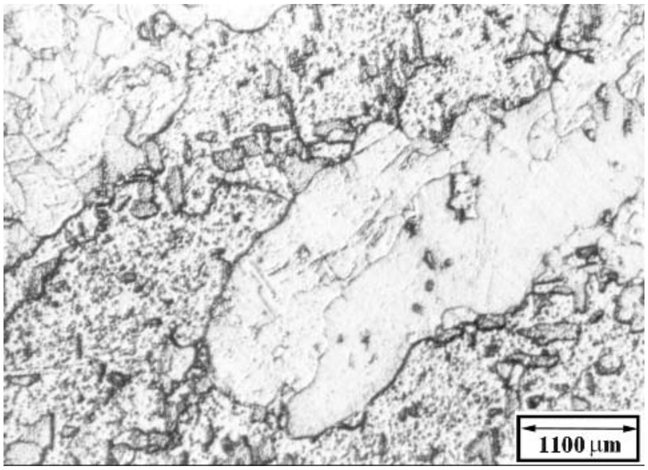

Fig. 3 Microstructure of pure copper (99.5%) annealed at 723 K

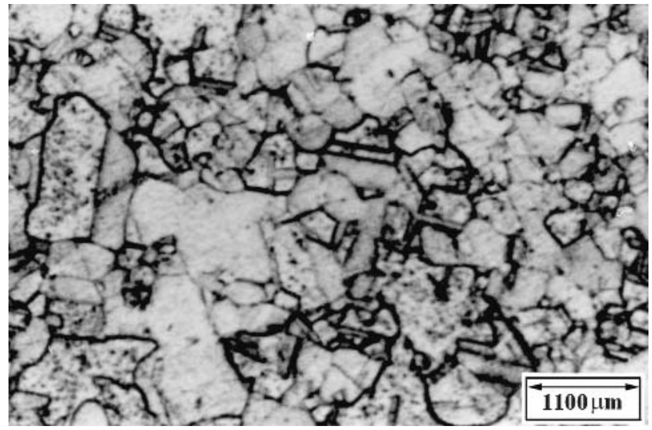

Fig. 5 Microstructure of pure copper (99.5%) annealed at 873 K

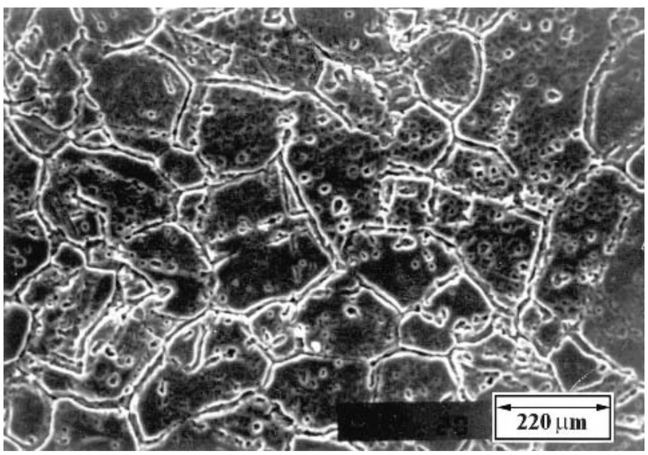

Fig. 4 Microstructure of pure copper (99.5%) annealed at 773 K

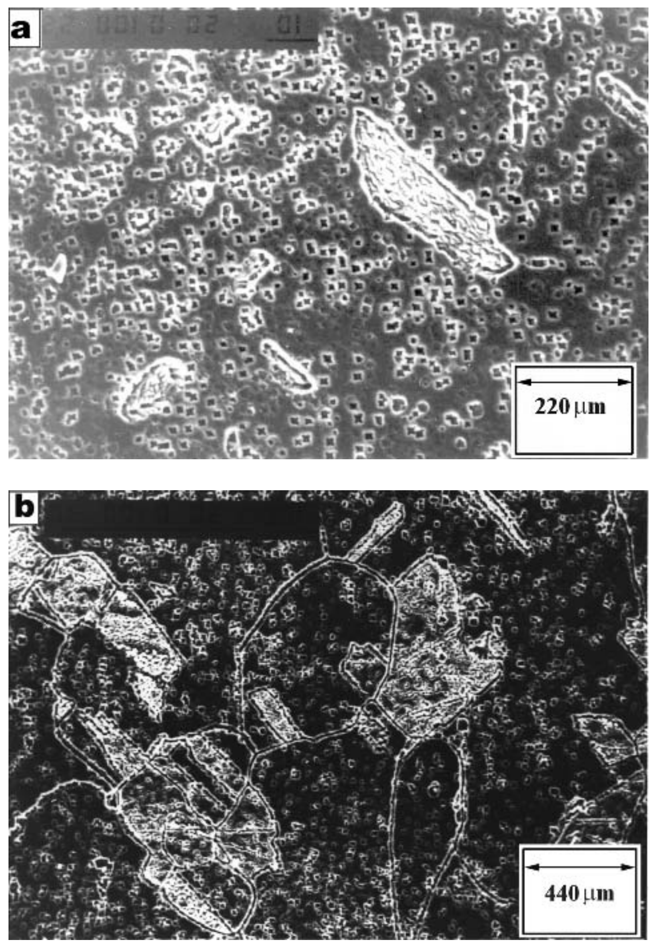

Fig. 6 Microstructure of pure copper (99.5%) annealed at (**a**) 1073 K and (**b**) 1173 K

and SEM observations confirmed these results (Fig. 2-4). A continuous decrease in τ_1 in these two regions is observed up to 773 K. At the end of the second region, up to the annealing temperature of 923 K, the value of τ_1 approaches the bulk value (122 ± 2 ps), indicating a release of the defects and primary recrystallization. This value is nearly the same as that obtained by Dave and Leblank[19] (122 ± 1.5 ps) and is higher than that obtained by Hehnkamp et al.[20] The difference in the measurement of the lifetimes may be referred to as the variation of the purity and history of the material. The lifetime of $\bar{\tau}$ never falls below the bulk lifetime value, which is evidence for the existence of positron-trapping centers during any heat treatments of the first two regions. For copper, the positron lifetimes in defects such as monovacancies and dislocation loops were found to be in the range of 180 ps.[21]

The increase of both $\bar{\tau}$ and I_2% in region three (823-973 K) is mainly due to the rearrangement of grains during the grain growth. Since there are no defects after complete recrystallization, the rule of grain boundaries thus will be the only parameter responsible for the increase of both the lifetime and intensity of positron annihilation.

However, the plateau of $\bar{\tau}$ in region four (973-1223 K) may be referred to as the stability of the microstructure after complete recrystallization and grain growth. The decrease of the intensity I_2 (Fig. 8) reflects that some vacancies were lost into

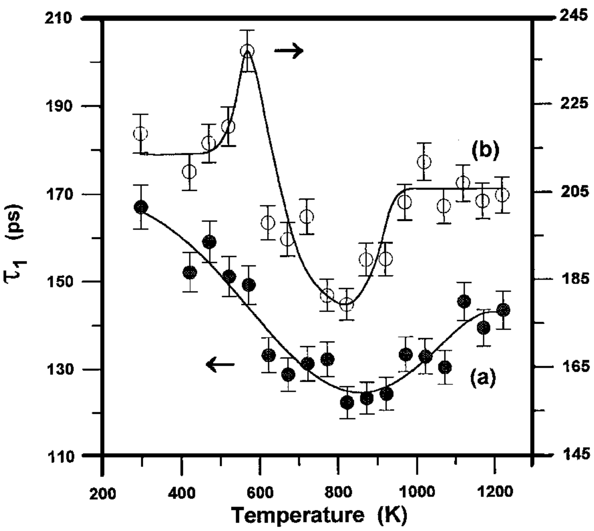

Fig. 7 The effect of annealing temperature on the RTs for pure copper (99.5%): (**a**) the short lifetime τ_1; and (**b**) the average lifetime

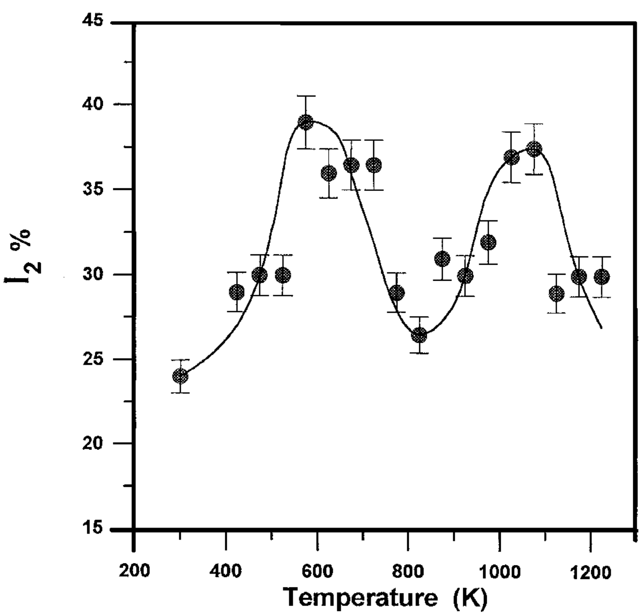

Fig. 8 The effect of annealing temperature on the intensity I_2 value

sinks or probably were recombined with self-interstitials during migration. In addition, the trapping rate/vacancy ratio decreases with increasing cluster size, which also contributes to the reduction of I_2.[21]

The positron-trapping rate (K) is defined as $K = \mu\, c_t = I_2\, (1/\tau_1 - 1/\tau_2)$, where c_t is the trap concentration and μ is the specific value of K. The variation of K with temperature (Fig. 9) shows a minimum occurring between two different maxima. This minimum corresponds to a complete recrystallization. This complete recrystallization is followed by grain growth and the formation of probable grain boundaries. This confirms our explanation about the classification of the four different regions.

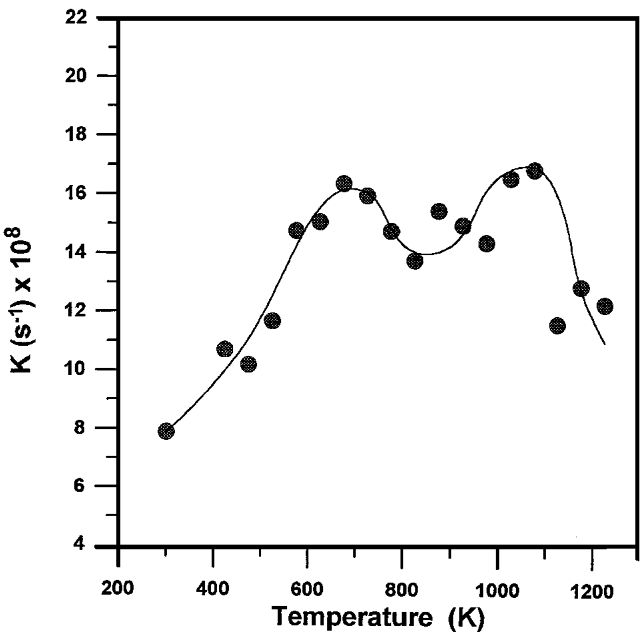

Fig. 9 The effect of annealing temperature on K

4. Conclusions

The results of the above study indicate that the variation of the short lifetime (τ_1) with temperature for pure copper (99.5%) is similar to that of microstructures but is more distinct and clearer. The relationships among isochronal annealing, the positron lifetimes, and the intensity I_2% reveals four regions, which are recovery (RT-573 K), partial recrystallization and primary recrystallization (573-823 K), grain growth (823-973 K), and stability of the microstructure after complete recrystallization and grain growth (973-1223 K). The variation of K with temperature, optical and SEM observations, and microhardness measurements confirms the presence of these regions.

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