Reocrystallization Kinetics in Copper: Comparison between Techniques

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Six different experimental techniques (electron backscattering (Kikuchi) patterns, calorimetry, micro- and macrohardness, orientation contrast scanning electron microscopy, and neutron diffraction) have been used to determine the volume fraction of recrystallized material \( X(t) \) in a series of partly recrystallized copper samples. Before recrystallization, the copper samples were cold-rolled to 92 pct reduction in thickness. The results obtained with the different experimental techniques are compared, and the applicability and accuracy of the six techniques are considered. While five of the techniques reveal the same kinetics, although with different degrees of accuracy, the texture measurements by neutron diffraction seem to indicate a faster recrystallization process. Finally, the recrystallization kinetics and stored energy aspects of the copper material are discussed with respect to the Johnson–Mehl–Avrami–Kolmogorov model.

I. INTRODUCTION

In studying recrystallization kinetics, it is necessary to determine the volume fraction of recrystallized material \( X \) in partially recrystallized samples. The most direct way to determine \( X \) is by microscopy using a series of partially recrystallized samples, and often optical microscopy is used. However, for example, in heavily deformed copper, it is difficult to distinguish deformed material from recrystallized material by optical microscopy.

Other, more indirect methods have also been used to determine the recrystallized fraction. Hardness indentations, calorimetry measurements, neutron diffraction texture data, electrical resistivity, and measurements of magnetic parameters have been employed for this purpose in the past.

The aim of the present article is to compare the results obtained by various techniques. As a basis for this comparison, data obtained by the electron backscattering pattern (EBSP) technique in the scanning electron microscope (SEM) are used. This method has the advantage of being a direct measurement of the traces of grains on a prepared external surface combined with orientation information, and not an indirect measurement like the other previously mentioned techniques. By EBSP, it is possible to determine almost objectively whether an area is recrystallized.

The material studied was copper cold-rolled to 92 pct reduction and annealed at 121 °C. The recrystallization in this material is very inhomogeneous, and it is therefore a good test material for the various techniques. The results of calorimetry, hardness (micro and macro), orientation contrast determination in the SEM, and neutron diffraction are related to the EBSP results and the agreements and disagreement are discussed.

II. EXPERIMENTAL

The material was copper of 99.96 pct purity, the main impurities being Pb, As, Ni, Ag, Co, and Fe. Initially the material was in the form of cold-drawn bars of square cross section (12 mm). These were cut into 20-cm-long pieces, each of which was sealed in a glass tube under 5N Argon gas and annealed at 450 °C for 1.5 hours. The average grain size after this treatment was 35 μm.

Subsequently, the pieces were deformed by rolling in steps of 1-mm reduction (12 mm, 7 mm), 0.5-mm reduction (7 mm, 2 mm), and 0.2-mm reduction (2 mm, 1 mm). This is equivalent to a total deformation of 92 pct. The choice of the reduction steps ensured that the condition for homogeneous deformation during rolling was fulfilled apart from the first reduction step. In the next step, samples of 6-mm and 10-mm diameters were punched out of the rolled sheet and stored at 6 °C. Before any experiments, all disks were etched in 50 pct nitric acid to remove surface contamination as well as any deformation fringe due to the punching.

Initially, some 6-mm specimens were recrystallized in a DSC-7 calorimeter made by Perkin–Elmer in order to determine the progress of the recrystallization with time under isothermal (121 °C) conditions. Thus, the times necessary to achieve certain recrystallized fractions could be specified within certain limits. These limits are due to the scatter among the nominal identical specimens under the same annealing conditions. This can be seen in Figure 1, where the heat release of three specimens at 121 °C (a) and the fraction recrystallized (b) derived from these data (see subsequent discussion) are plotted.

In order to derive the curves in Figure 1(b), the recovery contribution has to be subtracted. This concerns the initial part (up to 20 minutes), where an exponential-like decrease of the signal is superimposed on the beginning of the recrystallization peak (for further discussion, see References 9 and 10). Fitting this part by an exponential and subtracting it yields the "pure" recrystallization peak. This is then integrated and normalized to give curves like the ones shown in Figure 1(b).

Based on these curves (Figure 1(b)), it was decided to
heat treat samples for 1350, 1740, 2190, 2670, 3300, and 7800 seconds at 121 °C to obtain recrystallization fractions of approximately 10, 20, 40, 60, 80, and 100 pct, respectively. Each sample consists of a set of eight disks (10 mm in diameter) which were annealed simultaneously in a small furnace. Following that, the eight disks were marked by two small grooves at opposite points of the circumference indicating the transverse direction. The eight disks of each sample were then stacked together, positioned by a thin guidewire along the grooves. These stacks and an additional one consisting of unannealed disks were used for the neutron diffraction experiments described later. Subsequently, the stacks were split and the individual disks investigated employing the other experimental techniques described subsequently. For the EBSP measurements, additional specimens were prepared which were annealed at 121 °C for 750, 1050, and 1200 seconds as well as 4800 and 5100 seconds, corresponding to less than 10 pct and about 90 pct recrystallized fraction.

A. Electron Backscattering Pattern Measurements

A JEOL* 840 SEM equipped with a phosphor screen and a very low light TV camera mounted in the side port was used for the EBSP measurements. The principle of the EBSP technique is similar to that of the backscattered Kikuchi pattern technique. In the SEM, the incident electron beam is focused as a stationary probe on the specimen surface. When the sample is tilted (typically ~20 deg to the incoming beam), a large fraction of the beam is backscattered with sufficient contrast to form a clear Kikuchi pattern on the phosphor screen. Typical examples of EBSPs are shown in Figure 2. Such patterns contain full information about the crystallographic orientation of the selected ~1-μm-diameter sample area.

An EBSP from a recrystallized region (>1 μm) is sharp with a high signal-to-noise ratio (Figure 2(a)), whereas EBSPs from deformed regions are blurred or nonexistent with low signal-to-noise ratios (Figure 2(b)). Furthermore, when traversing a deformed region, the pattern is slightly meandering (this is due to local misorientations in the deformed microstructure), whereas a stable (nonmoving) pattern is observed when a recrystallized grain is crossed. Finally, recrystallized nuclei are at least partly surrounded by a high angle grain boundary, which is easily detectable. It is therefore fairly straightforward to distinguish deformed from recrystallized material.

For the present investigation, one to two disks were prepared for EBSP measurements by electropolishing in Struers A2. Each of them was inspected in the rolling plane section. The volume fraction recrystallized was determined by performing scans along the rolling and transverse directions. The individual intercept-free grain lengths, \( \lambda \), were recorded for each recrystallized grain encountered by noting the vernier positions of the stage. The value of \( X(t) \) was then calculated as \( X(t) = \Sigma \lambda / L \), where \( L \) is the total line length traversed. Due to the inhomogeneous recrystallization in the present material, quite long scans were needed; \( L \) was in the range of 0.5 to 5 cm. As mentioned previously, the scans were made along the rolling and transverse directions. When the two types of scans were compared, no significant differences could be observed, and the \( X(t) \) values to be reported in the following were calculated as the weighted average from the two types of scans. Ideally, in an anisotropic microstructure with, for example, flat grains, the scans should be in random directions to get a correct measure for \( X \). However, as the recrystallized grains are fairly equiaxed, no significant errors are expected due to this.

A measure of the sampling errors expressed as standard deviations, \( \sigma_n \), has been derived and is discussed in another article. It is based on an equation deduced by Hilliard and Cahn:

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\left( \frac{\sigma_n}{X} \right)^2 = \frac{1}{M} \left[ \left( \frac{\sigma_{\lambda r}}{\langle \lambda r \rangle} \right)^2 + \left( \frac{\sigma_{\lambda d}}{\langle \lambda d \rangle} \right)^2 \right]
\]

where \( M \) is the number of recrystallized grains encountered in a transverse of length \( L \); \( \langle \lambda r \rangle \) and \( \langle \lambda d \rangle \) are the mean...