Dynamic Recrystallization under Warm Deformation of Polycrystalline Copper

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Structural changes taking place under warm deformation of pure copper were studied in compression at temperatures ranging from 473 to 673 K (0.35 to $0.50T_m$) under strain rates of 10^{-3} – 10^{-1} s^{-1} . Dynamic recrystallization (DRX) takes place fully or partly at temperatures higher than 523 K, while no fine grains are evolved in the pan-caked original grains with serrated boundaries even after high strains at 473 K. The new grains are evolved by bulging mechanism associated with local migration of the original grain boundaries and the evolution of twins and dislocation boundaries. The relationship of peak flow stress to the new grain size evolved under warm deformation can be expressed by a power law function with a grain size exponent of about -0.35 , which is different from that for DRX taking place under hot deformation (*i.e.* -0.75). The correlation between the mechanisms of plastic deformation and the structural evolution, such as dislocation densities, cell sizes, and DRX grain sizes, and the mechanisms of low and high temperature DRX are discussed in combination with the analysis of deformation behaviour at moderate temperatures.

KEY WORDS: warm deformation; pure copper; dislocation substructure; dynamic recovery; dynamic recrystallization; mechanism of new grain formation.

1. Introduction

Grain refinement of metallic materials through thermo-mechanical treatments is of great practical importance because of the control and improvement of the mechanical properties of the products. Dynamic recrystallization (DRX) as well as static one (SRX) after DRX is the very important and interesting phenomena for grain refinement techniques, although their industrial application has not been succeeded yet.^{1–4)} The characteristics of DRX and also SRX after DRX have been fairly clarified and the several phenomenological mechanisms have been discussed.^{1–4)} There are, however, some problems remaining unsolved in the physical mechanisms of dynamic nucleation and of necklace DRX.^{5,6)} The critical conditions for the occurrence of DRX (*e.g.* materials, temperature and strain rate *etc.*) also have not been clarified, although they should give very important knowledge to the control of deformed microstructure.

The aim of the present work is to study the structural changes connected with a new grain formation during warm deformation of a DRX metal in which no phase change is involved. For this purpose, a polycrystalline pure copper was selected as the material and deformed at temperatures below half of the melting point ($0.5T_m$) under various strain rates. The effects of strain, strain rate and temperatures below $0.5T_m$ on the development of new grains are studied. Through the correlation

between flow behaviours and microstructure development the mechanisms operating are discussed in detail.

2. Experimental Procedure

A polycrystalline copper of 99.99 purity with an initial grain size of $60 \mu\text{m}$ was used as the samples for the present tests. The samples of 8 mm in diameter and 12 mm in height were compressed in vacuum with a graphite lubricant (DAG154) at warm temperatures under various constant true strain rates from 10^{-3} to 10^{-1} s^{-1} . The test temperatures were changed mainly from 473 to 673 K (0.35 to $0.50T_m$) and 723 K partly was used. The testing samples could be removed from the vessel and quenched within one second after deformation was ceased.⁷⁾ They were cut along a plane parallel to the compression axis, and then electropolished and etched by conventional technique for optical microscopy of copper.⁸⁾ The dislocation substructures of deformed samples were examined in a JEM-2000FX transmission electron microscope (TEM) operating at 200 kV. Misorientations on grain boundaries were studied using Kikuchi-line technique.⁹⁾ The accuracy of the analysis was about 0.5° because of high dislocation densities evolved during warm deformation.

3. Experimental Results

3.1. Flow Behaviours

Typical true stress–true strain (σ – ϵ) curves for the

Fig. 1. Effect of strain rate on true stress-true strain curves for 4N Cu at 523 and 623 K.

Fig. 2. Relationships between peak stress and strain rate for 4N Cu at 523 and 623 K.

copper at 523 and 623 K are shown in Fig. 1. The curves at 623 K exhibit a typical DRX type flow with a single peak stress followed by work softening.¹⁻³⁾ A strain corresponding to the peak stress increases and the following work softening decreases with decreasing temperature and increasing strain rate. On the other hand, the stress-strain curves below 523 K are similar to those usually observed under dynamic recovery and exhibit a quite small strain rate dependence. The flow stresses approach to a saturation value at strains above 1.

Figure 2 shows the strain rate ($\dot{\epsilon}$) dependence of the peak flow stress (σ_p). The relationship between σ_p and $\dot{\epsilon}$ can be approximated by a linear line with the stress exponent of about 30 and 150 at 623 and 523 K, respectively. These exponents are quite large compared with that for hot deformation of copper, *i.e.* 5 to 6.^{1,5)} It is considered, therefore, that the plastic flow taking place at around 523 K can be in the region of athermal flow.

Fig. 3. Effect of temperature on microstructural development at high strains above 1.2 for 4N Cu deformed at 10^{-3} s^{-1} : (a) 473 K, (b) 523 K, (c) 573 K and (d) 623 K.

3.2. Deformed Microstructures at High Strains

Figure 3 shows temperature dependence of the microstructures for the samples deformed to high strains above 1.2 at a strain rate of 10^{-3} s^{-1} . It can be clearly seen in Fig. 3 that DRX partially takes place at 523 and 573 K, and almost fully at 623 K. Decreasing of deformation temperature leads to the reduction in both DRX grain size and also the volume fraction of new grains from 100% at 623 K to 10% at 523 K. It is noted in Fig. 3(a), however, that no visible fine grains are evolved even at high strains at 473 K, and the original grains are pancaked accompanied with serrated boundaries.

It is well known that the stable DRX grain size (D_s) evolved at high strains is dependent sensitively on temperature and strain rate and so is a unique function of flow stress.^{1-3,5)} Figure 4 shows the relationship between peak stress (σ_p) and D_s under warm deformation

Fig. 4. Relationship between peak flow stress and stable dynamic grain size for 4N Cu under warm and hot deformation. Steady state flow stress (σ_s) vs. D_s relationship for 4N Cu⁵⁾ also is included for comparison.

of copper and, for comparison, includes the result for hot deformation at temperatures above $0.5T_m$.⁵⁾ The average values of D_s were measured only in the regions of DRX grains evolved. It is clearly noted in Fig. 4 that the σ_p - D_s relationship can be represented approximately by two straight lines which hold in the two domains to be distinguished at the critical condition if $\sigma_p \approx 180$ MPa. In the region of $\sigma_p < 180$ MPa, the σ_p - D_s curve is parallel to the σ_s - D_s one for hot deformation⁵⁾ and is similar to that for many other DRX metals.⁵⁾ In contrast, it shifts clearly towards the range of smaller grain sizes in the higher stress region of $\sigma_p > 180$ MPa. The slope of the σ_p - D_s relation is -0.35 in the region of $\sigma_p > 180$ MPa (i.e. under warm deformation), which differs from that of -0.75 in the region of lower stresses (i.e. under hot deformation).

3.3. Dislocation Substructures Developed at around a Peak Strain

A typical substructure developed by warm deformation to around a peak strain is shown in Fig. 5. The sample was deformed to about 0.7 at 623 K with 10^{-3} s^{-1} , instantaneously followed by quenching in water. It can be seen in Fig. 5 that the substructures evolved in grain interiors are mainly composed of the blocks of elongated cells subdivided by dense dislocation walls (DDWs). These look like the substructure developed under cold deformation of fcc metals.^{10,11)} The DDWs are arranged in parallel families that make elongated cells mutually disoriented on 2 to 4°, as indicated in Fig. 5. The characteristics of such dislocation substructures were scarcely changed by temperature or strain rate within the present test conditions, although with decreasing temperature the cell size (d) decreases and the dislocation density inside cells (ρ) increases.

Figure 6 represents the relationships between peak flow stress (σ_p) and ρ or d in log-log scale. The average value of d was measured in the transverse direction to DDWs.

Fig. 5. Typical TEM micrograph of dislocation substructure arranged in dense dislocation walls in 4N Cu deformed to a strain of 0.7 at 623 K under a strain rate of 10^{-3} s^{-1} .

Fig. 6. Relationship between peak flow stress (σ_p) and dislocation density (ρ) or cell size (d) observed at around a peak strain of warm deformed 4N Cu.

It can be seen in Fig. 6 that both the ρ - σ_p and d - σ_p relationships can be approximated by power laws. With the exception of stress region of around 200 MPa, the stress exponents for the ρ - σ_p and d - σ_p curves are about 2 and -1 , respectively. These values are roughly equal to those for dynamically recovered materials, which contain relatively homogeneous dislocation substructures developed in pan-caked grains.^{1,12)}

On the other hand, the dislocation substructures developed under DRX are different in each grain and distributed heterogeneously throughout.^{13,14)} It is quite possible, therefore, that the breakdown of power law in d or ρ vs. σ_p curve results from the changes in dislocation substructures from relatively homogeneous to hetero-

Fig. 7. Microstructural evolution in 4N Cu during deformation to various strains at 623 K with 10^{-3} s^{-1} : (a) $\varepsilon=0.4$ and (b) $\varepsilon=0.7$.

geneous nature. It is also interesting to note that such breakdowns appear at $\sigma_p \approx 200 \text{ MPa}$ and, on the other hand, the σ_p - D_s relation in Fig. 4 clearly changes at $\sigma_p \approx 180 \text{ MPa}$.

3.4. DRX Nucleation

An example of the progress of DRX taking place at 623 K with 10^{-3} s^{-1} is represented in Fig. 7. At a strain of 0.4 which is just below the peak strain, the original grains become pan-caked accompanied with the boundaries serrated. It is clearly seen in Fig. 7 that the new grain formation is accompanied with local migration of grain boundaries, followed by the evolution of necklace microstructure along these boundaries. At higher strains of over 1.2, DRX new grains having irregular shape and a few twins are almost fully developed (Fig. 3(d)). Such a progress of DRX is almost the same as that occurring under hot deformation at temperatures of above $0.5T_m$.^{1,3-5)}

The TEM observations confirm that the DRX grains were mainly formed by bulging mechanism even at the present medium temperatures, as shown in Fig. 8. It is clearly seen in Fig. 8(a) that local boundary migration can create nuclei of new grains and they already contain a few lattice dislocations. The boundary migration can be accompanied by twinning which can become like a boundary separating the nucleus from the parent grain in the lower right hand. It can also be noted in Fig. 8(b) that another type of such boundaries is composed of the dislocation boundaries with middle angle misorientation (e.g. 9.1° to 13.5°) which is clearly higher than an average misorientation of DDWs evolved in grain interiors (see Fig. 5). Accompanied with growth of the new grains, dislocation substructures are redeveloped and rearranged behind the moving boundaries by further straining. Once growing DRX grains with a dislocation density gradient

Fig. 8. Nuclei of new grains evolved along grain boundary (GB) of 4N Cu. The dislocation cell boundaries with a low misorientation of 1 to 2° are not indicated here schematically for simplicity. (a) 623 K; note twin boundaries and some dislocations are evolved in a new grain. (b) 523 K; note dislocation boundaries with middle angle misorientation are formed in a new grain.

may impinge on, the dislocation can increase more rapidly near the boundaries due to strain compatibility, leading to the development of fairly uniform substructures.¹⁴⁾ In a full DRX structure, regions belonging to three types of DRX grains, i.e. DRX nuclei, growing DRX grains and ones containing uniform dislocation substructures, are distributed fairly uniformly throughout, leading to a steady state flow at high strains.¹⁴⁾

On the other hand, a typical substructure evolved under deformation at 473 K with 10^{-3} s^{-1} is shown in Fig. 9. At this testing condition, there is no visible fine grains evolved at a strain of 1.2 (see Fig. 3(a)). Further straining to $\varepsilon > 4$, however, fine grains were frequently evolved in grain interiors, although the details will be

Fig. 9. Typical TEM micrograph of substructure evolved along grain boundary (GB) of 4N Cu deformed to a strain of 1.2 at 473 K under a strain rate of 10^{-3} s^{-1} . Note high density dislocation boundaries developed along GB and their misorientations decreasing with distance from GB.

described elsewhere. The substructures including high density dislocations are characterised by DDWs developed in grain interiors. It can be clearly seen in Fig. 9 that high density dislocations in the substructure evolved near the original grain boundary are arranged in parallel along the boundary, leading to the evolution of long DDWs in both the grains. It can also be noted here that the misorientations of such dislocation subboundaries decrease with distance from the grain boundary. It is suggested from Fig. 9 that such dislocation boundaries with middle angle misorientation can combine with the serrated initial boundary, leading to the formation of chains of subgrains outlined by middle to high angle boundaries. These subgrains can be evolved by dynamic recovery taking place predominantly along the grain boundaries because of high density dislocations stored. It should be noted, however, that this dynamic process of initial grain boundary serration does not connect with bulging mechanism operating under hot deformation.

4. Discussion

4.1. Deformation at Medium Temperatures

In the temperature region tested from $0.35T_m$ to $0.50T_m$, strain rate dependence of the σ - ϵ curves as well as the flow stresses are small compared with that for hot deformation. Their strain rate sensitivities approach to almost zero at around 523 K (see Figs. 1 and 2). **Figure 10** shows the relationship between the peak flow stress (σ_p) and temperature (T) under several strain rates in $\log \sigma_p$ vs. $1/T$ scale. It can be seen in Fig. 10 that the temperature dependence of σ_p clearly changes at a medium range of temperature which depends on strain

Fig. 10. Effect of strain rate on peak flow stress (σ_p) vs. temperature (T) relationship for 4N Cu in $\log \sigma_p$ - $1/T$ scale.

rate. The σ_p shows a strong temperature and strain rate dependence at higher temperatures above $0.5T_m$, i.e. under hot deformation, where DRX also frequently takes place.

In contrast, temperature and strain rate dependencies of σ_p are relatively and negligibly small at lower temperatures, respectively. Such plastic flow is considered to be in the region of athermal flow. It is concluded, therefore, that the temperatures in the present study are in a transitional region from thermal to athermal or from high to low temperature deformation. The high values of stress exponent in Fig. 2 are clearly changed with temperature. This suggests that the deformation behaviour of copper in the transitional region cannot be described by a simple power law equation. According to Frost and Ashby,¹⁵⁾ such a deformation region can be called as that of power law breakdown. They discussed that diffusion mechanisms assisting any dislocation movement become more difficult in this region and the strain rate dependence of stress decreases with decreasing temperature. The structure evolution taking place under warm deformation are very complex and usually connected with both cold and hot deformation, which will be described below.

4.2. DRX under Hot Deformation

The microstructure evolution taking place during hot deformation of copper is characterised by the operation of DRX. Sakai and Miura⁵⁾ pointed out that dynamic nucleation frequently takes place by bulging of the serrated grain boundaries, where inhomogeneous local strain gradients developed by grain boundary sliding. They concluded under such structural conditions that bulging mechanism, which Bailey and Hirsh¹⁶⁾ originally proposed for static nucleation, can operate much easier and quicker than that operating in cold-worked matrices, and so the serrated grain boundaries should be most preferable nucleation sites for DRX. These discussions

can be applied to DRX occurring even in the transitional region below $0.5T_m$. This is supported by the results of Figs. 3, 4, 7 and 8. It is concluded, therefore, that dynamic nucleation can be mainly controlled by bulging mechanism operating under DRX in the all range of medium to high temperatures.

It is well known that the stable dynamic grain size (D_s) evolved at high strains is dependent sensitively on temperature and strain rate, and so is a function of high temperature flow stress.^{1-3,5)} The data for single phase DRX materials obey the following relation,

$$\sigma = KD^{-N}, \dots\dots\dots (1)$$

where K is a constant and the exponent N lies in the range 0.7 to 0.8.^{1-3,5)} As shown in Fig. 4, the relationship between σ_p and D_s for copper can be approximately represented by the same equation with $N=0.75$ in the region of $\sigma_p < 180$ MPa (*i.e.* under hot deformation). The σ_p - D_s relation in the region of $\sigma_p > 180$ MPa, however, can be approximated by an another power law with $N=0.35$. Below the critical point where the two straight lines intersect, DRX microstructures are evolved fully at high strains. On the other hand, the volume fraction of new grains (as well as the grain size) decreases with decreasing temperature and goes to almost zero below 523 K.

4.3. Microstructure Evolution at Medium Temperatures

At low to moderate temperatures the substructures evolved at strains before a peak are mainly characterised by the formation of dislocation subboundaries much like to the DDWs subdividing the grain interiors into cell blocks. Such kind of substructure evolution generally takes place during low temperature deformation.^{10,11,17)} In this case, as expected from a simple Taylor relation, the free dislocation density (ρ) and cell size (d) are related to the applied stress as

$$\sigma \propto \rho^{1/2}, \dots\dots\dots (2)$$

and

$$\sigma \propto d^{-1}, \dots\dots\dots (3)$$

respectively. Such relationships also have been studied in creep and hot deformation.^{1,12,18)} It can be clearly seen in Fig. 6 that the ρ - σ_p and d - σ_p relationships can be approximated by these Eqs. (2) and (3), if the region of $\sigma_p \approx 200$ MPa is excepted. This deviation taking place around 200 MPa can be closely caused by changing deformation behaviour from thermal to athermal flow, and also by changing dislocations substructures from inhomogeneous (under DRX) to homogeneous distribution (under dynamic recovery).

It is interesting to note in Fig. 4 that the DRX grains evolved at $\sigma_p > 180$ MPa are much finer than those extrapolated from the data for hot deformation (*i.e.* $\sigma_p < 180$ MPa). This may suggest that nuclei of new grains can be evolved by another mechanism in addition to grain boundary bulging and also their growth rate can become lower in the region of $\sigma_p > 180$ MPa. As shown in Fig. 3(a), local grain boundary migration for a short distance takes place even at lowest temperature

of 473 K, leading to the evolution of serrated grain boundaries. It should be noted in the present study, however, that bulging mechanism hardly takes place in serrated boundaries at 473 K (Fig. 9). By the way, various kinds of dislocation boundaries including DDWs are developed under severe deformation at low temperatures. Their boundary misorientation can increase with deformation, leading to the evolution of high angle boundaries.^{10,11)} Near the latter new grains can be evolved assisted by dynamic recovery.¹⁹⁾ It is quite possible, therefore, that the nucleation sites can be developed in grain interiors and increase with severe deformation. Then the new grains dynamically evolved at lower temperatures may become much finer than those extrapolated from the data for hot deformation. These important events should be studied in more detail in future.

5. Conclusions

Microstructure evolution taking place during warm deformation of a pure copper were studied at temperatures from 473 to 673 K (0.35 to $0.50T_m$) under various strain rates and the main results are summarised as follows.

(1) Temperatures from 0.35 to $0.50T_m$ for copper are in the transient region changing from low to high temperature deformation. Namely, the deformed microstructures are changed from pan-caked original grains to new fine grains accompanied with the transition from athermal to thermal flow behaviours.

(2) The new grains evolved at high temperatures of the transient region are nucleated by bulging of serrated original grain boundaries and grow to a saturation size at high strains. The relationship of peak flow stress to the dynamic grain size can be expressed by a power law function with a grain size exponent of about -0.75 . This dynamic recrystallization (DRX) is almost the same as the results for hot deformation of copper.

(3) The substructures developed at low temperatures of the transient region are characterised by the formation of dense dislocation walls at low to medium strains, followed by the incomplete evolution of finer DRX grains at high strains. The experimental data also obey a power law relation of the peak flow stress to DRX grain size with an exponent of about -0.35 .

(4) Much finer DRX grains evolved at lower temperatures suggest that nucleation sites for new grains may be not only serrated grain boundaries, but also grain interiors.

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