Effects of stacking fault energy on the deformation mechanisms and mechanical properties of Cu and Cu alloys processed by rolling at different temperatures

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Received 30 April 2014; accepted 14 November 2014

The effects of stacking fault energy (SFE) on the deformation mechanisms and mechanical properties of Cu, Cu-2.5 at.% Al-2.5 at.% Zn, and Cu-12.1 at.% Al-4.1 at.% Zn alloys processed by different rolling temperatures have been systematically investigated. Tensile tests at room temperature indicate that, as SFE decreased, strength and ductility increased, and at constant SFE the samples deformed via rolling at liquid nitrogen temperature (77 K) exhibit higher strength and better ductility than those deformed via rolling at room temperature (293 K). With lowering SFE, the crystallite size decreases while the microstrain, dislocations and twin densities increase. With decreasing temperature, there is a transition of deformation mechanism from that dominate by dislocation activities to that dominate by deformation twinning. The results indicate that decreasing SFE is an optimum method for improving the ductility of Cu and Cu alloys without loss of strength, and that temperature plays a key role in the rolling process.

Keywords: Cu-Al-Zn alloys, Deformation mechanisms, Mechanical properties, Stacking fault energy, Temperature

In recent research, severe plastic deformation (SPD), has been regarded as one of the most effective approaches for fabricating nanocrystalline (NS) and ultrafine-grained (UFG) metallic materials, which exhibit increased strength compared to their coarse-grained (CG) counterparts. The microstructures and mechanical properties of NS/UFG metallic materials produced using several SPD processing techniques, such as high pressure torsion (HPT), accumulative roll bonding (ARB), equal channel angular pressing (ECAP), dynamic plastic deformation (DPD) and severe cold rolling, have been extensively studied over the past few decades. Rolling is the fundamental SPD process and is more convenient than high pressure torsion (HPT) or equal channel angle pressing (ECAP) for different processing temperatures.

Furthermore, alloying is a conventional strengthening mechanism that can improve in ductility. SFE, as an important intrinsic material parameter, has an intimate relationship with alloying that can significantly affect the microstructure and mechanical properties. Lowering SFE inhibits dislocation activities and increases deformation twinning, as dynamic recovery can be suppressed by dislocation cross-slip and climb which are thermally-activated processes, leading to higher defect density and finer grain size. Strength and uniform elongation may be simultaneously improved by reducing SFE.

Our previous research demonstrated that with a decrease in SFE in Cu-xZn (x = 0, 10, 20, and 30 wt%) alloys, a significant increase in the tensile strength was achieved without sacrificing ductility using a stress-relaxation approach. In another piece of research on a series of Cu-xAl-yZn (x = 2.5, 4.5, 12.1; y = 2.5, 22.9, 4.1 at%) alloys with various SFE values, a theoretical model that can be used to predict the minimum grain size of the Cu alloys was verified using experimental data, and it was revealed that decreasing SFE led to a decrease in crystallite size and an increase in microstrain, dislocations and twin densities in the Cu alloys. Therefore, lowering SFE can provide an effective method for achieving excellent mechanical properties including high strength and good ductility in Cu alloys.

Deformation temperature is known to be a crucial factor in different processing conditions. In previous studies, the effect of temperature on deformation behavior and deformation mechanisms in nanotwinned Cu (nt-Cu) samples has been studied. However, for Cu-Al-Zn ternary alloys, the effect of SFE on the deformation mechanisms and mechanical properties of samples processed by rolling at different
temperatures has seldom been investigated. Therefore, the purpose of the current study was to explore the influence of SFE on the deformation mechanisms and mechanical properties of bulk UFG Cu and Cu-Al-Zn alloys produced by rolling at liquid nitrogen temperature (LNT) and room temperature (RT). In addition, the contribution of solid solution strengthening to the total yield strength increment based on the yield strength of pure Cu has been discussed.

Experimental Procedure

Sheets of commercial copper (99.9% purity) with a thickness of 7.9 mm were used in the experiments. Cu-2.5 at.% Al-2.5 at.% Zn and Cu-12.1 at.% Al-4.1 at.% Zn were produced by induction vacuum melting. The SFE γ values of pure Cu, Cu-2.5 at.% Al-2.5 at.% Zn, and Cu-12.1 at.% Al-4.1 at.% Zn were 78, 40, and 7 mJ/m², respectively. These as-cast samples were made into plates with the same thickness of 7 mm via hot-rolling. Before cold-rolling, the plates were homogenized at 600°C for 4 h in an argon atmosphere, which diminished the effect of mechanical processing, in order to obtain homogeneous coarse-grained (CG) microstructures. The homogenized samples were processed by liquid nitrogen temperature rolling (LNR) and room temperature rolling (RR) in multiple passes. For LNR, the samples were immersed in liquid nitrogen for about 5 min before each rolling pass to cool the samples completely. These plates were cold rolled from the initial thickness of 7 mm to approximately 0.4 mm.

For the tensile testing, the RR and LNR samples were cut into dog-bone shaped specimens with a gauge length of 15 mm and a width of 5 mm. The specimens were cut along the direction of rolling. Uniaxial tensile tests were carried out at room temperature using a Shimazu Universal Tester operating at a strain rate of 1.0×10⁻⁴ s⁻¹.

X-ray diffraction (XRD) measurements of the rolled Cu and Cu-Al-Zn alloy samples were performed on an X-ray diffractometer equipped with a Cu target operating at 1.2 kW. The pure Cu sheets (99.95% purity) that were annealed at 700°C in a vacuum were used as an XRD peak-broadening reference for the grain size, microstrain, dislocation density, and twin density calculations. The divergence, anti-scattering, and receiving slits were set at 0.5°, 0.5°, and 0.3 mm, respectively. A series of 0–20 scans using Cu-Kα radiation were performed in order to record the XRD patterns at room temperature.

A JEOL 2010 transmission electron microscope (TEM) was employed to characterize the microstructures and operated at 200 kV. The cross-sectional thin foils for TEM observation were first mechanically ground to about 50 μm thick and then thinned by a double-jet using electro-polishing at -30°C with an electrolyte of 33% nitric acid and 67% methanol. The grain size and twins could be observed accordingly.

Results and Discussion

Figure 1 shows the tensile mechanical behaviors of the pure Cu, Cu-2.5 at.% Al-2.5 at.% Zn, and Cu-12.1 at.% Al-4.1 at.% Zn samples processed via the LNR route and the RR route. As shown in the figure, the ultimate strength and the yield strength of Cu and Cu-Al-Zn alloys deformed via two different deformation temperatures were both enhanced with decreasing SFE, regardless of the processing temperatures applied. In addition, the total elongation and the uniform elongation increase gradually with decreasing SFE. It is evident that the LNR samples of the Cu alloys exhibited higher strength and better ductility than did the RR samples with decreasing SFE. Furthermore, the lower the SFE is, the better the mechanical properties of the LNR samples. Therefore, the Cu-12.1 at.% Al-4.1 at.% Zn alloy exhibited the highest strength and the best ductility.

Figures 2(a) and (b) show the normalized work hardening rate (θ) against the true strain (ε), the samples of which were deformed via LNR and RR routes, respectively. The true stress and true strain were calculated from the engineering stress-strain...
curves in Fig. 1. It can be clearly seen that the work hardening rate $\theta$ of both LNR samples and RR samples increased with a reduction in SFE. The Cu-12.1 at.% Al-4.1 at.% Zn alloys had the highest work hardening rate.

Using XRD line broadening, the average grain size can be calculated via the Scherrer-Wilson method due to the existence of the $\{111\}$ and $\{110\}$ textures; furthermore, the microstrains ($<\varepsilon^2_{111}>^{1/2}$ and $<\varepsilon^2_{200}>^{1/2}$) were obtained using the pairs of (111)-(222) and (200)-(400) reflections. According to the grain size ($d$) and the microstrain ($<\varepsilon^2>^{1/2}$), the dislocation density ($\rho$) can be expressed using Eq. (1)

$$\rho = \frac{2\sqrt{3}<\varepsilon^2>^{1/2}}{d_{XRD}b} \quad \cdots (1)$$

where $b$ is the absolute value of the Burgers vector for FCC Cu alloys, $b = (\sqrt{2}/2)a$, and $a$ is the lattice parameter determined from the peak positions. The twin density ($\beta$), which is defined as the probability of finding a twin boundary between two neighboring $\{111\}$ planes, was calculated by the following equation:

$$\beta = \frac{\Delta C \cdot G \cdot (2\theta)_{111} - \Delta C \cdot G \cdot (2\theta)_{200}}{11\tan\theta_{111} + 14.6\tan\theta_{200}} \quad \cdots (2)$$

where $\Delta C \cdot G \cdot (2\theta)_{111}$ and $\Delta C \cdot G \cdot (2\theta)_{200}$ are the angular deviations of the gravity center from the peak maximum of the $\{111\}$ and $\{200\}$ XRD peaks, respectively.

Figure 3 depicts the variation in the microstructure parameters of the Cu-Al-Zn alloys deformed via LNR and RR routes along with SFE. As shown in Fig. 3(a), with decreasing SFE, the crystallite size clearly
reduced. Moreover, at the same SFE, the crystallite size of the LNR samples was smaller than that of the RR samples. In addition, the microstrain increased sharply with lowering SFE while the microstrain of the LNR samples was larger than that of the RR samples at the same SFE.

As SFE decreased, seen in Fig. 3(b), dislocation densities grew rapidly. However, the increment of the dislocation density deformed via the LNR route was larger than that deformed via the RR route. Meanwhile, it can be clearly seen that decreasing SFE led to an increase in twin density \( \beta \), regardless of the processing route adopted. In addition, the twin density of the LNR samples was higher than that of the RR samples at the same SFE.

Figures 4(a) and (b) show the microstructures of Cu-12.1 at.% Al-4.1 at.% Zn alloys deformed via LNR and RR routes, respectively. Deformation temperature showed a pronounced effect on the microstructural features, with larger volume fraction of twins and smaller grain size in the LNR samples than in the RR samples. The microstructure of the RR sample exhibited significantly elongated subgrains/grains with roughly parallel lamellar boundaries and only a few deformation twins (Fig. 4b). Cross-sectional TEM observations revealed that a high density of dislocation cells (DCs) existed in most of the grains for the LNR sample (Fig. 4a). Only very limited deformation twins were found in the form of bundles embedded in some of the deformed grains, marked with white arrows in Fig. 4(a).

The reason that there few twins were observed in the rolled Cu-12.1% Al-4.1% Zn alloy is twofold: (i) some twins may have merged during recrystallization or been devoured by the recrystallization of grains and the twin boundaries (TBs) may gradually have transformed into normal high-angle grain boundaries via accumulation of dislocations at the TBs\(^{30}\); (ii) the number of twins may have approached a saturated condition in the cryogenic rolled Cu-12.1% Al-4.1% Zn alloy\(^{31}\). From Fig. 4, it can also be seen that the grain size in the LNR series was finer than in the RR series. As shown in Fig. 4, the grain size detected by TEM was generally larger than that detected by XRD, because X-ray measures the volume of coherent diffraction, and therefore each volume measured is considered to be defect-free, while TEM measures the volume surrounded by visible boundaries and therefore each volume measured is considered a cell or a subgrain or a grain\(^{17}\).

**Effect of solid solution of atoms on strength**

In single phase alloys, there are three strengthening mechanisms: solid solution hardening, grain size strengthening and dislocation/twin strengthening. Firstly, we focus on comparatively assessing the solid solution strengthening contributions from Zn and Al.

According to Labusch and VÖhringer\(^ {32,33}\), the contribution of solid solution hardening \( (\sigma_s) \) to the yield strength of Cu-based alloy compared with Cu can be calculated by

\[
\sigma_s = M \times \left( \frac{G}{560} \right) \varepsilon L^{4/3} c^{2/3}
\]

where \( M = 3.1 \) is the Taylor factor, \( G = 45 \) GPa is the shear modulus of Cu, \( c \) is the solute concentration in at\% and \( \varepsilon_L \) is the Labusch parameter expressed as

\[ ... (3) \]

![Fig. 4—TEM microstructure of Cu-12.1 at.% Al-4.1 at.% Zn deformed by (a) LNR and (b) RR](image-url)
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In Eq. (4) \( \varepsilon_b \) and \( \varepsilon_G \) correspond to the misfit of atom size and shear modulus of solute and solvent in a solid solution, respectively. Because \( \varepsilon_b = 0.064 \) and \( \varepsilon_G = -0.61 \) between Cu and Al, while \( \varepsilon_b = 0.056 \) and \( \varepsilon_G = -0.38 \) between Cu and Zn. Therefore, \( \varepsilon_L \) is 1.068 and 0.8985 for Cu-Al and Cu-Zn, respectively. The calculated stress components of \( \sigma_{ss} \), for pure Cu and Cu-Al-Zn alloys in the current study are given in Fig. 5 together with corresponding Al and Zn content and SFE. Regardless of the possible interaction between Al and Zn, it can be seen that, with increased Al and Zn content, the contribution to yield strength from \( \sigma_{ss} \) occupied about 32.8 MPa and 67.9 MPa for Cu-2.5 at.% Al-2.5 at.% Zn and Cu-12.1 at.% Al-4.1 at.% Zn, respectively. As shown in Fig. 5, for RR and LNR Cu-2.5at.%Al-2.5at.%Zn alloys, the contribution to yield strength from \( \sigma_{ss} \) occupied about 33% and 24% of the total yield strength increment, respectively. Therefore, the higher strength of the Cu-Al-Zn alloy is related to the higher contribution to solid solution strengthening of Al and Zn compared with Cu. To a certain extent, it is why the strength of the Cu-12.1 at.% Al-4.1 at.% Zn and Cu-2.5 at.% Al-2.5 at.% Zn samples was higher than that of Cu. However, the remaining contribution to yield strength increment could be from grain size and twinning.

\[
\varepsilon_L = \sqrt{\left(15 \varepsilon_0 \right)^2 + \left( \frac{\varepsilon_G}{1+\varepsilon_0/|1/2|} \right)^2} \quad \cdots (4)
\]

Effect of SFE

Figure 1 clearly shows that the yield strength, ultimate strength and uniform elongation increased with decreasing SFE, which demonstrates that SFE plays a dominant role in the mechanical properties of pure Cu and Cu alloys. As shown in Fig. 3 (b), when SFE decreased from 78 mJ/m\(^2\) to 7 mJ/m\(^2\), the dislocation density of the LNR samples and the RR samples increased from 0.108\(\times10^{15}\) m\(^2\) to 4.526\(\times10^{15}\) m\(^2\) and from 0.033\(\times10^{15}\) m\(^2\) to 1.355\(\times10^{15}\) m\(^2\), respectively. Low SFE makes it difficult for a full dislocation to cross-slip and climb a barrier that hinders dislocation recovery via cross slip and climb. According to the Taylor equation, the yield strength \( (\sigma_{0.2}) \) is related to the dislocation density \( (\rho) \), given by:

\[
\sigma_{0.2} = \sigma_0 + \alpha M G b \sqrt{\rho} \quad \cdots (5)
\]

where \( \sigma_0 \) is the friction stress, \( \alpha \) is a constant, \( M \) is the Taylor factor, and \( G \) is the shear modulus. It can be seen that the strength can be well correlated to the dislocation density because the dislocations interact with the change of stacking fault energy.

It is known that the primary deformation mode of FCC metals is dislocation slip. Grain refinement is caused by dislocation gliding, accumulation, interaction, tangling, and spatial rearrangement. Dislocation slip played a major role in grain refinement in the early stage of deformation, so, the grain sizes of Cu-2.5 at.% Al-2.5 at.% Zn samples were so exceptionally small that dislocations were easily annihilated at grain boundaries without accumulations, which led to much higher stress being required for full dislocation to cross slip or climb. This is why the Cu-2.5 at.% Al-2.5 at.% Zn sample had higher strength and lower total elongation than those of the Cu samples. The pinning effect caused via solid solution atoms (which increase with lowering SFE) can hinder dislocation manipulation and rearrangement, and the critical shear stress for deformation twinning is lower than that for sliding. Therefore, twinning becomes the dominant deformation mechanism when SFE reduces to a certain degree.

As shown in Fig. 3 (b), when SFE decreased from 78 mJ/m\(^2\) to 7 mJ/m\(^2\), the twin density of the LNR samples and the RR samples increased from 0.097% to 1.266% and from 0.02% to 0.651%, respectively, which explains the increments of work hardening rate and uniform elongation. Twins can promote work...
Grain refinement effect

Other words, reducing SFE can produce an efficient grain refinement. Figure 2(a) and (b) demonstrate clearly that the samples with lower SFE and larger uniform elongation had higher work hardening rates. Twin boundaries can effectively subdivide the original grain into a new, more refined microstructure. In other words, reducing SFE can produce an efficient grain refinement effect. Figure 3(a) shows clearly that this phenomenon is consistent with the variation in grain size. Lowering SFE may improve ductility due to an increase in twinning capability. Hence, the elongation of Cu-12.1 at.% Al-4.1 at.% Zn was higher than that of Cu-2.5 at.% Al-2.5 at.% Zn.

Effect of temperature

As shown above, rolling at LNT leads to boosted grain refinement and associated strengthening compared to rolling at RT. The results show clearly that deformation with decreasing temperature not only effectively suppressed the dynamic recovery and recrystallization, but also resulted in enhanced yield strength with significant ductility for the samples. Ductility was improved in these materials at low temperature, owing to a uniform and efficient storage of dislocations. Uniform and efficient storage of dislocations results in a high strain hardening rate at low temperature, because the annihilation of dislocations through thermally activated cross-slip and climb and at grain boundaries is suppressed. It is well known for conventional FCC metals that their yield strength is less sensitive to temperature, whereas their strain hardening strongly depends on temperature. As clearly shown in Fig. 3(a), the measured microstrain of the LNR samples was larger than that of the RR samples. It illustrates that lowering the temperature can introduce more defects (dislocations), which corresponds to the measured lowering grain size (Fig. 3). Microstrain determined from XRD peak broadening revealed concentrations of lattice defects (dislocations and twins) in the samples.

Given the above, with lowering temperature, there will be a transition of deformation mechanism from that dominated by dislocation activities to that dominated by deformation twinning, and the transition temperature increases with reducing SFE. Moreover, the extent of the increase in strength can also be attributed to the formation of deformation twins. It has been analytically explained by Meyers et al. in comparison with the Hall-Petch slopes \( K_T \) for slip and \( K_T \) for twinning, it can be found that:

\[ K_T = (1.5–7) K_S \]

which implies that twin boundaries can hinder dislocation slip more effectively than general grain boundaries. In most FCC metals, twinning increases in importance when the temperature is lowered. This is often formally represented by a twinning stress versus temperature curve that increases less steeply at low temperatures than does the yield stress for plastic deformation by slip. Mahajan and Williams in their 1973 research suggested that the twinning stress of FCC metals actually has a positive dependence on temperature. So the transition from slip to twinning may be governed by lowering the temperature. The slow variation of twinning stress with temperature and the prevalence of twinning at low temperatures both support the conclusion that twin nucleation is not the effect of thermally activation but rather occurs at places of high stress concentrations. Once they have nucleated, there is evidence that, over an appreciable temperature range, twins can grow more readily than slip can propagate.

Compared to traditional high-angle grain boundaries, twin boundaries (TBs) are coherent boundaries with lower energy; but they can play the same role in the inhibition of dislocation movements as high-angle grain boundaries. Coherent TBs cannot only serve as storages of dislocations, but can also inhibit the transmission of dislocations from one side of the TB to the other. This is because the external force driving the motion of the partial dislocations is not sufficiently high to drive them across the grain, or because dislocation interactions stop partial dislocations and prevent them from slipping to the other end of the grain. Therefore, twins play an effective role in increasing the dislocation storage capacity. In addition, research has also indicated that twins can effectively suppress the dynamic recovery of dislocations; thus, an increase in twin density also contributes to an increase in dislocation density.

Grain refinement is caused by dislocation gliding, accumulation, interaction, tangling and spatial rearrangement. Lowering the SFE of the alloys led to a decrease in twinning stress, which made the formation of deformation twins significantly easier, thus resulting in a higher twin density. TB/GB intersections subdivided the original grains efficiently and led to the formation of a new refined microstructure, which resulted in finer grain size in the microstructure. In general, the cold-rolling process...
induces an obvious increase in the microstrain, which can be attributed to the local deviation of the atom from the equilibrium position due to the presence of impurities, point defects, dislocations or grain boundaries. Microstrain in the material is, to a certain extent, a signature of the high density of defects (dislocations or twins) within the grain boundaries and it increases with a reduction in grain size and increases in dislocation and twin densities.

As deformation temperature reduced, the driving stress of twin nucleation in the deformed samples increased and twinning was a much more regulated process than slip. Therefore, decreasing the deformation temperature facilitated the formation of deformation twins during the plastic deformation process. This is consistent with the data in Fig. 3(b). Consequently, it can be concluded that the LNR samples with low SFE were strengthened and toughened more significantly than those RR samples.

Conclusions

By rolling at different temperatures, the effects of the deformation temperature parameter on the deformation mechanisms and mechanical properties in Cu and Cu-Al-Zn alloys with different levels of stacking fault energy (SFE) have been investigated. This has shown that lowering SFE by use of enhanced Al and Zn content simultaneously improved both strength and ductility. Compared with RR samples with the same low SFE, LNR samples had higher strength and better ductility. The elongation of the LNR samples was larger than that of the RR samples, so that smaller grain size and higher microstrain were gained at liquid nitrogen temperature. Dislocation density and twin density both increased with decreasing temperature because of the suppression of dynamic recovery and the enhancement of twinning tendency through increasing strain rate or decreasing deformation temperature. Deformation twinning is a significant deformation mechanism that can improve the work hardening rate, strength and ductility of UFG materials. In addition, the LNR method is better for improving the mechanical properties of the materials than the RR method.

Acknowledgement

This research is financially supported by the National Natural Science Foundation of China (NSFC) through the NSFC grant (Grant No. 50874056 and 51361017).

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