Effect of stacking fault energy on mechanical properties and annealing behavior of brasses

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Abstract

The effect of stacking fault energy (SFE) on mechanical properties and annealing behavior of brasses was studied in Cu-20Zn alloy (SFE ~ 18 mJ/m²) and Cu-20Zn-1.2Si alloy (SFE ~ 9 mJ/m²) as well as Cu-20Zn-1.9Si alloy (SFE ~ 6 mJ/m²) alloy. These brasses have been rolled at room temperature up to different thickness reductions. The significant improvement of strength in Cu-20Zn-1.9Si alloy is attributed to the formation of fine grains and high densities of dislocations and deformation twins by decreasing the SFE. Thermal stability is enhanced due to the reductions of dislocation mobility and grain boundaries migration during annealing by a decrease in SFE and addition of Si. High fractions of the {236} {385} Brass-R and (55;30;0) in Cu-20Zn-1.9Si alloy with lower SFE should be ascribed to the

substantial Brass texture in deformed microstructures. Fine grains, deformation twins and abundant annealing twins were introduced into Cu–20Zn–1.9Si alloy by decreasing the SFE, resulting in superior strength–ductility combination.

Keywords: *copper alloy; rolling; mechanical properties; twins; annealing.*

1. Introduction

It has been widely acknowledged that the dislocation slip and twinning are two competitive plastic deformation mechanisms in face-centered cubic (fcc) metals [1,2]. Stacking fault energy (SFE) is an important parameter which has closely related to the deformation mechanisms. To date, most researches have concentrated on in metals with relatively high values for SFE so that dislocation slip would be the dominant deformation mechanism in plastic deformation [3–5]. With decreasing the SFE, the deformation mechanism will transfer from dislocation slip to twinning, and the grain refinement mechanism in plastic deformation will also change [6]. In metals with the high or medium SFE, the substantial dynamic recovery occurs by balancing multiplication and annihilation of dislocations, more interactions between dislocations and grain boundaries are in favor of the annihilation processes. Accordingly, no more dislocation boundaries can be formed, impeding the further subdivision of grains [7]. Twinning, a prevalent deformation mechanism in low SFE metals, plays a significant role in grain refinement. It primarily stems from the interactions between dislocations and twin boundaries resulting in the fragmentation of twin bundles. Meanwhile, the role of shear bands in refining the original coarse grains is also crucial. Therefore, it is reasonable to expect that the grain size can be refined down to the nanoscale for metals with low or extremely low SFE even under relatively gentle deformation conditions [8,9].

Previous study [7] indicated that the strength and uniform elongation were simultaneously improved by decreasing the SFE in Cu–Al alloys. This simultaneity was attributed to the formation of profuse deformation twins and shear bands, and their extensive intersections. Although the overall mechanical properties were

improved with decreasing the SFE, the uniform elongation was still limited. Therefore, in recent years much attention has been paid to developing strategies for improving the limited ductility [10–14]. Thermal annealing has been found to be effective on improving the ductility. It was found that SFE plays crucial role in thermal annealing [15–17].

Therefore, in this work, regarding the significance of SFE in plastic deformation and thermal annealing, the effect of SFE on the mechanical properties and annealing behavior of Cu–Zn and Cu–Zn–Si alloys was investigated. Meanwhile, the effect of SFE on the recrystallization texture and strength–ductility combination of these alloys during annealing was also studied.

2. Materials and Methods

Cu–20%Zn, Cu–20%Zn–1.2%Si and Cu–20%Zn–1.9%Si alloys (all compositions are in weight percent) were prepared by induction vacuum melting. The addition of Si, the valance of which is four, leads to a significant decrease in SFE [18]. The ternary alloy is especially designed to obtain minimum SFE in copper rich alloys. SFEs of the ternary alloys were calculated using the following equation and available data on the SFEs of binary alloys [19,20]:

$$\frac{\gamma - \gamma_{\text{Cu-Zn}}}{\gamma_{\text{Cu-Si}} - \gamma_{\text{Cu-Zn}}} = \frac{C_a - (\text{Al})_E}{(\text{Zn})_E - (\text{Al})_E} = \frac{2C_a}{e/a - 1} - 1 \quad (1)$$

where $\gamma_{\text{Cu-Zn}}$ and $\gamma_{\text{Cu-Si}}$ are SFEs for binary alloys with the same electron/atom ratio (e/a) as the ternary solid solution and with the respective atomic fraction contents $(\text{Al})_E$ and $(\text{Zn})_E$. C_a is the total atomic fraction of solute content. SFEs of Cu–20Zn, Cu–20Zn–1.2Si and Cu–20Zn–1.9Si alloys are 18 mJ/m², 9 mJ/m² and 6 mJ/m², respectively.

In order to homogenize the composition, the casting ingots with thickness of 15 mm were heated at 830 °C for 1 h and then hot-rolled to thickness of 5.5 mm. The plates were finally annealed at 490 °C for 50 min and then quenched in water. These alloys were then rolled at room temperature up to different thickness reductions. The

X-ray diffraction (XRD) measurements were carried out on a Rigaku X-ray diffractometer equipped with a Cu target. The Vickers hardness was measured at room temperature under a load of 1000 g and for a duration time of 30 s. Measurements at least 9 different points for each sample were taken to obtain an average hardness. Tensile specimens have gauge lengths of 6 mm and widths of 1 mm. Tensile tests were conducted at an initial strain rate of $3 \times 10^{-3} \text{ s}^{-1}$ at room temperature. Electron backscatter diffraction (EBSD) observations were made in the longitudinal plane containing the rolling direction (RD) and normal direction (ND), the rolling plane containing the RD and transverse direction (TD), and the cross plane containing the TD and ND. To make reliable measurements, all low angle grain boundaries with a misorientation less than 2° were excluded. The misorientation of 15° was used as the criterion to distinguish the low angle grain boundaries (LAGBs) and high angle grain boundaries (HAGBs). Microstructures in the rolling plane and longitudinal plane were also determined using the transmission electron microscope (TEM) technique. Thin foils were prepared by utilizing a conventional jet polishing technique in a solution of 25% HNO_3 and 75% Methanol.

3. Results and Discussion

3.1 Deformed microstructures and tensile properties of brasses

The three-dimensional EBSD microstructures of the Cu–20Zn and Cu–20Zn–1.9Si alloys after a thickness reduction of 30% are shown in Fig. 1. Cu–20Zn alloy exhibits relatively coarse grains compared to Cu–20Zn–1.9Si alloy in all three planes. Cu–20Zn alloy shows deformed grains, which are elongated along the RD and TD in the longitudinal plane and cross plane, respectively. However, the severely fragmented and elongated grains were observed in Cu–20Zn–1.9Si alloy. As compared to Cu–20Zn alloy, the length of the elongated grain size is less in Cu–20Zn–1.9Si alloy.

Fig. 2 shows the micrographs of Cu–20Zn and Cu–20Zn–1.9Si alloys after a thickness reduction of 90%. A heavily deformed microstructure and dislocation cells

were observed in Cu–20Zn alloy, as shown in Fig. 2a. The microstructures of Cu–20Zn alloy exhibit lamellar grains lying parallel to RD in Fig. b. As compared to Cu–20Zn alloy, a severely deformed microstructure with ill-defined grain boundaries and much higher dislocations density as well as lamellar grains were observed in Cu–20Zn–1.9Si alloy. Moreover, deformation twins (marked by circles in Fig. 2b, d) inclined to the RD by an angle of 45° were observed in Cu–20Zn–1.9Si alloy.

The dislocation density, twin density and grain size of the Cu–20Zn, Cu–20Zn–1.2Si and Cu–20Zn–1.9Si alloys were measured by using XRD analysis. The dislocation density ρ can be calculated based on microstrain, using the following equation [19]:

$$\rho = 16.1 \times \varepsilon^2 / b^2 \quad (2)$$

where ε is the microstrain, b is the Burgers vector and $b = (\sqrt{2}/2)a$, and a is the lattice constant in fcc metals. The twin density β , defined as the probability of finding a twin boundary between any two neighboring $\{111\}$ planes, may be calculated based on the equation [21, 22]:

$$\beta = \frac{\Delta C.G.(2\theta)_{111} - \Delta C.G.(2\theta)_{200}}{11 \tan \theta_{111} + 14.6 \tan \theta_{200}} \quad (3)$$

where $\Delta C.G.(2\theta)_{111}$ and $\Delta C.G.(2\theta)_{200}$ are the angular deviations of the gravity center from the peak maximum of the $\{111\}$ and $\{200\}$ XRD peaks, respectively. The grain size can be calculated by the Williamson–Hall method [23]. It is known that XRD analysis often yields a smaller grain size because it measures the sizes of coherent-diffraction domains [24]. The calculated results of these three main factors are shown in Table 1. An increase in the dislocation density and twin density from $1.6 \times 10^{15} \text{ m}^{-2}$, 0.05% to $6.1 \times 10^{15} \text{ m}^{-2}$, 0.26% when the SFE decreases from 18 to 6 mJ/m² for Cu–20Zn alloy and Cu–20Zn–1.9Si alloy, respectively. The grain size decreases from 103 to 42 nm with decreasing SFE from 18 to 6 mJ/m².

It is well known that SFE has close relationship with the deformation mechanisms [25]. On one hand, a decrease in SFE facilitates the full dislocations to split into two partials with a wide stacking fault between them, the lower the SFE, the wider the

stacking fault [26]. The stacking fault can acts as a barrier for full dislocations to cross slip or climb, which suppresses the dislocation recovery via cross slip and climb [23]. On the other hand, with decreasing SFE, the stress necessary for dislocation slip is greater than that for twinning, and deformation twins would easily form with decreasing SFE [1]. Moreover, deformation twins caused by lowering SFE may serve as locations for dislocation accumulation [27]. Consequently, a lower SFE usually leads to a higher dislocation density and twin density. As proved by the results of TEM and XRD.

SFE determines not only the deformation mechanisms but also the grain refinement mechanisms. For metals with medium to high SFE, dislocation subdivision is the dominant grain refinement mechanism due to the extensive dislocation movement. With a decrease in SFE, deformation twins would be prevalent in plastic deformation [28,29]. The grain refinement mechanism gradually transfers from the dislocation subdivision to twin fragmentation. The twin fragmentation is more effective in grain refinement [30]. Therefore, finer grain size is achieved in metals with lower SFE.

Fig. 3 shows the engineering stress–strain curves of Cu–20Zn, Cu–20Zn–1.2Si and Cu–Zn–1.9Si alloys after a thickness reduction of 90%. A significant improvement in ultimate tensile strength (UTS) from 675 MPa to 1002 MPa, and an improvement in uniform elongation (UE) and total elongation from 3.8%, 8.5% to 5.4%, 9.6% when the SFE decreases from 18 to 6 mJ/m² for Cu–20Zn alloy and Cu–20Zn–1.9Si alloy, respectively. The large enhancement in UTS with decreasing SFE derives from fine grain and high densities of dislocations and deformation twins, as indicated in previous experiments [16]. The solid solution of atoms plays a role in improving the strength. However, its influence is negligible compared with other strengthening mechanisms [7]. The inset shows curves of normalized hardening rate–true strain. The work hardening rate θ can be defined by Eq. (4) [31]:

$$\theta = \frac{1}{\sigma} (\partial \sigma / \partial \varepsilon) \dot{\varepsilon} \quad (4)$$

where σ is the true stress, ε is the true strain and $\dot{\varepsilon}$ is the strain rate applied. It can be seen that the work hardening rate increases with decreasing SFE. The work

hardening rate is closely related to the evolution of dislocation density in plastic deformation [32]. The higher work hardening rate in Cu–20Zn–1.9Si alloy is attributed to the high dislocation accumulation capacity caused by deformation twins and suppression of dynamic recovery.

3.2 Annealing behavior of brasses

Fig. 4 illustrates the hardness evolution of Cu–20Zn and Cu–20Zn–1.9Si alloys annealing at different temperatures for 1 h. Three stages can be seen in Fig. 4. During stage 1, a slight decrease in hardness was detected in Cu–20Zn alloy when the annealing temperature is below 240 °C. It indicates that recovery dominates these processes. However, Cu–20Zn–1.9Si alloy exhibits annealing hardening phenomenon at stage 1. An apparent increase in hardness was observed and maximum hardness was achieved at 260 °C. The annealing hardening phenomenon can be attributed to the addition of Si, leading to solute segregation to dislocation which is analogous to the formation of Cottrell atmospheres in interstitial solid solution during recovery [33]. During stage 2, a dramatic decrease in hardness was observed in Cu–20Zn and Cu–20Zn–1.9Si alloys when annealing temperatures are beyond 240 °C and 280 °C, respectively. This must be closely related to the beginning of recrystallization. The higher temperature for the beginning of recrystallization implies that the thermal stability of Cu–20Zn–1.9Si alloy is relatively better than that of Cu – 20Zn alloy. The introduction of Si results in an interaction between dislocations and solute atoms, which reduces dislocation mobility and the grain boundaries migration during recovery. Furthermore, a decrease in SFE hinders dislocation movement [16,34]. Therefore, thermal stability is enhanced during annealing. In the final stage 3, the hardness in Cu–20Zn alloy decreases very slowly due to the conventional grain growth after recrystallization. While the hardness in Cu–20Zn–1.9Si alloy scarcely change. It indicates that the grain growth is inhibited in Cu–20Zn–1.9Si alloy. It is well known that grain growth involves the migration of grain boundaries [35]. Addition of Si pins grain boundaries, which would impede further grain growth after

recrystallization.

Fig. 5 shows unique grain color map of Cu–20Zn and Cu–20Zn–1.9Si alloys after annealing at 340 °C and 360 °C, respectively. Grain size distributions (based on diameter) of these two alloys are also shown in Fig. 5. Every color stands for a unique grain with a misorientation larger than 15°. Most grains in Cu–20Zn alloy appear equiaxed with its size in the range of 1µm to 5µm. Meanwhile, abnormal grain growth (secondary recrystallization) has started to produce an area fraction (~ 5%) of coarser (5.5–8.5 µm) grains, as shown in Fig. 5a, b. This is consistent with a slight decrease in hardness at stage 3 in Fig. 4. The equiaxed grains are distributed homogeneously in Cu–20Zn–1.9Si alloy and there is no abnormal grain growth. The average grain size $\bar{d}$ in Cu–20Zn–1.9Si alloy (1.4µm) is finer than that in Cu–20Zn alloy (2.9 µm).

Fig. 6 presents the distributions of the misorientation angle of Cu–20Zn and Cu–20Zn–1.9Si alloys after annealing at 340 °C and 360 °C, respectively. The misorientation distributions are characterized by a sharp peak near 60°, which are attributed to the significant fraction of $\Sigma 3$ twin boundaries. The $\Sigma 3$ twin boundaries fractions in Cu–20Zn alloy and Cu–20Zn–1.9Si alloy are 24.4 % and 36.2 %, respectively. Annealing twins can be formed during recrystallization driven by minimizing the total excess energies of boundaries separating newly formed grains, and stacking faults would serve as nuclei for annealing twins [36]. As shown in Fig. 7, the annealing twins originated at the stacking faults (marked by circles) and extended into the dislocation-free recrystallized grain. Therefore, profuse annealing twins were introduced into the Cu–20Zn–1.9Si alloy by decreasing the SFE. As expected, a large number of annealing twins would increase the proportions of $\Sigma 3$ twin boundaries.

A lower SFE results in a higher defect density, leading to an enhancement in driving force for recrystallization. Consequently, there are more nucleation sites, resulting in finer size of SRX grains [37]. Furthermore, the high formation rate of annealing twins in lower SFE metals may also play crucial roles in hindering grain growth during recrystallization [35]. Compared with conventional HAGBs, $\Sigma 3$ twin boundaries are more stable due to the excess energy of $\Sigma 3$ twin boundaries is one

order of magnitude lower than that of HAGBs [35,36]. These $\Sigma 3$ twin boundaries can impede grain boundaries migration and then delay the growth of newly generated grains [35]. Accordingly, abnormal grain growth is inhibited and finer grain size is achieved in Cu–20Zn–1.9Si with lower SFE.

Fig. 8 shows distributions of the main texture components of Cu–20Zn and Cu–20Zn–1.9Si alloys annealed at 300 °C and 340 °C, respectively. $\Sigma 3$ twin boundaries are designated by thin gray lines. For comparison, several ideal rolling and recrystallization textures in fcc metals are listed in Table 2. Both Cu–20Zn and Cu–20Zn–1.9Si alloys mainly comprised {236}<385> Brass–R and (55;30;0). The fractions of rolling textures were small. The fractions of the {236}<385> Brass–R and (55;30;0) in Cu–20Zn–1.9Si alloy are 22.2% and 15.1%, respectively. However, lower fractions of these two textures in Cu–20Zn alloy are 12.8% and 8.5%, respectively.

The recrystallized texture is dependent on the character of the deformed microstructures. The formation of Brass texture has been attributed to the suppression of dislocation movement. Brass texture would be the dominant texture in the deformed microstructures with a decrease in SFE [38–40]. Brass–R texture, the major component of the recrystallization texture of fcc metals with low SFE, is most favorable to growth into the microstructures with the brass type rolling textures [41]. Moreover, a decrease in SFE would facilitate the formation of deformation twins. Due to the deformation twins in the deformed microstructures, the newly formed deformation twins possess the {236}<385> Brass–R orientation, and this can grow very quickly during annealing process.[42]. Owing to (55;30;0) orientation is relatively close to Brass. It is possible that the (55;30;0) grains derived from orientation spread of the Brass component[39]. Therefore, it is reasonable to expect that higher fractions of the {236}<385> Brass–R and (55;30;0) obtained in Cu–20Zn–1.9Si alloy.

Fig. 9 shows the effect of SFE on the strength–ductility relationship. The strength–ductility trend of Cu–20Zn–1.9Si alloy with lower SFE is shifted upwards compared to the Cu–20Zn and Cu–20Zn–1.2Si alloys. With the same uniform

elongation, the UTS of Cu-20Zn-1.9Si alloy is higher than that of Cu-20Zn and Cu-20Zn-1.2Si alloys by 100–300 MPa. Or, with the same UTS, Cu-20Zn-1.9Si alloy exhibits a 2–10% higher UE than Cu-20Zn and Cu-20Zn-1.2Si alloys. Clearly, compared with Cu-20Zn and Cu-20Zn-1.2Si alloys, the Cu-20Zn-1.9Si alloy exhibits a superior strength–ductility combination after annealing.

The superior strength–ductility combination of Cu-20Zn-1.9Si alloy may be attributed to several factors. In terms of strength, the high strength with decreasing SFE may originate from two factors. First, the fine grains after annealing impart high strength, as expected from the Hall–Petch relationship. Second, dislocation movement is impeded due to the deformation twins and abundant annealing twins with a decrease in SFE and thus the strength is improved. [7,16,43]. The enhancement in ductility can be attributed to the following three factors. First, it is well known that the UE is determined by the work hardening rate [16]. With a decrease in SFE, cross slip is significantly blocked as a dynamic recovery mechanism and thus the work hardening rate is improved [16]. Second, with decreasing SFE, the formation of high density of annealing twins may in favor of accumulating more dislocations to improve the work hardening ability. Moreover, stacking fault may interact effectively with slipping dislocations, leading to an increase in work hardening ability. Therefore, the ductility is enhanced [44-46]. Third, the easy formation of deformation twins during tensile test in the recrystallized grains not only provides ample room for dislocation storage but also improves the work hardening rate, thereby to further enhance the ductility [47,48].

4. Conclusions

The effect of stacking fault energy (SFE) on mechanical properties and annealing behavior of brasses was studied and can be summarized as follows:

- (1) Investigations by X-ray diffraction indicate that a decrease in SFE leads to both a decrease in the grain size and an increase in the densities of dislocations and deformation twins.

- (2) A significant increase in the strength has been observed by lowering SFE. This is due to the formation of fine grains and high densities of dislocations and deformation twins.
- (3) Thermal stability is enhanced due to the reductions of dislocation mobility and grain boundaries migration during annealing by a decrease in SFE and addition of Si.
- (4) High fractions of the {236}<385> Brass-R and (55;30;0) in Cu-20Zn-1.9Si alloy with lower SFE may result from the substantial Brass texture in deformed microstructures.
- (5) The superior strength-ductility combination in Cu-20Zn-1.9Si alloy with lower SFE originates from the fine grains, deformation twins and abundant annealing twins.

Acknowledgements

The authors gratefully acknowledge support from the National Science Foundation of China (NO. 51174234).

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Figures captions

Fig. 1. Three-dimensional EBSD micrographs of the Cu-20Zn and Cu-20Zn-1.9Si alloys at a thickness reduction of 30%: (a) Cu-20Zn alloy; (b) Cu-20Zn-1.9Si alloy.

Fig. 2. TEM micrographs of Cu-20Zn and Cu-20Zn-1.9Si alloys at a thickness reduction of 90%; Cu-20Zn alloy: (a) rolling plane (b) longitudinal plane Cu-20Zn-1.9Si alloy: (c) rolling plane (d) longitudinal plane.

Fig. 3. Engineering stress-strain curves of the Cu-20Zn, Cu-20Zn-1.2Si and Cu-20Zn-1.9Si alloys at a thickness reduction of 90%. The inset shows curves of normalized hardening rate-true strain.

Fig. 4. Hardness data of Cu-20Zn and Cu-20Zn-1.9Si alloys annealed at different temperatures for 1 h.

Fig. 5. (a) Unique grain color map and (b) Grain size distributions of Cu-20Zn alloy annealed at 340 °C. (c) Unique grain color map and (d) Grain size distributions of Cu-20Zn-1.9Si alloy annealed at 360 °C.

Fig. 6. Misorientation angle distributions of (a) Cu-20Zn alloy after annealing (b) Cu-20Zn -1.9 Si alloy after annealing.

Fig. 7. TEM micrograph of annealing twins originated from stacking faults (marked by circles).

Fig. 8. Distributions of the main texture components within the EBSD maps: (a) Cu-20Zn annealed at 300 °C; (b) Cu-20Zn-1.9Si annealed at 340 °C. ($\Sigma 3$ twin boundaries are designated by thin gray lines)

Fig. 9. Variation in UTS with UE for Cu-20Zn, Cu-20Zn-1.2Si and Cu-20Zn-1.9Si alloys under different annealing conditions.

Fig. 1.

Fig. 2.

Fig. 3.

Fig. 4.

Fig. 5.

Fig. 6.

Fig. 7.

Fig. 8.

Fig. 9.

Table 1 Dislocation density ρ , twin density β and average grain size $\bar{d}$ of brasses

	Cu-20Zn	Cu-20Zn-1.2Si	Cu-20Zn-1.9Si
$\gamma, mJ / m^2$	18	9	6
$\rho, 10^{15} m^{-2}$	1.6	4.55	6.1
$\beta, \%$	0.05	0.14	0.26
$\bar{d}, nm$	103	51	42

Table 2 Several ideal rolling and recrystallization textures for face-centered cubic metals.

orientation	Euler angles (deg)			Miller indices	
	φ_1	ϕ	φ_2	Rolling plane	Rolling direction
Rolling textures					
Brass	35	45	0	{011}	$\langle 211 \rangle$
Goss	0	45	90	{011}	$\langle 100 \rangle$
Cube	0	0	0	{001}	$\langle 100 \rangle$
Dillamore	90	27	45	{4;4;11}	$\langle 11;11;8 \rangle$
Recrystallization textures					
Cube-RD	0	22	0	{013}	$\langle 100 \rangle$
Brass-R	80	31	35	{236}	$\langle 385 \rangle$