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Effect of Cold Rolling and Annealing Temperature on the Characteristics of Cu-28Zn-1.1Bi Alloy Produced by Gravity Casting

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Abstract. The microstructures, mechanical properties, deformation mechanism, and recrystallization behavior of Cu-Zn-Bi alloys for cartridge case application have been investigated in this research. Cu-28Zn-1.1Bi wt. % alloys were produced by gravity casting and subjected to a homogenization – cold rolling – annealing sequences with variations on reduction level and annealing temperature. Samples characterizations were done through optical emission spectroscopy, optical microscopy, SEM-EDS imaging, and X-ray mapping modes, while hardness measurements were performed using micro Vickers method. The presence of Bi was found to increase cartridge brass hardness through a dispersoid strengthening mechanism in which dislocation movements are rendered by Bi particles. Higher deformation levels resulted in higher microhardness of the alloy. Recrystallization took place at grain boundaries and areas surrounding Bi dispersoid at 400 ^{o}C, while further heating resulted in grain growth phenomenon. Bismuth addition accelerated the recrystallization process in cartridge brass by a particles stimulated nucleation (PSN) mechanism.

Introduction

In its purpose for cartridge case application, cartridge brass (Cu-(28-30)Zn wt. %) alloys are required to possess fine deep-drawability, machinability, and internal pressure tightness, which all are observed in lead-brass alloys [1]. All alloys composition in this paper is mentioned in wt. % unless otherwise stated. Bi was developed to substitute alloying constituent Pb in cartridge brass due to their similar properties and a more environmental-friendly nature [2]. Investigation by Emelina [3] on Cu-30Zn-1Bi found that Bi was not completely soluble in brass furthermore tended to segregate at grain boundaries area. A corresponding finding was observed in a research by Basori [4] on microstructures of Cu-29Zn alloys with addition of 0.269, 0.409, and 0.797 Bi. This research also emphasized that optimal mechanical properties of the respective alloys was found at 0.269 Bi addition, while further addition resulted in significant decrease in tensile strength, yield strength, and elongation of the alloys [4].

Cartridge brass alloys have transitioning deformation mechanisms at different reduction levels which are identified as *slip*, *twinning*, and *shearing* mechanism [5]. Cu-30Zn deformation at 40 % and above resulted in shear bands, while slip mechanism was predominant at low deformation level (< 20%) [6]. Previous research by Sofyan and Basori [7] on deformation mechanism of cold-rolled Cu-32Zn alloys observed the presence of slip, twinning, and shear bands at 20 %, 40 %, and 70 % reduction level, respectively. However, it was found that only cross slip was present in cold-rolled Cu-29Zn-0.6 Bi at 40 % reduction due to the occurrence of Bi segregation, responsible for hindering the formation of twinning and shear bands [1].

Conventionally, annealing treatment is used to recover alloys ductility and toughness after deformation process in order to achieve a certain specification on materials properties. Resulted materials properties are highly dependent on annealing temperatures and duration. Cold-rolled

Cu-32Zn of highest deformation level by Sofyan and Basori [7] was subjected to annealing at varying temperatures of 300, 400, and 600 ^{o}C for 30 min., with results showing that recrystallization was visible at 300 ^{o}C. Basori [1] has found that the addition of 0.6 Bi to Cu-28Zn alloys accelerated the recrystallization process of the cold worked alloys where samples with Bi content underwent a full recrystallization at 400 ^{o}C in 15 min. while Cu-32Zn base alloys required more than 30 min. at the same annealing temperature and cold working level. It was suggested that Bi addition generated a Particle Stimulated Nucleation (PSN) mechanism – nucleation at areas with high interface energy, preferably areas with great disorientation angle between sub-grain and matrix – which was responsible for accelerating the recrystallization rate of cartridge brass alloy [9]. In this research, a higher Bi content of 1.1 was used to further determine the effect of Bi inclusion at grain boundaries area to deformation mechanism and recrystallization phenomenon of cartridge brass alloys.

Experimental Method

Materials. Bi-alloyed cartridge brass alloys were prepared using 99.50 % Cu rods from UD Metallindo Sejahtera, 99.99 % Zn ingots from Korea Zinc Co. Ltd. Onsan Complex, and 99.99% Bi ingots with borax flux melted in an inductance melting furnace and gravity casted at 1150 ^{o}C into a 110 x 110 x 6 mm^3 AISI H-13 steel mold. Resulting plates were subjected to homogenization at 800 ^{o}C for 2 h followed by air cooling. Nominal composition of as-homogenized Cu-Zn-Bi alloys was investigated using Oxford Instrument Foundry-Master Pro Optical Emission Spectroscopy (OES) with result presented in Table 1, showing that chemical composition of produced alloy matched the initial design.

Cold Rolling. Specimens with initial thickness of 6 mm prepared from the as-homogenized plates were directly cold rolled using ONO Roll two-high mill set for 5 %, 10 %, and 20 % thickness reduction at 18000 kgf rolling load. Cold rolled specimens of highest reduction level were annealed at 300, 400, and 600 ^{o}C for 30 min in a muffle furnace to observe the effect of increasing thickness reduction to the recrystallizaion behavior of the brass alloy. As-annealed specimens were water quenched.

Characterizations. The longitudinal section samples from as-homogenized, rolled, and annealed alloys were prepared using standard metallographic preparation in accordance to ASTM E3-11 and etched for 6-7 s by ferric chloride (10 g $FeCl_3$ + 100 mL distilled water) for microstructure imaging using Zeiss optical microscope (OM) and JEOL JSM-6510 LA Scanning Electron Microscope (SEM). Energy Dispersive Spectroscopy (EDS) and X-ray Mapping investigation were done simultaneously with SEM imaging using Oxford Quanta 650 instrument to identify the present phases. Micro Vickers hardness measurements were done on the specimens according to ASTM E 384 with 300 kgf indentation force and 10 s dwelling time.

Table 1. Nominal composition of as-homogenized Cu-Zn-Bi alloys in wt. %.

Zn	**Bi**	Cr	Sn	Co	Sb	Pb	Fe	**Cu**
27.60	**1.10**	0.451	0.116	0.047	0.027	0.025	0.015	*bal.*

Results and Discussion

Microstructure Evaluation. As-homogenized images of Cu-28Zn-1.1Bi wt. % alloys captured using SEM instrument are given in Figure 1. Red arrows indicate the presence of globular particles on brass matrix that is confirmed as Bi dispersoid by EDS analysis result presented in Figure 2 and Table 2. High Bi content was found in both spectra 30 and 31, while no trace of Bi was observed in the α-brass matrix (spectrum 32). It is suggested that the Bi dispersoid were formed due to the low solubility of Bi in both Cu and Zn. Higher Bi alloying content in brass alloys resulted in more quantity of Bi dispersoid with larger particle diameter, as found in comparison of as-homogenized Cu-Zn-Bi alloys images between this work and the previous research by Basori [4]. Further testing using X-ray mapping method with results shown in Figure 3 confirmed the identity of the globular particles as Bi. Color codes were used to determine the distribution of each investigated element; red indicating Cu, green for Zn, and deep orange for Bi elements. The globular particles did not contain any traces of Cu and Zn.

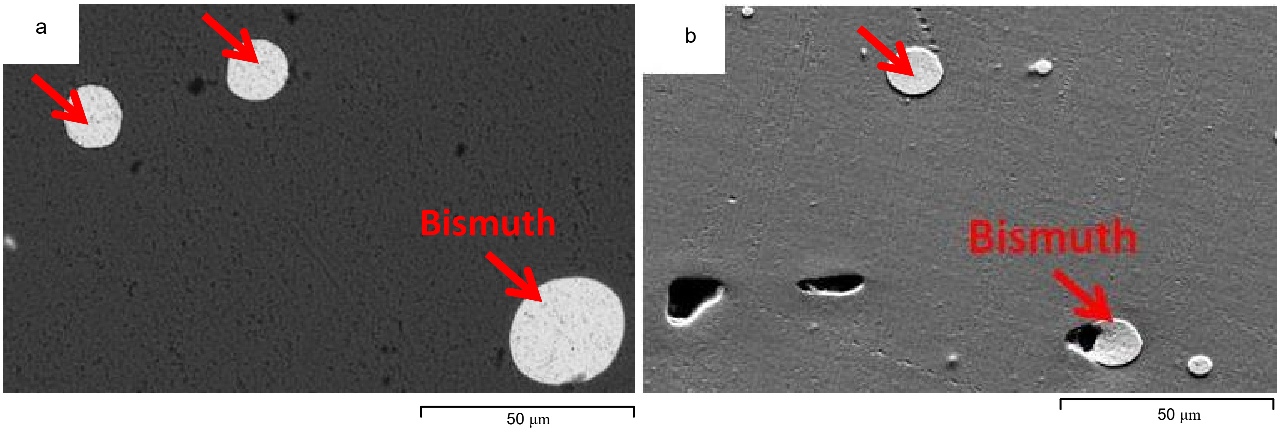

Figure 1. Microstructure of (a) Cu-28Zn-1.1Bi and (b) Cu-28Zn-0.8Bi [4] alloys at 2000x magnification.

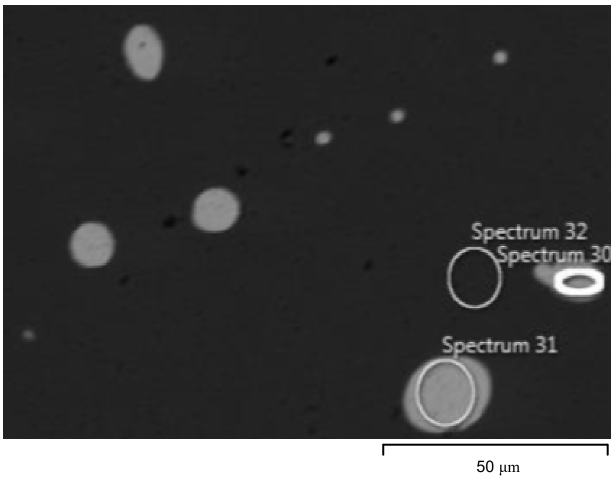

Figure 2. Result of EDS analysis on Cu-28Zn-1.1Bi alloys positioned on spectra 30-32.

Table 2. Nominal EDS analysis results on Cu-28Zn-1.1Bi alloys at positions shown in Fig. 2.

Position	Elements (at. %)		
	Cu	Zn	Bi
30	15.86	7.60	76.54
31	7.60	3.25	89.15
32	71.38	28.62	-

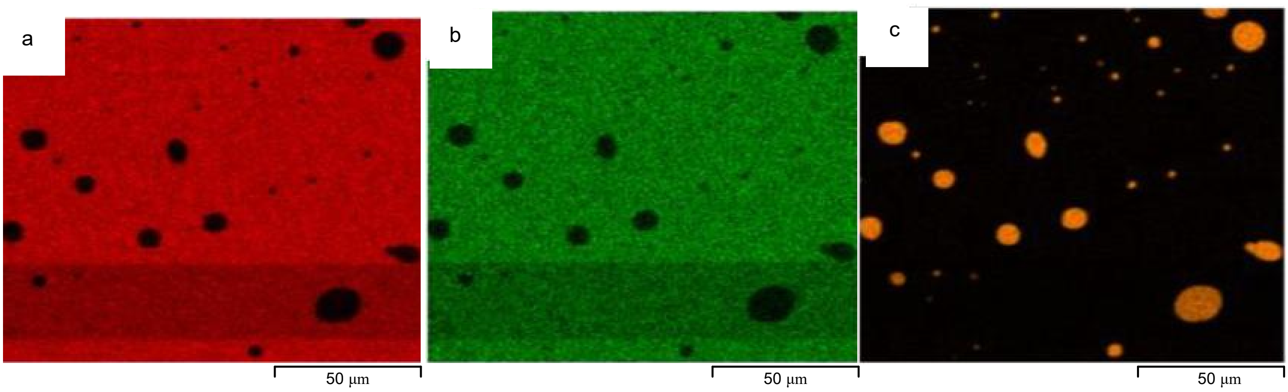

Figure 3. X-ray mapping analysis results on as-homogenized Cu-28Zn-1.1Bi wt. % alloys. Bright fields indicating distribution of each element: (a) Cu, (b) Zn, and (c) Bi.

Deformation Characteristics. Microstructure evolution of as-homogenized Cu-28Zn-1.1Biwt. % alloys during cold rolling at 5 %, 10 %, and 20 % thickness reduction is presented in Figure 4. It can be seen from images comparison in Figure 4 (a,d,g,j) that both brass matrix and globular Bi dispersoid (Figure 4 (b), red circles) were flattened along the rolling direction (Figure 4 (e), green circles), however calculations were only done on the length/diameter (L/d) ratios of initial and deformed dispersoid sizes. L/d ratios of Bi dispersoid in Cu-28Zn-1.1Bi alloy increased from 1.01 to 1.07, 1.32, and 2.64 at 5 %, 10 %, and 20 % thickness reduction, respectively. This indicates that Bi underwent a flattening mechanism from its initial globular shape in as-homogenized samples during cold rolling.

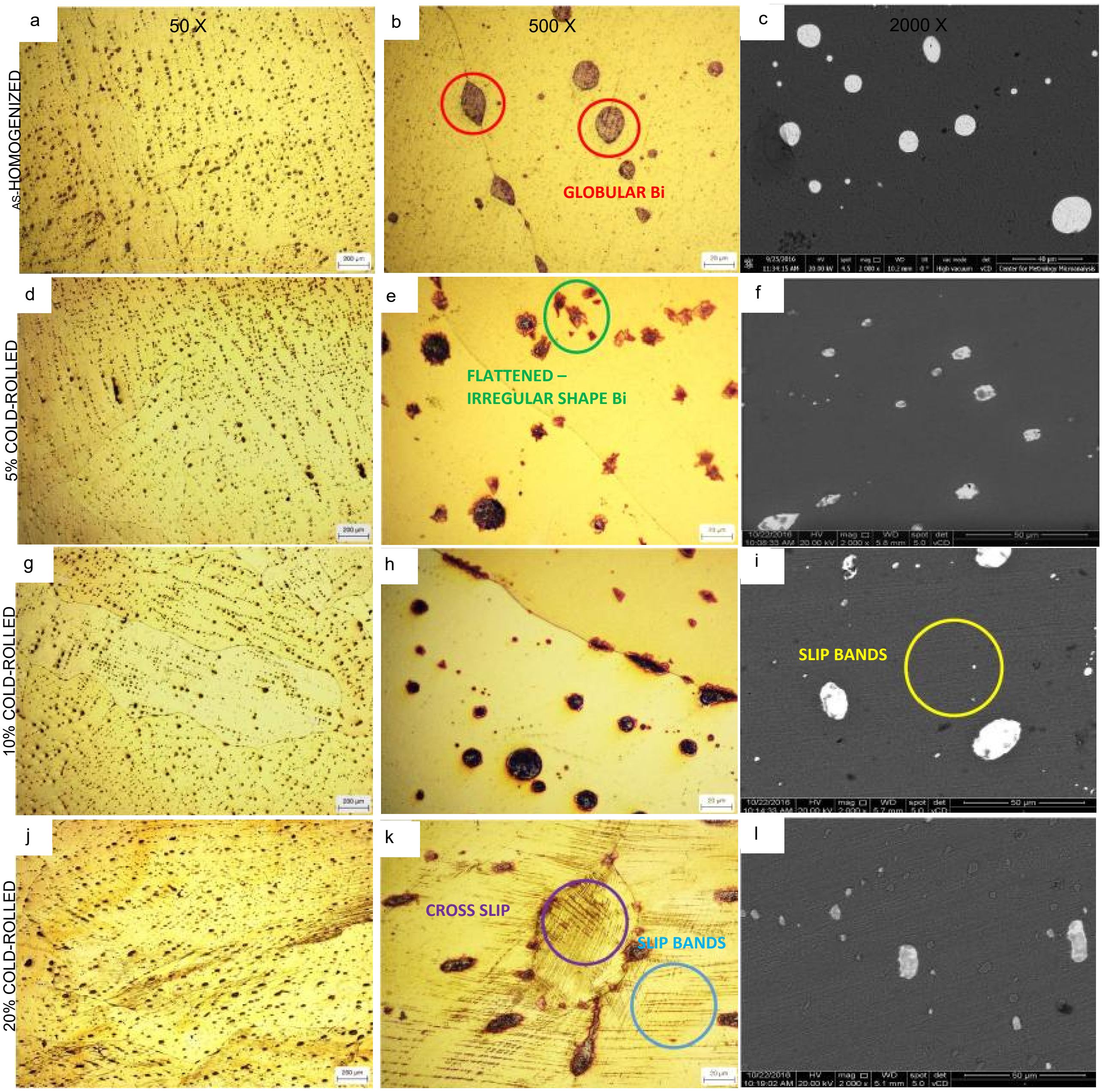

Figure 4. Evolution on the microstructure of Cu-28Zn-1.1Bi alloy in (a-c) as-homogenized state; and after cold rolling deformation at (d-f) 5 %, (g-i) 10 %, and (j-l) 20 %.

Evidences of deformation mechanisms were not visible at 5 % thickness reduction as observed in Figure 4 (d-f), suggestively due to the very low thickness reduction level applied. Bi shape transformation from globular to an irregular-flattened shape, however, is visible after one rolling pass. Rolling at 10 % thickness reduction resulted in slip bands formation in brass matrix, as appointed by the yellow circle area in Figure 4 (i). The slip bands were developed by dislocation movements due to the presence of applied strain, resulting in plastic deformation of the samples [9]. Figure 4 (j-l) shows the deformation microstructure of the alloys after cold rolling with 20 % thickness reduction. The amount of slip bands (shown in blue circle of Figure 4 (k)) in the samples increased remarkably with several appearances of cross slip, indicated by area within purple circle in Figure 4 (k). From these evidences it can be inferred that slip was the predominant deformation mechanism that developed at low level of applied strain as a result of dislocation movements in particular slip planes. While cross slip appeared as intersecting slip as a result of slip mechanism in more than one slip plane happening simultaneously inside a grain [9] as observed in Figure 4 (k). However, no mark of twinning was found in all experimental deformation level. Basori [1] reported a similar finding on microstructure observation of 20 % rolled Cu-29Zn-0.6Bi alloy, where no sign of twinning was observed. It is suggested that Bi segregation at grain boundaries and the presence of Bi dispersoid inhibited the formation of twin boundaries, as twinning is described as a uniform and simultaneous volume shifting inside a grain [6].

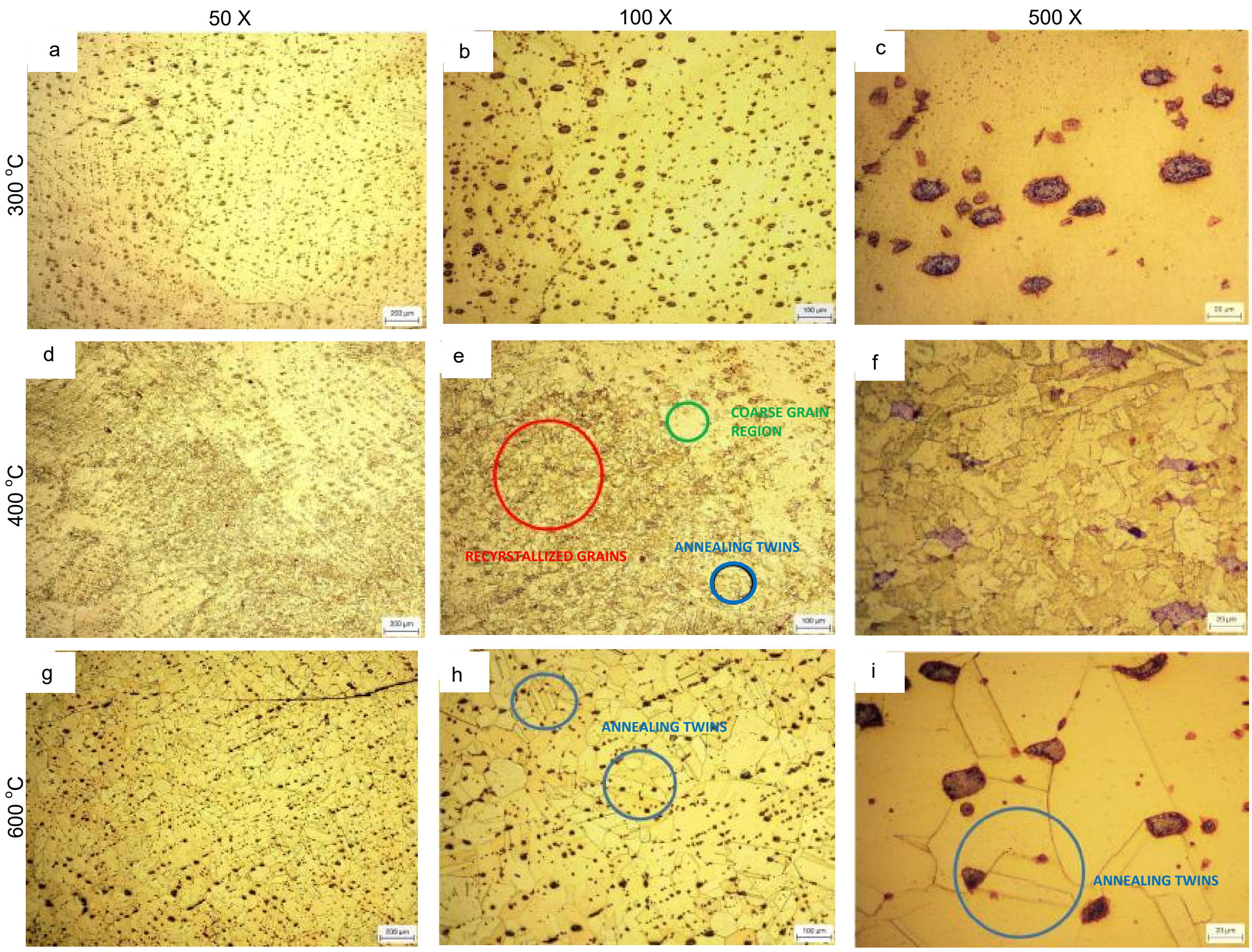

Figure 5. Microstructures of Cu-28Zn-1.1Bi with 20 % thickness reduction and subjected to annealing treatment for 30 min at (a-c) 300, (d-f) 400, and (g-i) 600 ^{o}C showing the occurrence of recrystallization and grain growth at 400 and 600 ^{o}C, respectively, with appearance of annealing twins.

Annealing Behavior. Figure 5 (a-c) shows little to none difference to the microstructure of 20 % rolled Cu-28Zn-1.1Bi alloy after annealing at 300 ^{o}C. Sofyan and Basori [7] reported that annealing treatment at 300 ^{o}C for 30 min subjected to 20 % rolled Cu-32Zn alloys aided the recovery and stress relieving process as the preliminary stage of recrystallization [9], but did not contribute to any microstructure evolution of the respected alloys.

A difference in the alloys microstructure was found after annealing at 400 ^{o}C as shown in Figure 5 (d-f), where the red circle shows recrystallized grains that could be clearly distinguished from the coarse grain region (shown by the green circle). Newly formed grains tended to nucleate at grain boundaries and areas surrounding the Bi dispersoid due to their high interface energy [8]. The grain nucleation around Bi dispersoid was stimulated by a *Particle Stimulated Nucleation* (PSN) mechanism, described as the appearance of *particle deformation zone* (PDS) – a disoriented sub-grain unit to its matrix resulted from stress between a large particle/dispersoid (> 1 µm) and alloys matrix. A high angle of disorientation will result in new grain nucleation in the respected area, owing to its high interface energy [8]. Additionally, annealing twins started to develop at this stage as shown in the blue circle area.

Further heating at 600 ^{o}C resulted in grain growth due to grain boundaries migration as illustrated in Figure 5 (g-i). During the grain boundaries migration, large grains grow continuously until smaller grains are subsided – known as *grain cannibalism* [9]. Annealing twins with sharp boundaries are observed as pointed by blue circle in Figure 5 (h-i).

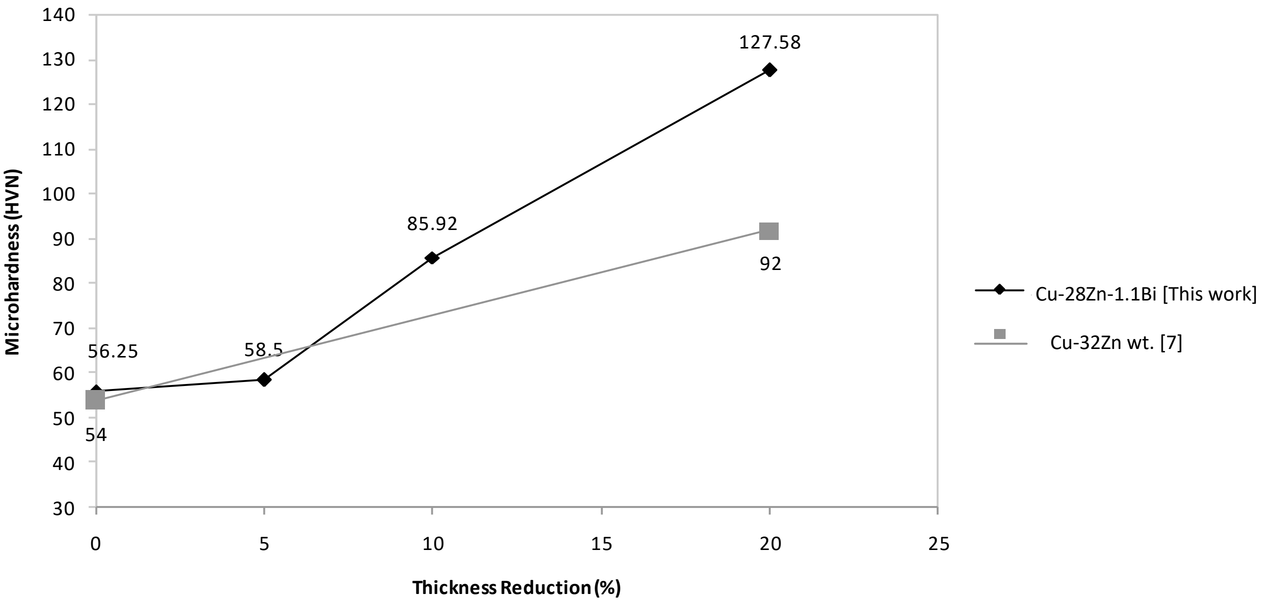

Figure 6. Increasing microhardness of Cu-28Zn-1.1Bi alloys simultaneous to higher thickness reduction level.

Microhardness. In the previous study by Basori [1], microhardness of as-homogenized and 20 % cold rolled Cu-29Zn-0.6Bi alloys were found to be 52.21 and 124.21 HVN, respectively, while produced Cu-28Zn-1.1Bi in this research possessed slightly greater microhardness of 56.25 and 127.58 HVN in as-homogenized and 20 % rolled state, respectively. It is indicated that higher Bi alloying content contributed in strengthening the alloys following a dispersoid strengthening mechanism – when particles of different crystallographic structure are present within the metal matrix, dislocation movements will be rendered [4].

Figure 6 shows an increasing trend of microhardness of Cu-28Zn-1.1Bi alloys along with higher deformation level from 56.25 to 58.5, 85.92, and 127.58 HVN at 5 %, 10 %, and 20 % thickness reduction. Higher deformation level resulted in escalated dislocation density that furthermore hardened the alloys – a strain hardening event [9].

Conventional decrease in microhardness of deformed alloys subjected to annealing was observed in both Cu-28Zn-1.1Bi [this work] and Cu-29Zn-0.6Bi alloys [8]. Recovery and stress relieving mechanisms occurred at 300 ^{o}C, resulting in a very slight decrease of alloys micro hardness. At 400 ^{o}C recrystallization was visible, with newly formed, stress-free equiaxed grain morphology as an outcome, replacing grains with high internal stress, hence re-activating the dislocation movements [9]. Thus, recrystallization resulted in decreasing alloys microhardness. A remarkable hardness decrease was observed at 600 ^{o}C. Full recrystallization had been achieved in this temperature, followed by a grain growth phenomenon. Hardly any traces of slip bands and cross slip deformation mechanism were found after annealing at 600 ^{o}C. Previous research by Sofyan and Basori [7] reported that recrystallization of Cu-32Zn wt. % alloys started at 500 ^{o}C, hence it can be inferred that Bi accelerated the recrystallization rate of brass alloys due to PSN mechanism as indicated in findings of this work.

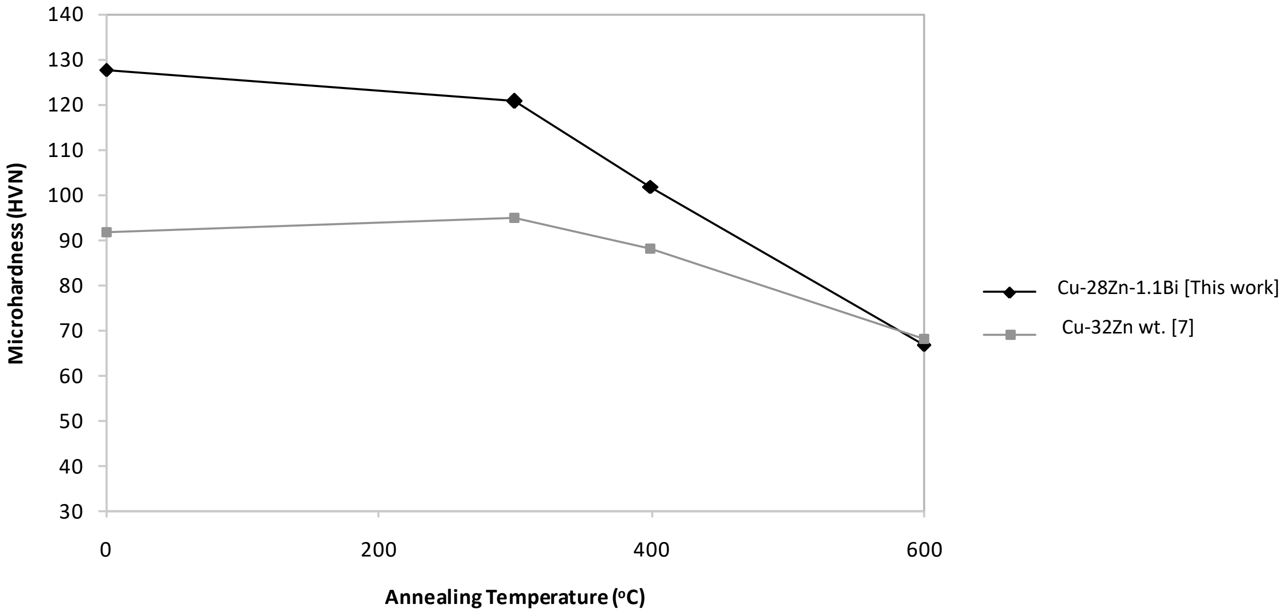

Figure 7. Effect of variations in annealing temperature to microhardness of Cu-28Zn-1.1Bi in this work and Cu-32Zn alloys[7] after 20 % thickness reduction.

Conclusion

In this work, the influence of cold rolling and annealing heat treatment on the characteristics of CuZnBi alloys were investigated. The main conclusions are summarized below"

1. Cold deformation resulted in flattened grain and Bi particles morphology with L/d ratios of Bi particles described as follows: 1.07, 1.32, and 2.64 at 5%, 10%, and 20% thickness reduction, respectively.
2. Slip was the predominant deformation mechanism of the experimental Cu-28Zn-1.1Bi alloys with evidence of slip bands and several cross slip visible in microstructure images of specimens of 10 % and 20 % thickness reduction.
3. An inclining trend in microhardness was observed along with higher thickness reduction level, where alloys micro hardness increased from 56.25 to 58.50, 85.92, and 127.58 HVN at 5 %, 10 %, and 20 % thickness reduction, respectively.
4. Bi contributed to alloys hardness through dispersoid strengthening mechanism in which Bi particles rendered the dislocation movements during cold deformation.
5. Recrystallization of deformed Cu-28Zn-1.1Bi wt. % alloys took place at 400 ^{o}C with nucleation sites concentrated at grain boundaries and areas around the large, insoluble Bi particles following a PSN mechanism which accelerated the recrystallization rate,. Grain growth phenomenon was observed after full recrystallization at 600 ^{o}C.

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