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Effect of thickness reduction on consolidation and crystallization of Cu based bulk metallic glass alloy during powder rolling in supercooled liquid region

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ABSTRACT

Cu based bulk metallic glass (BMG) strips were produced by a single pass of powder rolling of the metallic glass (MG) powder canned in a copper container. The effects of the thickness reduction on the consolidation and deformation of BMG alloys after powder rolling were examined. The BMG strips were produced successively by controlling the rolling reduction. Insufficient movement and deformation of MG particles during rolling with a reduction of ≤65% caused incomplete densification. In contrast, the excessive heat generated from the rolling deformation gave rise to the crystallization of the BMG strips leading to the formation of cracks during rolling with a reduction of ≥85%.

1. Introduction

Thin gauge strips of various bulk metallic glass (BMG) alloys are generally produced by a rapid solidification process (RSP), such as melt-spinning or direct casting (e.g. [1,2]). However, the direct production of thick strips of BMG alloys using RSP is still limited due to the insufficient glass forming ability (GFA) of BMG alloys. Various metallic glass (MG) powders having rather lower GFA can be produced by high pressure gas atomization in a clean closed system (e.g. [3]). Because MG alloys display the characteristic superplastic deformation behavior at temperatures in the supercooled liquid region (SLR), consolidation of the MG powder and subsequent rolling in this region can produce BMG sheets with various thicknesses [4,5].

Metal powders can be compacted continuously by powder rolling. The compacted green strips obtained by powder rolling undergo further processing by sintering and re-rolling to produce an end product in the final gauge. Sintering and compaction can also occur during powder rolling at elevated temperatures. BMG sheets were produced successfully by powder rolling in the SLR [6,7]. However, there are few reports of the consolidation and deformation of BMG alloys during powder rolling.

In this study, Cu–Ni–Zr–Ti BMG strips were produced by a single pass of powder rolling of MG powder canned in a copper container. Rolling was performed in the SLR of the BMG alloy by controlling the temperatures of the samples and roll surfaces. The effect of thickness reduction on the consolidation and crystallization of BMG alloys during powder rolling was examined by microstructure observations and structural characterization.

2. Experimental procedure

MG powder of $Cu_{54}Ni_6Zr_{22}Ti_{18}$ was atomized by a high pressure gas in a closed system [7]. MG powder with diameters ranging from 63 to 90 μm was selected by screening. Copper cans with an outer diameter of 36 mm and a thickness of 3 mm were filled with MG powder and evacuated to a pressure of 10^{-3} Pa at 470 K in order to minimize the oxidation. After evacuation, the samples were sealed and pre-compacted in the rolling machine. During pre-compaction, the outer thickness of the can decreased to 22 mm. The copper cans were then heated to the rolling temperature of 733 K and rolled by a single pass to a final outer thickness of 9.9, 7.7, 5.5 and 3.2 mm, corresponding to a thickness reduction of 55, 65, 75 and 85%, respectively. The working roll diameter was 400 mm and the temperature of roll surfaces was maintained at approximately 500 K in order to minimize the decrease in sample temperature during rolling.

Structural characterization was carried out by X-ray diffraction (XRD; Rigaku, RINT2200) with Cu Kα radiation. The consolidation and deformation of samples after rolling were observed by optical microscopy (OM). High resolution transmission electron microscopy (HRTEM, FEI Technei TM G2) was used for microstructural observations.

3. Results and discussion

The as-atomized MG powder of $Cu_{54}Ni_6Zr_{22}Ti_{18}$ used in this experiment had a supercooled liquid region from the glass transition temperature, $T_g = 712$ K, to the crystallization temperature, $T_x = 765$ K. As mentioned previously, the BMG strips were produced by a single pass of powder rolling of MG powders canned in a copper container. Fig. 1 shows optical micrographs observed

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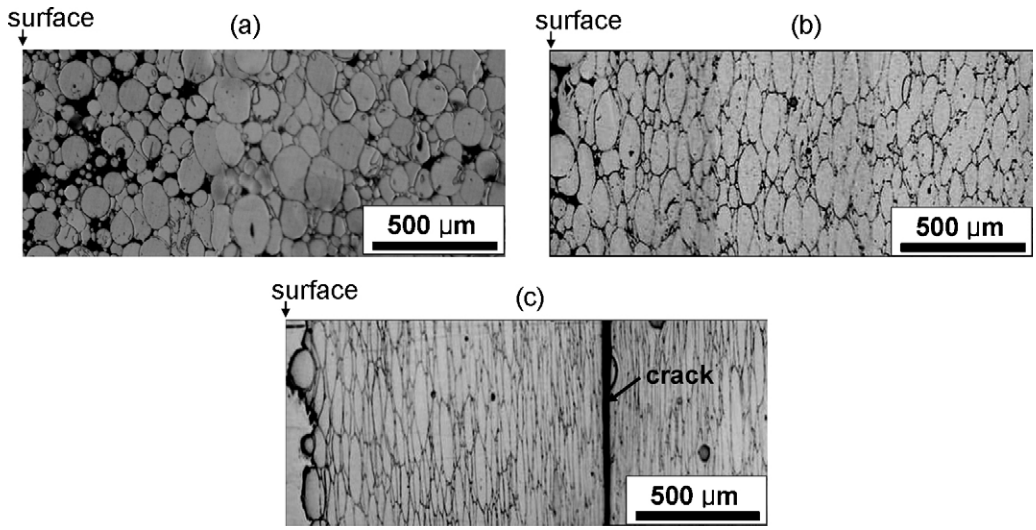

Fig. 1. Microstructure observed from the longitudinal section of the samples rolled to a thickness reduction of (a) 65%, (b) 75%, (c) 85%.

from the transverse direction of the rolled samples. The reduction in thickness indeed plays a dominant role in the evolution of microstructures during powder rolling. Rolling with a reduction of ≤65% caused an incomplete densification (Fig. 1(a)). In the present work, a thickness reduction of 75% provides a successful route for the production of BMG strips having a fairly homogeneous microstructure nearly free from pores (Fig. 1(b)). However, the sample rolled to 85% in one powder rolling pass displayed a quite inhomogeneous microstructure with some cracks (Fig. 1(c)).

With increasing rolling reduction, an increase in the density was observed. The relative density reached to 98% when the samples rolled to the reduction higher than 75%. A quite low fracture strength $\sigma_f < 1.4$ GPa was determined in the samples rolled to the reduction lower than 65%, which was attributed to an incomplete densification.

During powder rolling of MG powders canned in the copper container, simultaneous deformation of the MG particles and outer copper occurred. The change in thickness of the inner BMG layer and outer copper was measured and their corresponding strains were calculated, as shown in Fig. 2. The absolute thickness strain of the inner BMG layer was larger than that of the outer copper can, which was attributed to the densification of MG particles during powder rolling. Because a low imposed reduction mainly caused densification, a larger difference in thickness strain between the BMG sheet and copper was observed at a lower reduction.

Proper etching visualizes the changes in the MG particle shape by revealing the surface of the prior MG particles. Using this method, the deformation of BMG layers can be analyzed by observing the shape of the MG particles after powder rolling as shown in Fig. 1. Table 1 lists the aspect ratio (length/height) of the MG particles and the corresponding strain. The MG particles barely deformed during powder rolling with a thickness reduction of ≤65%. This suggests that the movement of MG particles occurs mainly for consolidation. In contrast, a thickness reduction of ≥75% resulted in the pronounced deformation of MG particles.

Table 1
Changes in the average aspect ratio and corresponding thickness strain of the metallic glass particles after powder rolling.

Reduction in thickness	Aspect ratio (length/height)	Corresponding thickness strain
55%	1.0	0.0
65%	1.2	−0.08
75%	3.06	−0.56
85%	8.0	−0.91

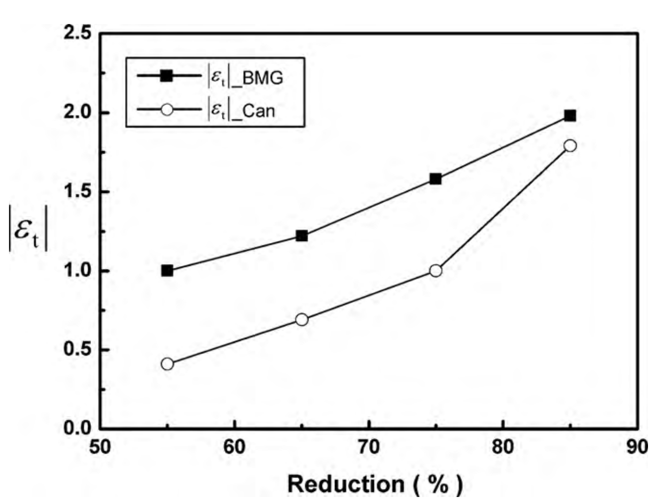

Fig. 2. Changes in thickness strain in the inner BMG layer and outer copper after powder rolling.

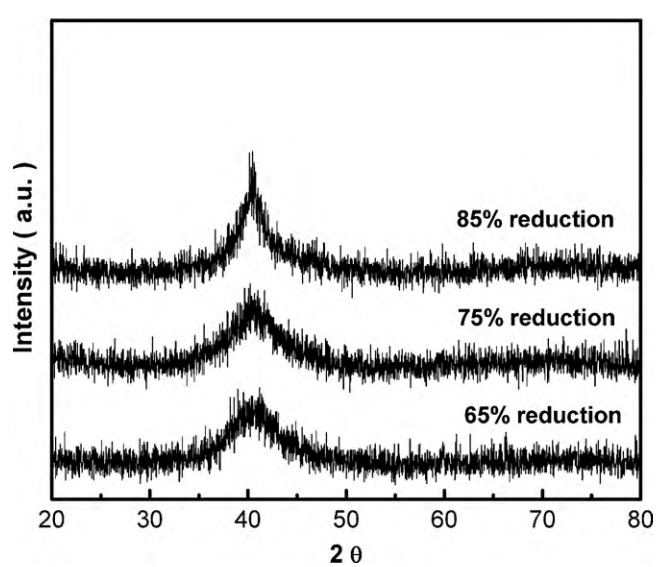

Fig. 3. XRD patterns of the rolled strips after powder rolling.

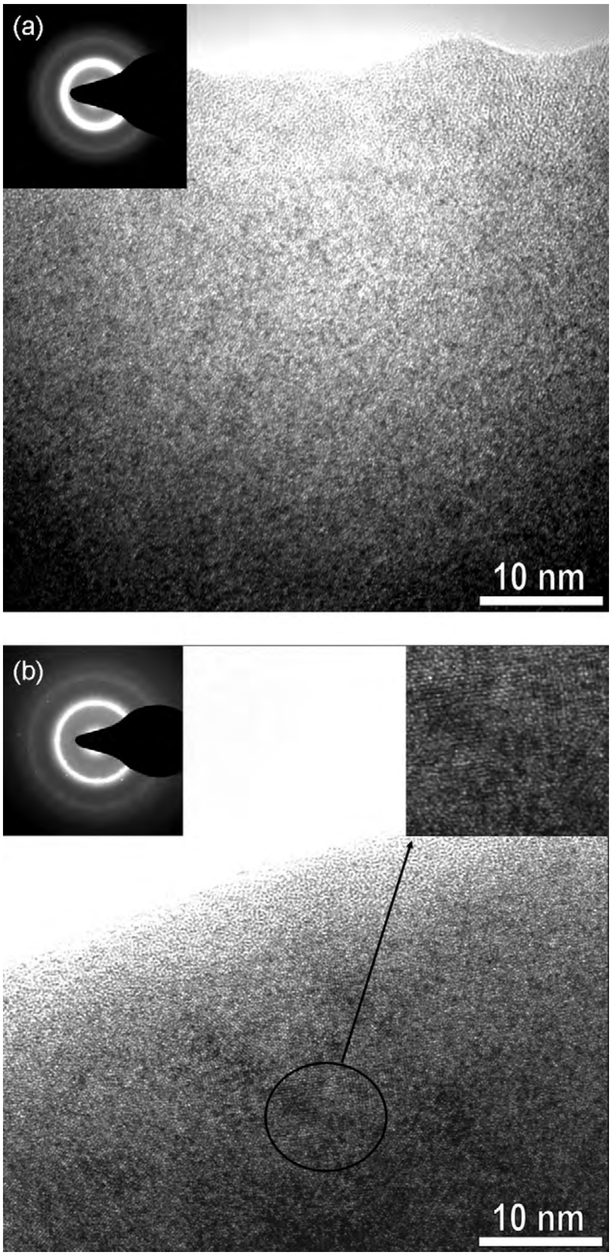

Fig. 4. High resolution TEM images and diffraction patterns of the sample rolled to a thickness reduction of (a) 75% and (b) 85%.

Fig. 3 shows the XRD patterns of the BMG strips after powder rolling. The XRD patterns of the samples rolled with a thickness reduction of ≤75% displayed a broad halo peak ranging from 33 to 47°, which is characteristic of an amorphous structure. However, the sample rolled to an 85% reduction showed crystalline peaks of $Cu_{51}Zr_{14}$, $Cu_{10}Zr_7$. HRTEM observations confirmed the results obtained from the XRD patterns. A fully amorphous structure without lattice fringes was observed from the samples produced with a reduction of ≤75%. In contrast, nano-crystallites were distributed homogeneously in the amorphous matrix were observed in the sample rolled to 85%, as shown in Fig. 4. The diffraction rings further confirmed the presence of nano-crystallites in this sample.

In order to understand the formation of nano-crystallites in the BMG sample produced by power rolling with an 85% reduction, the deformation work and corresponding increase in temperature ΔT were considered. If deformation is adiabatic, the increase in

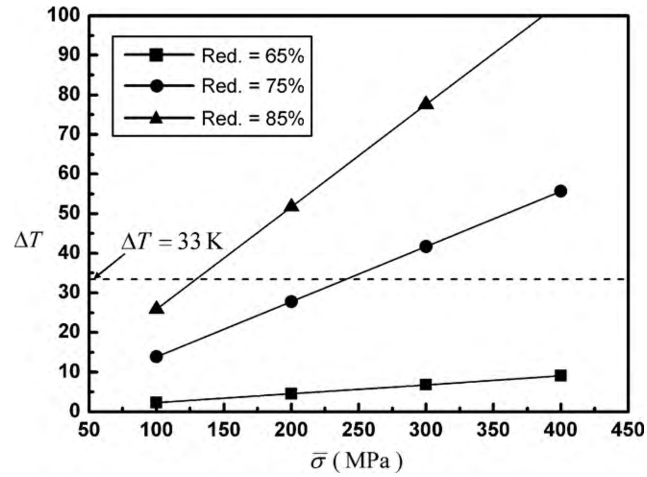

Fig. 5. Calculated increase in temperature as a function of the average flow stress.

temperature due to deformation can be approximated by Eq. [1].

$$\Delta T = \frac{\alpha \cdot \bar{\sigma} \cdot \bar{\varepsilon}}{\rho C} \quad (1)$$

where $\bar{\sigma}$ is the average flow stress, $\bar{\varepsilon}$ is the effective strain, ρ is the density, $C = 0.611$ J/g K is the mass heat capacity of the $Cu_{54}Ni_6Zr_{22}Ti_{18}$ alloy at 733 K in the SLR, and α is the fraction of energy stored [8]. In this study, an adiabatic system with $\alpha = 1$ was assumed because rolling was carried out with a high strain rate.

The strain and the average flow stress should be known in order to complete the temperature increase calculation. The strain observed in the MG particles was simply taken from the data in Table 1, which was calculated from the observed aspect ratios. Since rolling deformation can be approximated by plane strain, the effective strain of the rolling deformation is given by $\bar{\varepsilon} = 2/\sqrt{3}\,\varepsilon$ [9,10]. An evaluation of the exact value of $\bar{\sigma}$ is beyond the scope of this work because the strain and strain state of the rolled material vary from the entrance to the exit in a roll gap and the flow stress varies with the strain and strain rate [8,11]. Instead, several values of $\bar{\sigma}$ were assumed and the increase in temperature ΔT was calculated, as shown in Fig. 5.

In this work, powder rolling of the MG powder was carried out at 733 K and the crystallization temperature of the present MG alloy was $T_x = 765$ K, which shows that a $\Delta T > 33$ K leads to the onset of crystallization. As mentioned above, powder rolling with an 85% reduction resulted in the formation of crystalline phases, while no crystalline phases developed during powder rolling with a reduction of ≤75%. Fig. 5 shows that powder rolling of the MG alloy with an average flow stress of $\bar{\sigma} > 250$ MPa leads to crystallization in both samples rolled to 75 and 85%. In contrast, ΔT is less than 33 K even in the sample rolled to an 85% reduction when $\bar{\sigma} < 100$ MPa is assumed. Accordingly, Fig. 5 reveals that an average flow stress $\bar{\sigma}$ ranging from 150 to 225 MPa prevailed during powder rolling of the present MG alloy.

4. Conclusion

The effect of a thickness reduction on the consolidation and crystallization of Cu–Ni–Zr–Ti metallic glass (MG) powder was examined during powder rolling. Bulk metallic glass strips were produced successively by proper control of the rolling reduction. Insufficient movement and deformation of MG particles during rolling with a reduction of ≤65% caused incomplete densification, while the heat generated from the deformation work promoted the crystallization of BMG leading to the formation of cracks during rolling with a reduction of ≥85%.

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