Journal of Alloys and Compounds 504S (2010) S271–S274

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jallcom

Production of Zr-based bulk metallic glass thin strips by means of sandwich rolling in the supercooled liquid region

H.J. Kim [a], J.C. Bae [a], E.S. Park [b], M.Y. Huh [b,*], D.H. Kim [c]

[a] Department of Materials Science and Engineering, Korea University, Seoul 136-701, Republic of Korea
[b] Eco-Materials and Processing Department, Korea Institute of Industrial Technology, Inchon 406-130, Republic of Korea
[c] Center for Non-Crystalline Materials, Department of Metallurgical Engineering, Yonsei University, Seoul 120-749, Republic of Korea

ARTICLE INFO

Article history:
Received 4 July 2009
Received in revised form 21 January 2010
Accepted 6 April 2010
Available online 14 April 2010

Keywords:
Bulk metallic glass
Sandwich rolling
Finite element method

ABSTRACT

In order to produce thin gauge bulk metallic glass (BMG) sheets, thick Zr-based BMG plates were sandwiched between upper and lower outer copper sheets and then warm-rolled jointly in the supercooled liquid region. Proper control of the outer sheet thickness during sandwich rolling provided successful rolling of the BMG sheets. The finite element method simulation disclosed that sandwich rolling with thick outer sheets minimized the shear strains leading to a strain state close to the plane strain in the rolled BMG sample.

1. Introduction

Bulk metallic glass (BMG) strips in the form of an intermediate-shaped material have been used in the manufacture of various final products [1,2]. However, the application of BMG strips is still very limited due to the lack of processes for the mass production of BMG strips. Since BMG alloys display the characteristic superplastic deformation behavior in the supercooled liquid region (SLR), which is defined by the temperature range between glass transition (T_g) and crystallization temperature (T_x), rolling of BMG alloys can be utilized for the production of BMG strips with various thicknesses [3,4].

Previously, BMG sheets were produced successfully by powder rolling of amorphous powder in the SLR [5,6]. Strict control of the process variables, such as the reduction per a rolling pass, heating rate and rolling temperature, during powder rolling was required for the production of BMG sheets. Improper control of the temperature of BMG samples gives rise to the crystallization of the BMG leading to the catastrophic fracture of BMG samples during rolling. Accordingly, powder rolling of amorphous powder is a difficult and expensive process for commercialization.

In this study, a thick BMG plate was first produced by vacuum centrifugal casting. In order to obtain homogeneous deformation of BMG during rolling, the as-cast BMG plate was sandwiched between the outer copper sheets and then jointly warm-rolled in the supercooled liquid region. The effect of the outer copper sheet on the evolution of strain states in the sandwiched samples was studied by finite element method (FEM) simulations.

2. Experimental procedure

A bulk metallic glass (BMG) plate of $Zr_{41.2}Ti_{13.8}Cu_{21.5}Ni_{10}Be_{22.5}$ with a thickness of 3.0 mm was first fabricated using the vacuum centrifugal casting method. The BMG plate was then sandwiched between upper and lower outer copper sheets with a thickness of either 0.2 or 0.5 mm. The size of the sample was approximately 25 mm × 60 mm (width × length). The sandwiched composite plates were heated to the rolling temperature in the supercooled liquid region (SLR) at a heating rate of 80 K/min in a box furnace and then warm-rolled jointly by a single pass without holding. The rolling temperature of 700 K was chosen to prevent the inhomogeneous deformation and crystallization. Fig. 1 shows an illustration of the sandwich rolling process. The working roll diameter was 300 mm and the roll speed was 20 rpm, giving a linear velocity of the roll of 314 mm/s. The structural characterization was carried out by the X-ray diffraction (XRD) and the deformation behavior of the BMG sheets after rolling was observed by optical microscopy (OM).

The two-dimensional finite element method (FEM) simulation was carried out using a commercial package DEFORM-2D to interpret the evolution of strain states in the sandwiched samples during rolling. During the FEM calculation, the friction between the roll and outer copper sheets was assumed by the friction coefficient of $\mu = 0.25$ and the interface between the copper can and BMG sheet was assumed to be inseparable. For details on the FEM simulation scheme regarding the rolling deformation of composite samples, please refer to [7,8].

3. Results and discussion

For warm rolling, the composite sample was heated to 700 K in a box furnace at a heating rate of 80 K/min. At this heating rate, the differential scanning calorimetry (DSC) curve was measured and a

---

* Corresponding author. Tel.: +82 2 3290 3271; fax: +82 2 928 3584.
E-mail address: myhuh@korea.ac.kr (M.Y. Huh).

0925-8388/$ – see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.jallcom.2010.04.035

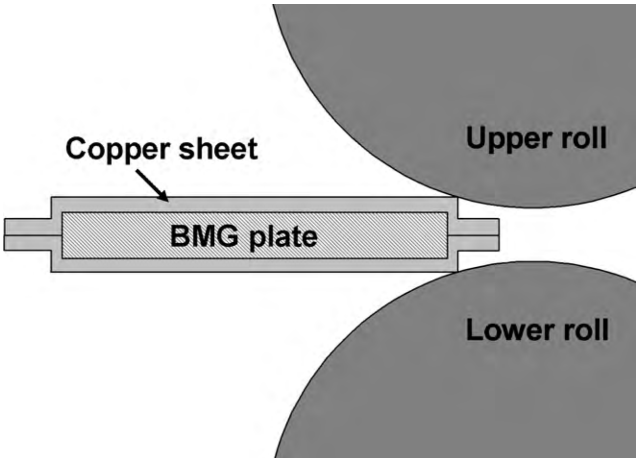

Fig. 1. Schematic diagram of sandwich rolling.

glass transition temperature (T_g) and crystallization temperature (T_x) of 665 and 755 K, respectively, were confirmed.

Three samples were prepared: one BMG sample without outer copper sheets, and two composite samples comprising an inner BMG plate and outer copper sheets with a thickness of either 0.2 or 0.5 mm. The samples were warm-rolled by a single pass to a thickness reduction of 30%, corresponding to a logarithmic strain of $\varepsilon = 0.35$. Fig. 2 shows the macrostructures of a longitudinal section of the samples after warm rolling. The outer copper sheet played an important role in the evolution of strain states in the inner rolled BMG sheet and resulting macrostructures.

Warm rolling of a BMG sheet without and with the 0.2 mm thick outer copper sheets led to the formation of cracks, as shown in Figs. 2(a) and (b), respectively. Vain pattern was observed in fracture surfaces formed along cracks. Interestingly, cracks developed in two directions in the BMG sample rolled without the outer sheets, while cracks formed in only one direction in the inner BMG sample rolled with the thin outer copper sheets. Warm rolling of the composite comprising an inner BMG plate and 0.5 mm thick outer

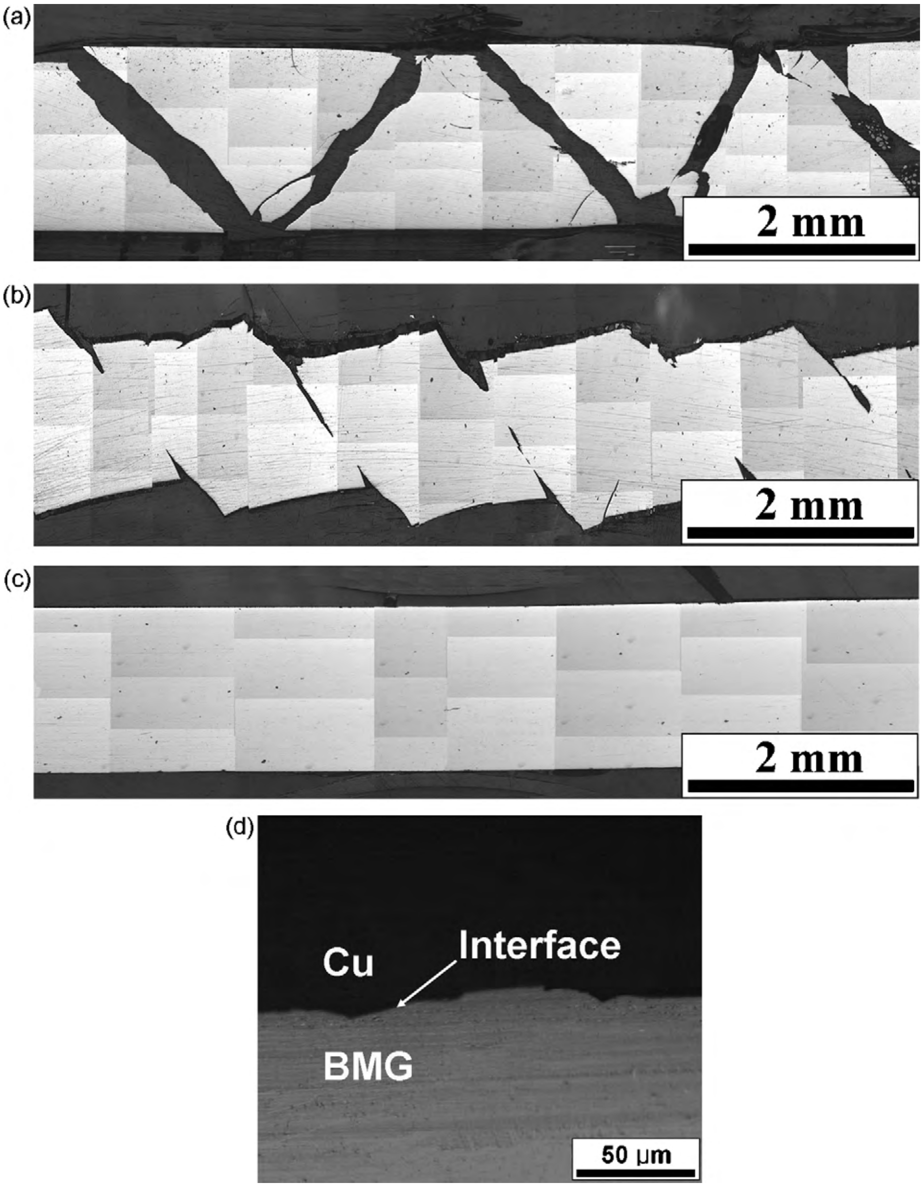

Fig. 2. Macrostructures of a longitudinal section after rolling. BMG samples (a) without the outer sheets, and sandwiched between the outer sheets with a thickness of (b) 0.2 mm, (c) 0.5 mm, (d) magnified view of the interface between BMG and outer copper sheet in (c).

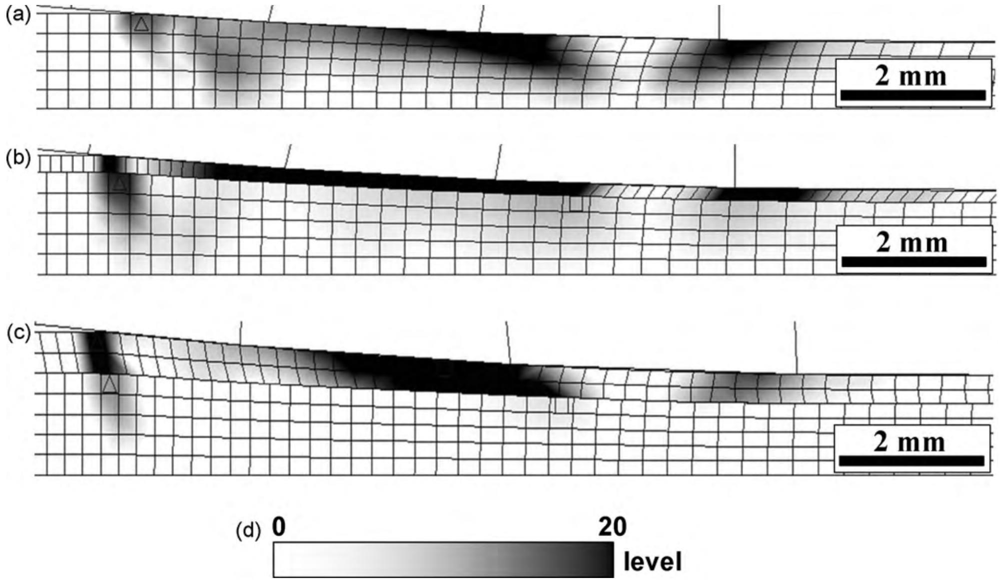

Fig. 3. Distributions of $\dot{\varepsilon}_{13}$ calculated by the FEM simulations. BMG samples (a) without the outer sheets, and sandwiched between the outer sheets with a thickness of (b) 0.2 mm and (c) 0.5 mm.

copper sheets gave rise to the successful rolling deformation of the BMG plate, as shown in Fig. 2(c). Additional experiments showed that the use of more than 0.5 mm thick outer sheets always allowed successful rolling of the inner BMG plate.

A series of finite element method (FEM) simulations with the commercial FEM package DEFORM$^{\text{TM}}$-2D was carried out to determine the role of the outer sheets. The entire set-up (Fig. 1) was modeled with bilinear 4-node plane strain elements. Friction at the various contact surfaces was incorporated using Coulomb's friction law with the friction coefficients μ. The normal strain per a rolling pass was $\varepsilon_{11} = 0.35$ and the time of the sample in the roll gap was $t \approx 0.04\,s$ which gives the mean applied strain rate of $\dot{\varepsilon}_{11} \approx 10^1\ \text{s}^{-1}$ during rolling. Since the stress-strain relationships of rolled materials at this strain rate were required for FEM calculations, compression tests in a Gleeble tester were carried out and experimental data were fed into the FEM simulation [9].

To a very rough approximation of a macroscopic sheet, normal rolling deformation is the plane strain compression for which $\dot{\varepsilon}_{11} = -\dot{\varepsilon}_{33}$ and all other strain rates $\dot{\varepsilon}_{22} = \dot{\varepsilon}_{12} = \dot{\varepsilon}_{13} = \dot{\varepsilon}_{23} = 0$. Here, the rolling direction (RD), transverse direction (TD) and normal direction (ND) of a sheet are identified with directions 1, 2 and 3, respectively. However, shear deformation $\dot{\varepsilon}_{13}$ occurs in the thickness layers away from the center plane. Rolling with a large rolling draught and high friction causes large variations in $\dot{\varepsilon}_{13}$ along a given streamline in the roll gap [10–12].

Fig. 3 shows the distribution of $\dot{\varepsilon}_{13}$ in the three samples examined in this experiment. In the BMG sample rolled without outer copper sheets, the variation of $\dot{\varepsilon}_{13}$ increases when approaching the surface layer in contact with the roll surface, while the component $\dot{\varepsilon}_{13}$ barely develops at the center layer. Therefore, the shear strain rate $\dot{\varepsilon}_{13}$ shows strong gradients in the BMG sample rolled without the outer sheets. There were significantly smaller variations in $\dot{\varepsilon}_{13}$ in the inner BMG sample after rolling with the outer sheets, as shown in Fig. 3(b) and (c). The operation of shear strain rate $\dot{\varepsilon}_{13}$ was almost negligible in the BMG sample rolled with the thick outer sheets.

The macroscopic shear strain rate $\dot{\varepsilon}_{13}$ is composed of the two components $\dot{e}_{13}$ and $\dot{e}_{31}$ of the displacement rate gradient $\dot{e}_{ij}$ according to $\dot{\varepsilon}_{13} = 0.5(\dot{e}_{13} + \dot{e}_{31})$. In brief, the geometrical shape changes of an element in the roll gap lead to the evolution of the shear rate $\dot{e}_{31}$, while the friction between the roll and sheet surface accounts for the shear rate $\dot{e}_{13}$ [13,14]. Both components $\dot{e}_{13}$ and $\dot{e}_{31}$ vary considerably along a given streamline in the roll gap and also depend on the through-thickness layer.

Fig. 4 shows the shape changes of a square element before and after the operation of the displacement gradients of $\dot{e}_{13}$ and $\dot{e}_{31}$ in a roll gap. Obviously, the operation of each shear displacement $\dot{e}_{13}$ and $\dot{e}_{31}$ gives the shear deformation in different directions.

Fig. 5 shows the variations of the displacement rate gradients $\dot{e}_{13}$ and $\dot{e}_{31}$ along the quarter thickness layer of the inner BMG sample. As indicated in Fig. 3, the variations of $\dot{e}_{13}$ and $\dot{e}_{31}$ depends strongly on the configuration of the samples examined in this work. The BMG sample rolled without the outer sheets displays large variations of $\dot{e}_{13}$ and $\dot{e}_{31}$ comprising a large negative maximum of $\dot{e}_{31}$ and a large positive maximum of $\dot{e}_{13}$ (Fig. 5(a)). The operation of two large shear components leads to two strong localized strain gradients in the roll gap, which can lead to the formation of cracks in two directions in this sample (Fig. 2(a)). As shown in Fig. 5(c), the inner BMG sample rolled with the thick outer sheets was rolled successively without the formation of cracks because a plane strain state with only small $\dot{e}_{13}$ and $\dot{e}_{31}$ values prevailed throughout the entire sample. Fig. 5(b) shows that the BMG sample rolled with the

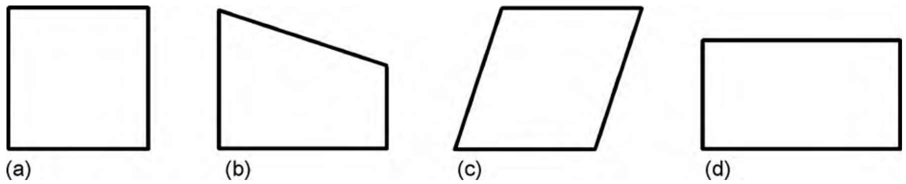

Fig. 4. Shape changes of elements before and after operation of shear displacement gradients in a roll gap. (a) Before entry of rolls, (b) after operation of $\dot{e}_{31}$, (c) after operation of $\dot{e}_{13}$, and (d) after exit of rolls.

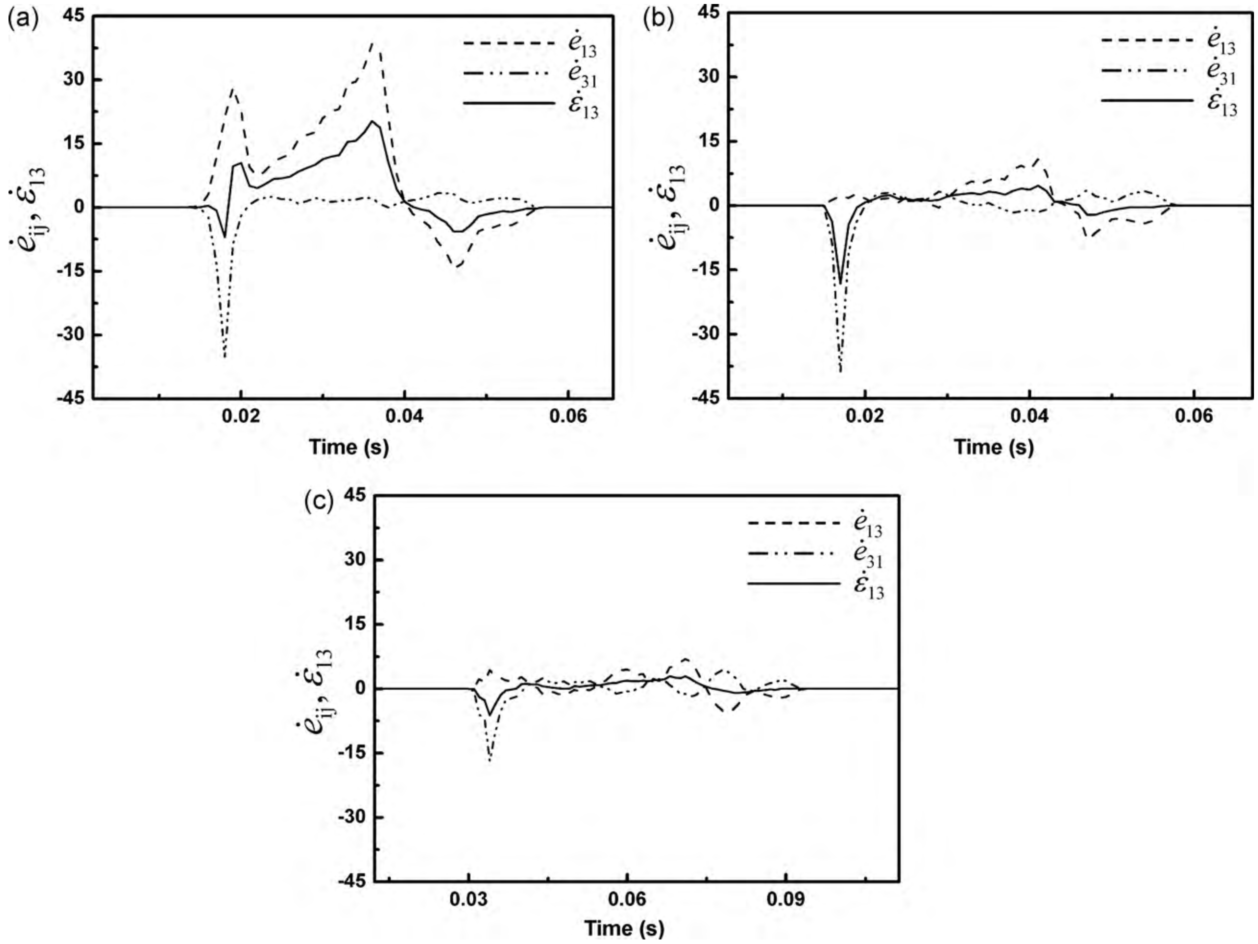

Fig. 5. Variations of $\dot{e}_{13}$ and $\dot{e}_{31}$ along the quarter thickness layer of the inner BMG samples (a) without the outer sheets, and sandwiched between the outer sheets with a thickness of (b) 0.2 mm, and (c) 0.5 mm.

thin outer sheets displayed the operation of only one large shear component $\dot{e}_{31}$ in the roll gap, which may give rise to the formation cracks in one direction in this sample (Fig. 2(b)). Accordingly, the formation of cracks in the rolled BMG was attributed to the evolution of the shear strain gradients of $\dot{e}_{13}$ and $\dot{e}_{31}$ in the roll gap.

4. Conclusion

In order to produce thin gauge BMG sheets, thick BMG plates were sandwiched between upper and lower outer copper sheets and then warm-rolled jointly in the supercooled liquid region. Warm rolling of the BMG plate with thick outer copper sheets allowed the successful rolling deformation of the BMG plate. The finite element method simulation disclosed that sandwich rolling with thick outer sheets minimized the shear strains leading to a strain state close to the plane strain in the rolled BMG sample.

References

[1] A. Inoue, Stabilization of metallic supercooled liquid and bulk amorphous, Acta Mater. 48 (2000) 279–306.
[2] J. Jayaraj, Y.C. Kim, H.K. Seok, K.B. Kim, E. Fleury, Mater. Sci. Eng. A 449–451 (2007) 30–33.
[3] J. Lu, G. Ravichandran, W.L. Johnson, Acta Mater. 51 (2003) 3429–3443.
[4] Y. Kawamura, T. Shibata, A. Inoue, T. Masumoto, Appl. Phys. Lett. 69 (1996) 1208–1210.
[5] H.J. Kim, K.W. Lee, J.K. Lee, T.S. Kim, J.C. Bae, E.S. Park, M.Y. Huh, D.H. Kim, Mater. Sci. Eng. A 449–451 (2007) 929–933.
[6] M.H. Lee, J.S. Park, J.H. Kim, W.T. Kim, D.H. Kim, Mater. Lett. 59 (2005) 1042–1045.
[7] M.Y. Huh, K.R. Lee, O. Engler, Int. J. Plast. 20 (2004) 1183–1197.
[8] H.G. Kang, J.K. Kim, M.Y. Huh, O. Engler, Mater. Sci. Eng. A 452 (2007) 347–358.
[9] E.S. Park, M.Y. Huh, H.J. Kim, J.C. Bae, High strain rate response of Zr-based bulk metallic glass in supercooled liquid region, Intermetallics (2010), doi:10.1016/j.intermet.2010.02.040.
[10] G.E. Dieter, Mechanical Metallurgy, 3rd ed., McGraw-Hill, 1986.
[11] J.J. Nah, H.G. Kang, M.Y. Huh, O. Engler, Scripta Mater. 58 (2008) 500–503.
[12] H.G. Kang, M.Y. Huh, S.H. Park, O. Engler, Steel Res. Int. 79 (2008) 489–496.
[13] O. Engler, M.Y. Huh, C.N. Tomé, Metall. Mater. Trans. A 31 (2000) 2299–2315.
[14] W.F. Hosford, R.M. Caddell, Metal forming Mechanics and metallurgy, 2nd ed., Prentice-Hall, Michigan, 1972.