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Micro-lateral extrusion of $Zr_{55}Cu_{30}Al_{10}Ni_5$ bulk metallic glass under low-frequency vibration loading

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Abstract. The effect of vibration on the micro-forming ability of $Zr_{55}Cu_{30}Al_{10}Ni_5$ bulk metallic glass in its supercooled liquid region was studied. The experiment of micro-extrusion was carried out under different amplitude (38 ∼ 760 N) and different frequency (0.1 ∼ 2.0 Hz) at a fixed temperature of 723 K. The extrusion length was taken as a measure to characterize the micro-forming ability. Results reveal that the extrusion length of bulk metallic glass is effectively improved under vibration loading, and increases with increasing loading frequency and amplitude, whereas the frequency dependence is stronger. The viscosity of bulk metallic glass declines under vibration loading because of a larger free volume concentration and surface effect caused by vibration. This research indicates that the vibration forming is an effective method to enhance the micro-forming ability of bulk metallic glasses.

1. Introduction

Bulk metallic glasses (BMGs) are multi-component alloys with attractive mechanical properties such as high strength, high hardness and corrosion resistance at room temperature. BMGs exhibit low viscosity and good formability when reheated into the supercooled liquid region (SCLR) [1, 2], the temperature region where the glass relaxes into a metastable liquid before it eventually crystallizes. Benefiting from the superplasticity of a BMG in its SCLR, it can be used for thermoplastic forming. To date, this unique processing opportunity has been used for a wide range of applications, including micro- and nano-forming, surface replication, extrusion and micro-hot embossing [3]. All of these works exhibit potential application of BMGs in the miniaturization of modern industry [4, 5].

It is noted that most of the previous studies are focusing on choosing optimum processing parameters such as temperature and processing time to get the highest formability of the BMG former in its supercooled liquid region. However, these two processing parameters are interactional. At higher temperature, the viscosity of BMG is significantly reduced but, at the same time, the processing time is shortened because a risk of possible crystallization may occur in such forming conditions [3]. Previous studies showed that vibration can be applied in metal micro-forming to increase forming ability through

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the reduction of forming load and reduction of the friction at the interface between die and workpiece [6]. Jinpin Qu [7] studied the influence of polymer melt flowing under vibrating field by introduced electromagnetic vibration and mechanical vibration into the amorphous polymer forming, found that the vibration can significantly reduce the apparent viscosity of the polymer melt, and then deducing the constitutive relation of the apparent viscosity of polymer melt under vibrating field according to the "Free Volume" and "Dynamic Behaviour of Polymer Chains" theory. Goshawk and Thien [8, 9] studied the flow behaviour of non-Newtonian fluid in the tube under vibration field, concluded that vibration can effectively reduce the viscosity of the fluid. Based on the fact that amorphous alloys and polymers have some similar characteristics, we can introduce vibration loading as a new method to improve the micro-forming ability of amorphous alloys. Very few studies on micro-forming of BMGs under vibration field have been reported so far.

The objective of this paper is to investigate the micro-forming ability of $Zr_{55}Cu_{30}Al_{10}Ni_5$ BMG in its supercooled liquid region under a vibration field. Lateral extrusion experiment was carried out under vibration loading, and compared with those under a constant loading in addition as the original sample to demonstrate the efficiency of this method. Particular attention is given to the variation of extrude length of this alloy that means the BMG micro-forming ability, under different amplitude, different frequency. Finally, the physical mechanism of the phenomenon is rationalized in terms of the free volume theory, material loss modulus and surface effect.

2. Experiment

$Zr_{55}Cu_{30}Al_{10}Ni_5$ BMG was selected for this study due to its high glass-forming ability as well as its wide supercooled liquid region. BMG rods of 2 mm in diameter were prepared by copper mould casting in vacuum. The glassy structure of the as-received rod was confirmed using X-ray diffractometry. Differential scanning calorimetry (DSC) study of the as-received rod revealed that the temperatures at the onset of the glass transition (Tg) and at the onset of the crystallization (Tx) are 693 K and 763 K, respectively when subject to a heating rate of 20 K/min. So the temperature in this experiment was chosen at the supercooled liquid region of $693 \sim 763$ K. Then, the BMG rods were cut into billets of 4 mm in length and both ends of the specimens were carefully polished to ensure parallelism. In this study, lateral extrusion experiment was selected as the extrude length can be a standard to characterize the micro-forming ability of BMG in the SCLR with vibration loading. Figure 1 shows the schematic illustration of dies with a semi-circular channel of 1 mm in diameter. In order to determine the extruded length under different vibration parameters, the experiments were carried out on a Zwick Z020 machine equipped with an environmental chamber, at a fixed temperature of 723 K and the forming time was set to 5 min, which is shorter than the period at the onset of the crystallization at the temperature of 723 K. The BMG specimens were subjected to oscillatory force applied on the punch in a sinusoidal mode with loading frequencies ranging from 0.1 to 2.0 Hz and force amplitude ranging from 38 to 760 N. The average force is $\sim$1900 N, which calculated from flow stress by $\sigma = 3\eta\dot{\varepsilon}$, where η is the viscosity from the previous work [4] and $\dot{\varepsilon}$ is the max strain-rate can be gained according to the equivalent volume principle.

The lateral extrusion experiment operation started from setting the temperature of air furnace at 723 K. Loading mode of the machine was cyclic compression. Then put the BMG specimen in the die with proper graphite lubricant in addition. The heating of furnace was stopped when the temperature approximately increases up to the set temperature and the assembled die was placed with a fixture on the machine and then closed the furnace. Continue heating to the set temperature and maintain the temperature for 30 seconds, then take compression for 5 min. After the vibration forming process, stop heating and the die was removed from the furnace and the specimen was separated from the die through a pushing process by the punch after air-cooling.

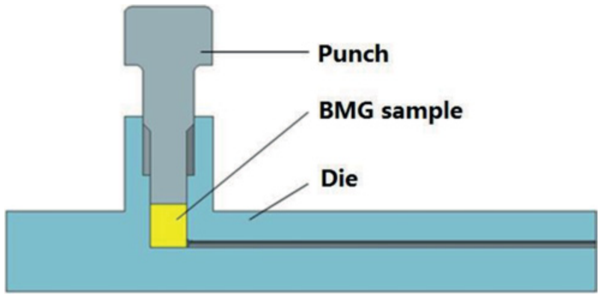

Figure 1. Schematic illustration of lateral extrusion dies.

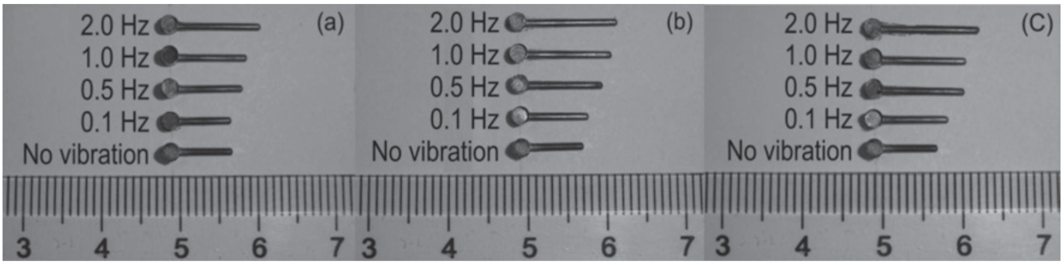

Figure 2. Photographs of the $Zr_{55}Cu_{30}Al_{10}Ni_5$ BMG after vibration forming under various loading frequency ranging from 0.1 to 2.0 Hz and the same force amplitude: (a) 38N; (b) 190N; (c) 380N.

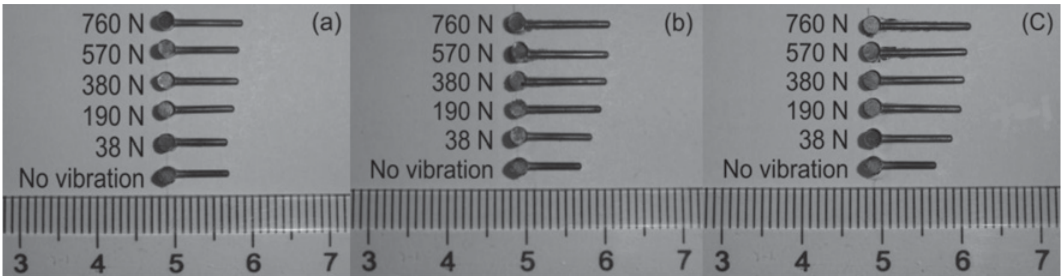

Figure 3. Photographs of the $Zr_{55}Cu_{30}Al_{10}Ni_5$ BMG after vibration forming under various force amplitude ranging from 38 to 760 N and the same loading frequency: (a) 0.1 Hz; (b) 0.5 Hz; (c) 1.0 Hz.

3. Results and discussion

Experimental results show that vibration can improve the micro-forming ability of Zr55 BMG. Figure 2 exhibits experimental Zr55 BMG specimens with various loading force amplitude of 38 N, 190 N, 380 N, and frequencies ranging from 0.1 to 2.0 Hz. It can be seen that the extrude length of Zr55 BMG increases significantly with the increasing loading frequencies under a certain force amplitude. The corresponding extrude length-frequency curves are shown in Fig. 4a), at the force amplitude of 38 N, the extrude length is ~7.3 mm at the loading frequency of 0.1 Hz, while it increases up to 11.1 mm when the loading frequency increase to 2.0 Hz. Similarly, the extrude length increases up to 51.9% at the amplitude of 190 N and 41.2% at that of 380 N from 0.1 Hz to 2.0 Hz. It is obviously from the Fig. 4a) that the extrude length almost shows linear increasing tendency with the increment of the loading frequency at the force amplitude of 190 N and 380 N.

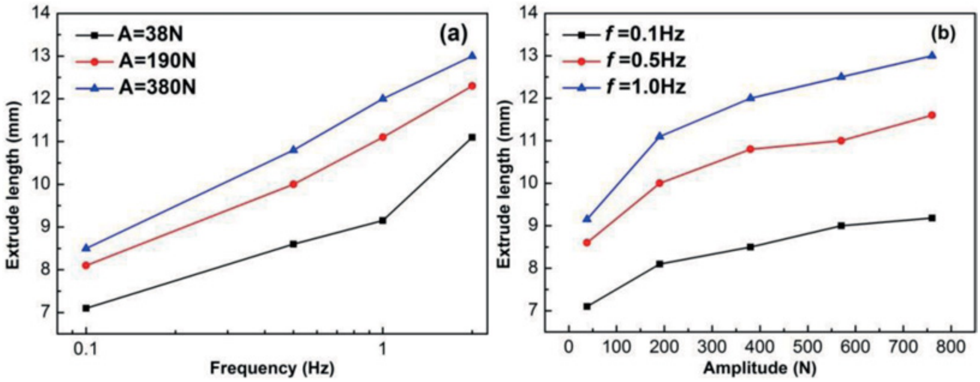

Figure 4. The extrude length-frequency curves (a) and the extrude length-amplitude curves (b) under vibration field.

However, as described in Fig. 3 and Fig. 4b), the extrude length increasing trend with the force amplitude is not as obvious as that with the loading frequency at the frequency of 0.1, 0.5 and 1.0 Hz; especially in the amplitude range 190 to 760 N, the extrude lengths are not distinctly changing. It is indicated that the micro-forming ability of $Zr_{55}Cu_{30}Al_{10}Ni_5$ in the SCLR significantly improved under vibration field compared with the sample with no vibration, and the frequency dependence is stronger than amplitude dependence.

The above experimental results indicate that the extrude length increases with the increasing frequency, which means the viscosity of Zr55 BMG decline and the micro-forming ability increases in the SCLR. In fact, when applying sinusoidal stress on viscoelastic material as: $\sigma = \sigma_0 \sin \omega t$, there will be a phase difference (δ) between the observed strain and the stress, which can be expressed as follows: $\varepsilon = \varepsilon_0 \sin(\omega t + \delta)$. Accordingly, the dynamic modulus can be described as: $E = E' + iE''$, where E' is storage modulus that represents the elastic part and E'' is loss modulus that refers to viscosity part. At lower loading frequency, most deformation is contributed by viscous flow, which causes irreversible energy loss. However when the frequency is increased, the time of viscous flowing is very short, that means there is no time to rearrange for atomic structure. The elastic deformation, most of elastic energy is recoverable, will become dominant in the whole period of deformation. So, the energy loss will be reduced with the increasing loading frequency, this leads the dynamic viscosity greatly decline and enhance the micro-forming ability of BMGs.

According to the free volume theory, the plastic deformation of bulk metallic glass is determined by the transition of partial single atom as like atomic diffusion. Spaepen [10] represented that in order to move or diffuse, the atom has to push the other peripheral atoms to obtain free volume. Evolution equation of the relationship between the flow rate and the free volume was constructed, indicating that as the free volume concentration increases, the atomic flowing will become distinctly easy, the flow rata will also increase and therefore viscosity will decline with that. Li N [11] had made uniaxial tension and compression experiment of $Zr_{35}Ti_{30}Be_{26.75}Cu_{8.25}$ BMG in the SCLR under vibration field, revealing that the concentration of free volume increases with increasing frequency, which reduces flow viscosity and improve the micro-forming ability.

In vibration plastic deformation, the direction of friction force at the tool-workpiece interface reverses at every cycle of vibration, this "surface effect" changes the contact pressure characteristics at the interface and decreases friction [6]. Thus, the plastic deformation force of the material is reduced and the forming efficiency of metal is improved.

It is interesting to observe that the increasing trend of extrude length with increasing frequency and amplitude are different, the length increases more slowly with increasing amplitude, indicating that the influence mechanism of frequency dependence and amplitude dependence may not be the same, but what the underlying physical origin is for this difference are remain unanswered, which needs further experimental and theoretical studies.

4. Conclusions

Vibration load, as an effective method, was introduced to improve the micro-forming ability of $Zr_{55}Cu_{30}Al_{10}Ni_5$ BMG in the supercooled liquid region. The results of experiment showed that the extrude length increases, which means flow viscosity of BMGs was reduced by vibration. It was revealed that the micro-forming ability of bulk metallic glasses in the SCLR is effectively improved under vibration loading, and increased with increasing loading frequency and amplitude. But the frequency dependence is stronger. Under vibration load, dynamic viscosity of BMGs in the SCLR decreases by the change of loss modulus of the viscoelastic material. Free volume theory indicates that the concentration of free volume increases with increasing frequency, which reduces the material flow viscosity and enhances therefore the micro-forming ability of BMGs.

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