# Preparation of Zr<sub>52.8</sub>Cu<sub>29.1</sub>Ni<sub>7.3</sub>Al<sub>9.8</sub>Y<sub>1</sub> bulk metallic glass by hereditary process

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**Abstract:** Zr-based bulk metallic glass possesses the highest potential as a structural material among metallic glasses. However, its potential application has been restricted by a number of issues, such as fragility, small size and difficult fabrication into different shapes. In this paper, an attempt is made to evaluate the possibility of preparing a solid  $Zr_{52.8}Cu_{29.1}Ni_{7.3}Al_{9.8}Y_1$  bulk metallic glass by using binary precursors. It is found that the GFA and the stability of  $Zr_{52.8}Cu_{29.1}Ni_{7.3}Al_{9.8}Y_1$  bulk metallic glasses prepared in the hereditary process increase with the increasing quenching temperature, while the supercooled liquid region  $\Delta T_x$  increase from 55 K to 83 K. The flexural strength increases to 1823 MPa at a quenching temperature of 1723 K.

#### Introduction

In recent decades various research groups have discovered a wide range of bulk metallic glasses. Some are metal-metal alloys based on elements such as zirconium, copper, iron and nickel; others combine metals with near metalloids such as silicon, carbon and phosphorus [1-4]. Some metallic glasses are attractive for their magnetic properties; iron-boron glass, for example, is commonly used in distribution transformer cores [4]. Specifically, BMGs exhibit a rare and tantalizing combination of traits: their amorphous, defect-free microstructure makes them one of the strongest engineering materials known, and because they can occupy a peculiar thermodynamic middle ground between solid and liquid, they can be processed like plastics into nanoscale textures, seamless hollow containers, and other shapes that are impossible to make with traditional metals [3,4].

Moreover, some changes in composition can lead to large changes in properties. Several empirical parameter with glass-forming ability, but only some of these correlations are useful for predicting universal glass-forming ability. These rules still require knowledge of properties of the alloy to allow prediction of glass-forming ability. Models that only require information about the constituent elements as inputs remain a challenge [5].

As a consequence, only a minute fraction of potential BMG-forming compositions have been explored thus far. To investigate the vast remaining phase space and properties, more efficient techniques and methods are required [6-8]. While the studies have been focusing on the discussions about the inhomogeneous flow, its relation in structural disorder and mechanical property [9-12].

The objective of this research is to explore the preparing a  $Zr_{52.8}Cu_{29.1}Ni_{7.3}Al_{9.8}Y_1$  bulk metallic glass using binary precursors and evaluate the aim BMG properties. This work demonstrates that a  $Zr_{52.8}Cu_{29.1}Ni_{7.3}Al_{9.8}Y_1$  bulk metallic glass preparing from binary precursors has an excellent glass-forming ability and flexural strength.

### **Experimental**

The Zr<sub>52.8</sub>Cu<sub>29.1</sub>Ni<sub>7.3</sub>Al<sub>9.8</sub>Y<sub>1</sub> metallic glass was selected as the objective in the current study. The Zr<sub>41</sub>Cu<sub>59</sub>, Zr<sub>36</sub>Ni<sub>64</sub>, Zr<sub>73</sub>Al<sub>27</sub> and Al<sub>88</sub>Y<sub>12</sub> alloys were selected as the precursors for the Zr<sub>52.8</sub>Cu<sub>29.1</sub>Ni<sub>7.3</sub>Al<sub>9.8</sub>Y<sub>1</sub> metallic glass. The idea is that these binary alloys have relatively low melting points that are comparable to that of the metallic glass. All alloys were prepared by the medium-frequency induction furnace melting a mixture of pure metals and die casting into a copper mold with 3 mm thickness. The hereditary processes of preparing metallic glass was used and the GFA and flexure strength were analyzed.

The hereditary process of preparing metallic glass was shown in Fig. 1, high purity Zr (99.0 wt.%), Cu (99.99 wt.%), Ni (99.99 wt.%), Al (99.99 wt.%), Y (99.0 wt.%) metal were mixed according to the binary alloy ( $Zr_{41}Cu_{59}$ ,  $Zr_{36}Ni_{64}$ ,  $Zr_{73}Al_{27}$  and  $Al_{88}Y_{12}$ ) components ratio and melted by using the medium-frequency induction furnace , and then die casted into a copper mold. Next, the binary alloys got from quenching were mixed and melted again as precursors to prepare the aim metallic glass.

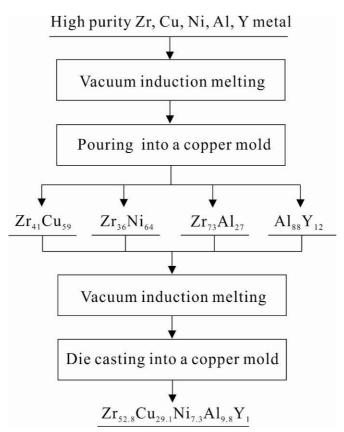


Fig. 1. The hereditary process of preparing Zr<sub>52.8</sub>Cu<sub>29.1</sub>Ni<sub>7.3</sub>Al<sub>9.8</sub>Y<sub>1</sub> bulk metallic glass using precursor

The specimens prepared were analyzed by X-ray diffraction (XRD) (X' Pert Pro, PANalytical Corporation, Netherland) with Cu K $\alpha$  radiation. Differential scanning calorimetry (DSC) was performed using a TG-DSC (SDTQ600, TA Instruments, America) in an argon atmosphere with the heating rates of 20 K/s and the sample mass of  $20\pm1$  mg. The flexure strength was measured by using all-powerful material test machine (AG-X 100kN, SHIMADZU Corporation, Japan).

#### **Results and discussion**

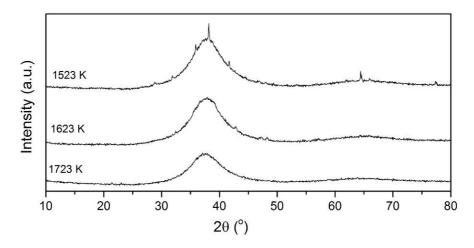


Fig. 2. X-ray diffraction patterns of metallic glasses quenching at different temperatures

Fig. 2 shows the XRD patterns of samples casted at different quenching temperatures. It can be seen that the XRD patterns consist of a broad diffuse peak between diffraction angles  $30^{\circ}$  and  $45^{\circ}$ . The sample quenching at 1523 K shows some little shape peak which indicates some crystalline phase mixed with the primary amorphous structures. When the quenching temperature is up to 1623 K, there is no apparent crystalline phase corresponding to the sharp crystallization peak, indicating that this sample is in the amorphous structures. As the quenching temperature is 1623 K, though its pattern shows broad diffuse backgrounds, but the amorphous diffuse peak are shape, the amorphous diffuse peak of 1623 K is sharper than that of 1723 K, showing that it has the trend of further crystallization. Thus, the threshold overheating temperature for fully amorphous structure of  $Zr_{52.8}Cu_{29.1}Ni_{7.3}Al_{9.8}Y_1$  bulk metallic glass is at least 1623 K, below which it may have an intersection with the crystallization position.

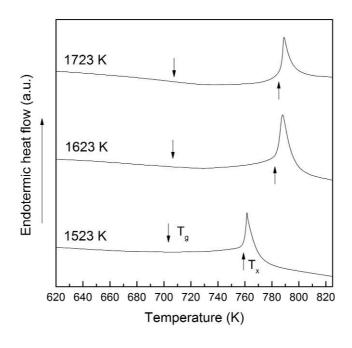


Fig. 3. DSC curves of metallic glasses quenching at different temperatures

The DSC curves of  $Zr_{52.8}Cu_{29.1}Ni_{7.3}Al_{9.8}Y_1$  bulk metallic glass prepared by hereditary process are shown in Fig. 3. All samples show an endothermic event, which is characteristic of glass transition. For the purpose of comparison, the specific temperatures of the three kinds of alloys quenching at different temperatures are listed in Table 1.

As shown in Table 1, the specific temperature  $T_x$  and  $\Delta T_x$  of the samples with lower quenching temperature clearly decline, which means that the short-term thermal stability of these samples is improved by using hereditary process. The hereditary process in the content range investigated does not evidently influence the basic form of the DSC curves at different quenching temperature, however, the specific temperature  $T_x$  and  $\Delta T_x$  of the samples at high quenching temperature evidently rises, which means the short-term thermal stability of the metallic glass is enhanced by overheating [13-15].

Table 1 Thermodynamic parameters of metallic glasses quenching at different temperatures

Sample No.	Temperature (K)	$T_{g}(K)$	$T_{x}(K)$	$\Delta T_{\mathrm{x}}\left(\mathrm{K}\right)$
1	1523	703	758	55
2	1623	704	784	80
3	1723	703	786	83

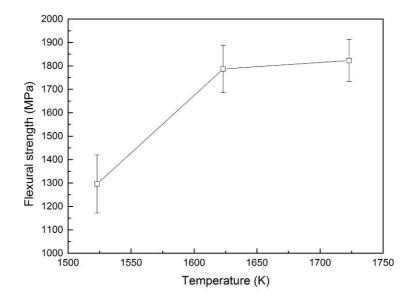


Fig. 4. The flexural strength dependence of quenching temperatures for Zr<sub>52.8</sub>Cu<sub>29.1</sub>Ni<sub>7.3</sub>Al<sub>9.8</sub>Y<sub>1</sub> samples

Fig. 4 shows compressive fracture strength dependence of quenching temperatures Zr<sub>52.8</sub>Cu<sub>29.1</sub>Ni<sub>7.3</sub>Al<sub>9.8</sub>Y<sub>1</sub> samples with 3 mm thickness, 12.45 mm width and 100.2 mm length. It is found that the flexural strength increases with the increasing quenching temperature. When the quenching temperature is up the threshold overheating temperature 1623 K, the flexural strength have no obvious change. The flexural strength can reach to 1823 MPa at a quenching temperature of 1723 K.

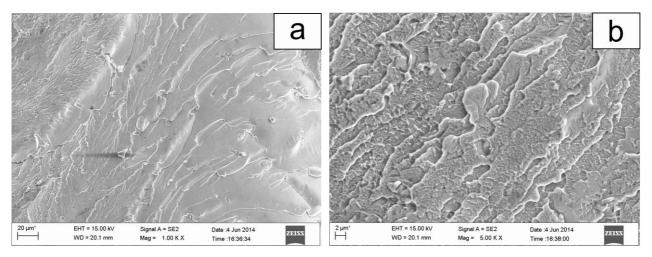


Fig. 5. Fracture morphologies of Zr<sub>52.8</sub>Cu<sub>29.1</sub>Ni<sub>7.3</sub>Al<sub>9.8</sub>Y<sub>1</sub> sample at a 1623 K quenching temperature

As can be seen that the typical fracture morphology of full bulk amorphous alloy is shown in Figs. 5(a) and (b), which is a typical characteristic of fracture feature with well-developed vein patterns. The local melting and the softened alloy, which look like liquid droplets, can also be observed on the fracture surface. And there are some little area, irregular smooth featureless zones distributing in Fig. 5(a). This is consistent with the excellent strength at the quenching temperature of 1623 K.

Based on aforementioned results, it is clear that mechanical properties of  $Zr_{52.8}Cu_{29.1}Ni_{7.3}Al_{9.8}Y_1$  metallic glass to a large extent is related to TRT (thermal rate treatment) process [16]. While the hereditary process also has obvious influence on mechanical properties.

## Conclusion

The  $Zr_{52.8}Cu_{29.1}Ni_{7.3}Al_{9.8}Y_1$  bulk metallic glass prepared in hereditary process shows excellent GFA and flexural strength. The glass-forming ability and flexural strength improve with the increasing quenching temperature. When the quenching temperature is up the threshold overheating temperature, the supercooled liquid region  $\Delta T_x$  increase from 55 K to 83 K, and the flexural strength can reach to 1823 MPa at a quenching temperature of 1723 K.

#### References

- [1] W.H. Wang, Roles of minor additions in formation and properties of bulk metallic glasses, Prog. Mate. Sci. 52 (2007) 540-596.
- [2] W.L. Johnson, K. Samwer, Auniverasl criterion for plastic yielding of metallic glasses with a  $(T/T_g)$  Phys. Rev. Lett. 95 (2005), 195501-195506.
- [3] C.J. Byrne and M. Eldrup, Bulk metallic glasses, Science 321 (2008), 502-503.
- [4] M. Chen, A brief overview of bulk metallic glasses, NPG Asia Mater. 3 (2011) 82-90.
- [5] J. Schroers, The superplastic forming of bulk metallic glasses, J. Metals 57 (2005), 35-39.
- [6] J. Schroers, Bulk metallic glasses, Physics today 66 (2013) 32-37.
- [7] P.W. Anderson, Through the glass lightly, Science 267 (1995) 1615-1616.
- [8] E.M. Axinte and M.P.I. Chirileanu, Recent progress in the industrialization of metallic glasses, Recent Patents on Materials Science 5 (2012) 213-221.
- [9] B. Idzikowski, P. Švec, M. Miglierini, Properties and Applications of Nanocrystalline Alloys from Amorphous Presursors, Kluwer Academic Publishers, U.S.A., 2003.

- [10] F. Spaepen, Homogeneous flow of metallic glasses: a free volume perspective, Scripta Mater. 54 (2006) 363-367.
- [11] W.H. Wang, Metallic glasses: family traits, Nat. Mater. 11 (2012) 275-276.
- [12] S. Torquato, Glass transition: hard knock for thermodynamics, Nature 405 (2000) 521-523.
- [13] Z.J. Yan, J.F. Li, S.R. He, H.H. Wang, Y.H. Zhou, Effect of repeated melting of the ingots on the glass-forming ability of Zr-based alloys, Meter. Lett 57 (2003) 2829-2833.
- [14] S.G. Zhang, M.X. Xia, G.H. Hu, J.G. Li, Discrepancy of structural stability against temperature between metallic liquids and their pglases, J.Non-cryst. Solids 356 (2010) 2223-2227.
- [15] Y.C. Zhao, S.Z. Kou, H.L. Suo, R.J. Wang, Y.T. Ding, Overheating effects on thermal stability and mechanical properties of Cu36Zr48Ag8Al8 bulk metallic glass, Meter. Design 31 (2010) 1029-1032.
- [16] G.Q. Liu, S.Z. Kou, C.Y. Li, Y.C. Zhao, H.L. Suo, Effect of TRT process on GFA, mechanical properties and microstructure of Zr-based bulk metallic glass, Prog. Nat. Sci. Vol. 21 (2011), 53-58.