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Bulk metallic glass tube casting

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ABSTRACT

Tubular bulk metallic glass specimens were produced, using a custom-built combined arc-melting tilt-casting furnace. Zr55Cu30Al10Ni5 tubes with outer diameter of 25 mm and 0.8–3 mm wall thicknesses were cast, with both tilt and suction casting to ensure mold filling. Tilt casting was found to fill one side of the tube mold first, with the rest of the tube circumference filled subsequently by suction casting. Optimized casting parameters were required to fully fill the mold and ensure glass formation. Too small melt mass and too low arc power filled the mold only partially. However, too large melt mass and higher arc power which lead to the best mold filling also lead to partial crystallization. Variations in processing parameters were explored, until a glassy ring with 1.8 mm thickness was produced. Different sections of the as-cast ring were investigated by X-ray diffraction (XRD), differential scanning calorimetry (DSC), and instrumented indentation to ensure amorphous microstructure. Atomic force microscopy (AFM) was used to compare the surface qualities of the first- and last-filled sections. These measurements confirmed the glassy structure of the cast ring, and that, the tilt cast tube section consistently showed better surface quality than the suction cast section. Optimized casting parameters are required to fully realize the potential of directly manufacturing complex shapes out of high-purity bulk metallic glasses by tilt casting.

1. Introduction

Metallic glasses are amorphous metals that solidified without detectable crystallization. Upon heating from the solid state these alloys exhibit a glass transition, after which they remain metastable for a finite length of time in the super-cooled liquid region, before crystallizing. For some alloys, rapid quenching is not necessary to avoid crystallization. If the diameter of the glass is larger than 1 mm, it can be called a bulk glass. Enhanced stability against crystallization is usually achieved by alloying multiple elements with significant difference, greater than 12%, in atomic radius and negative heats of mixing among constituent elements. The critical casting diameters of known BMG alloys typically range from 1 mm to 100 mm. BMG alloys have been found in many different alloy groups (Pd-, Mg-, Ln-, Zr-, Ti-, Fe-, Co-, Ni- and Cu-based systems) and new alloys have been discovered and reported with a variety of different properties. By casting BMG alloys, without cold working or heat treatment, complex shapes can be produced with excellent mechanical properties: purely elastic deformation up to a yield strain of typically 2%, resulting in tensile or compressive strength of 1500–5500 MPa, with Young's modulus of 70–275 MPa [1]. The lack of grain boundaries in the BMG materials also results in very accurate surface finish and enhances corrosion resistance. Several recent reviews testify to the widespread interest in these materials both from a fundamental science perspective and for practical applications [2–4].

For practical applications, the ease of forming complex shapes out of metallic glass is particularly important, but most reports in the academic literature focus on simple shapes achievable with simple molds. Recently a lot of attention has been focused on the use of thermoplastic forming processes or "visco-forming" for manufacturing metallic glass products from cast preforms [5]. Thermoplastic forming allows to form complex shapes that would be practically impossible to produce directly by casting, but does not obviate the need for casting complex shapes. Rather, they are complementary processes, wherein the preform produced by casting provides the right amount of material in the right starting position for the thermoplastic forming step. For example, blow-molding a bottle starts with a parison in the shape of a tube closed at one end [6]. The casting of annular or tubular specimens, in molds requiring a core, allows to investigate some of the issues arising when bulk metallic glasses are cast in complex molds. Such shapes are also relevant for potential practical applications such as annular gaskets, active solder materials [7], jewelry, and enclosures for electronic components. Metallic glass tubes can be used to construct precise Coriolis mass flowmeters [8]. Bulk metallic glass rings or tubes have previously been manufactured by centrifugal casting [9], machining [10], modified water quenching [7], copper mold

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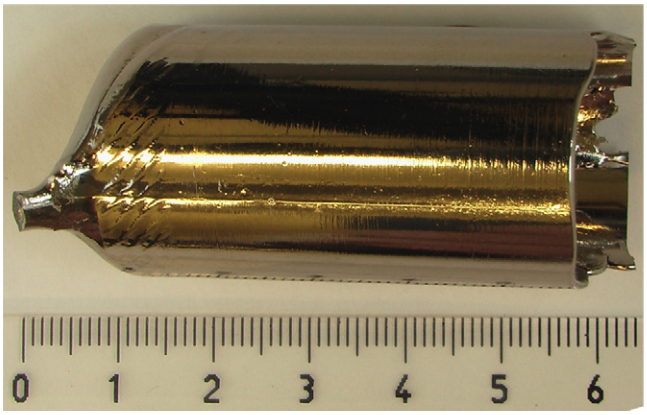

Fig. 1. A fully filled casting.

casting [11–13] and coreless mold suction casting [8,14]. Centrifugal casting [9] and suction casting [8,14] were done without a core. Modified water quenching [7] used a fused silica core. The copper mold casting [12,13] used vertical argon pressure ejection of induction-heated melt from a quartz crucible into the ring mold with minimal vertical runner length. MingZhen et al. [11] used a more traditional lip pour ladle with both vertical and horizontal runners before the mold with a copper core.

The published methods for bulk metallic glass tube and ring casting use two furnaces: one for alloy production and another for casting. Often the melt is cooled only from one side, possibly limiting the achievable tube wall thickness. When a core is used, the melt size has been very large, e.g., 780 g in [11], or the length of the ring was the smallest dimension [12,13]. When temporary molds are used [7], the molten alloy comes in contact with fused silica, which may cause embrittlement [15].

Here we study the possibility of casting bulk metallic glass tube into a core mold in one process. With a custom-built furnace, we start from raw materials and break the vacuum only once the tube has been cast. With the same arc melting electrode used in both alloy production and casting, we aim to keep the ingot size under 100 g and waste as little as possible of the melt charge on runners using tilt-casting to avoid the need for vertical runners (Figs. 1 and 2).

2. Experimental procedures

Alloy constituents for the target composition of $Zr_{55}Cu_{30}Al_{10}Ni_5$ (in at.%) were weighed from the elements with purity (in mass%) of 99.99% for Zr(+Hf), 99.999% for Cu, 99.99% for Ni, and 99.99% for Al. The cut and weighed pieces were melted in a custom built combined arc melting and tilt casting furnace under reduced pressure titanium gettered argon atmosphere. The vacuum-chamber was evacuated to a vacuum of better than 5×10^4 Pa before filling the chamber with 99.9999% purity argon. Any remaining oxygen in the chamber was reduced by titanium gettering. To ensure an even distribution of the alloying elements, the ingot was melted, observed while it was allowed to crystallize, flipped over and remelted repeatedly, until the crystallization process appeared to follow a uniform and repeatable pattern over several remelting cycles. Once the ingot was deemed mixed, it was melted again for casting. The suction casting line valve was opened and the furnace tilted to tilt cast the melt into a cylindrical mold located at the edge of the copper hearth. The furnace is designed so that these steps can be carried out without opening the chamber to air between the first melting and the final casting, maintaining the high-purity atmosphere throughout the entire process.

In our casting set-up, schematically shown in Fig. 3, filling of the mold begins as the chamber is tilted, when the melt flows over the hump separating the hearth and the mold orifice. The melting arc is still powered during the tilting, keeping the rest of the melt molten as it flows over the hump. However, until the chamber has finished tilting, the melt pours into one side of the mold first. The chamber tilting rate depends on the constriction caused by a pneumatic valve feeding the two pneumatic actuators that tilt the chamber. For best results, the valve is fully opened and tilting takes less than 2.5 s. A small longitudinal draft is used in the core; the diameter of the core increases towards the bottom of the mold to facilitate withdrawing the core from the cast tube.

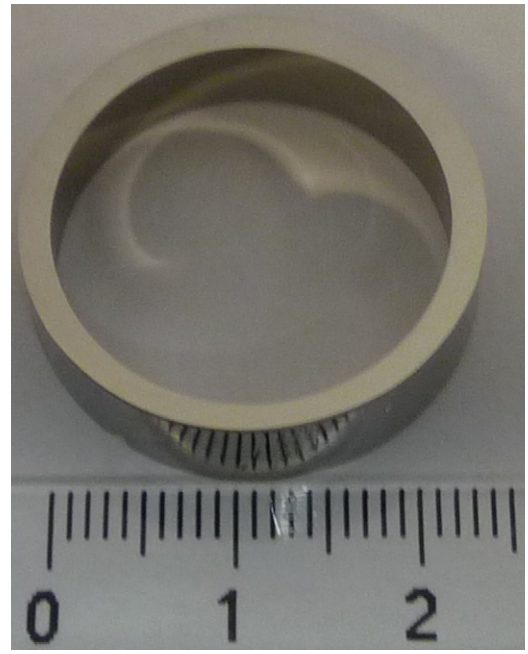

Fig. 2. A glassy ring cut in cross-section from the optimized parameter casting.

The X-ray diffraction (XRD) patterns of the cast specimens were measured with a Siemens Kristalloflex 710H X-ray generator and Inel CPS-120 position sensitive detector. Instrumented indentation tests were conducted with a Berkovich diamond indenter on a CSM Instruments nanoindentation tester. Linear loading with few seconds of holding time was used. The indentation force used was 40 mN.

Contact mode atomic force measurements were done with an AFM-S-AE-004X atomic force microscope ($xy = 40\,\mu m$, $z = 4\,\mu m$) integrated into the nanoindenter platform. A 450 μm long silicon contact probe with ≤10 nm tip was used in the measurements (Fig. 4).

3. Experimental results and discussion

The wall thickness of 0.8–3 mm used in this study is relatively large, allowing the molten alloy to flow into the first-filled portion of the mold cavity during tilt casting, without the application of a suction pressure difference. As a result, preliminary trials using smaller melt charges only filled one side of the mold. A larger melt charge allowed the filling of the entire mold circumference, so that the rest of the mold could be filled by suction casting after the one side was filled by tilt casting. However, there was a longitudinal cold shut running on both sides of the tube. This casting defect, which impacts the mechanical integrity, varied in severity along the length of the tube. Thus, there were very large differences in thermal history between different sections of the casting, with variations both longitudinally along the tube and circumferentially. In essence, one side of the casting was filled quickly, fed from a high-

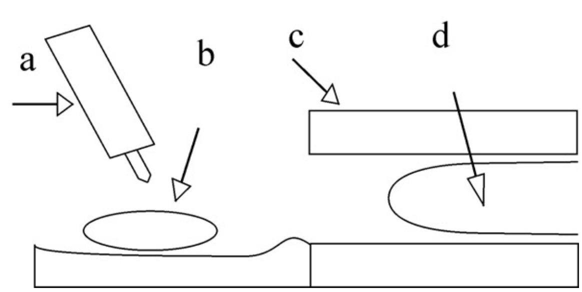

Fig. 3. An illustration of the used casting setup with (a) arc melting electrode, (b) molten alloy, (c) mold and (d) core. In casting the setup is tilted clockwise to pour the melt into the mold.

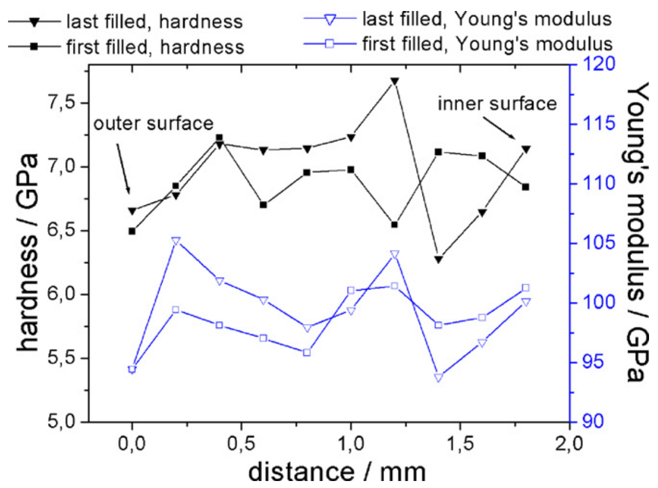

Fig. 4. Young's modulus and hardness profiles as a function of the distance from the outer surface to the inner surface for the as-cast ring. Results for both the first-filled and the last-filled sections are shown.

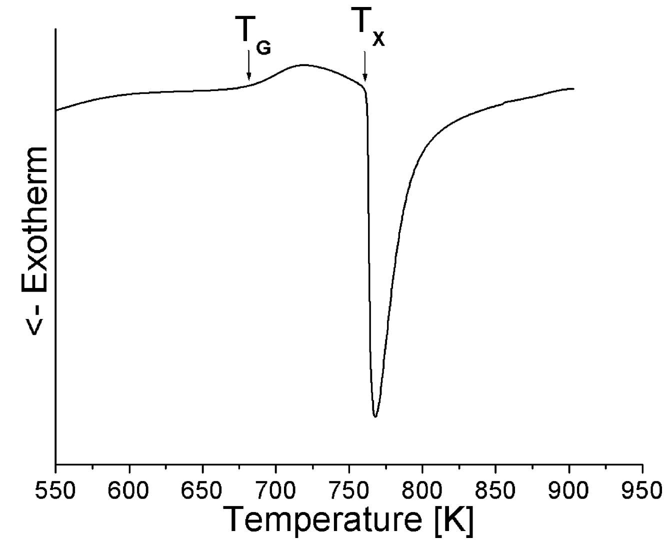

Fig. 6. Differential scanning calorimetry measurement from a cast tube segment showing distinctive glassy region between T_G and T_X.

temperature running melt, and near the bottom already solidified, before the other side was filled more slowly, fed from a lower-temperature standing melt, in a mold already preheated by the material on the first-filled side. Nevertheless, both tube halves were found to be fully amorphous when tested by X-ray diffraction and differential scanning calorimetry to be amorphous.

MingZhen et al. [11] discuss the effects of latent heat release from the melt and heat transfer to the mold upon freezing of the melt as the mold is filled. The heat transfer from the melt to the mold is much affected by any created thermal shrinkage gaps between the mold and the specimen surface, and the atmosphere in these gaps [16]. The latent heat is affected by both the size of the melt charge and the power with which it is heated just before casting. To improve the rate at which the second side of the mold is filled during suction casting, in an effort to eliminate the cold shuts, their conclusions indicate a melt charge with more latent heat to begin with should be used. More latent heat would thus need to be removed from the melt before it solidified, simultaneously increasing the time available to fill the mold, and lowering the melt viscosity so that the mold cavity would fill more quickly. In addition, the suction line of the casting setup was modified, increasing its diameter and improving its vacuum sealing. These modifications also reduced the mass of the mold. With improved suction, higher melt mass and higher arc power just before casting, the tube shown in Fig. 1 was produced. The mold was completely filled and the cross-section showed no cold shut formation. Unfortunately, despite the high gloss surface quality, the cross-section instrumented indentation results pointed to a partially crystallized microstructure, as shown in Fig. 7. The instrumented indentation measurement provides a convenient method of evaluating variations in process parameters: fluctuations in the measured hardness, over a cross section of the specimen, indicate partial crystallization.

Clearly, the modifications had the intended effect, so much so that the melt was cooled too slowly to form fully glassy (or amorphous) structure. Optimal process parameters were clearly located between these two values. Hence, with lower arc power and melt mass, the glassy ring cross-section shown in Fig. 2 was produced. Both tube halves were found to be fully amorphous when tested with X-ray diffraction and differential scanning calorimetry. Typical results of these measurements are shown in Figs. 5 and 6. Unlike in the tube shown in Fig. 1, the indentations taken from the ring cross-section shown in Fig. 2 show almost no difference between the first-filled and last-filled portions and thus indicate no signs of partial crystallization, as shown in Figs. 4 and 7 and in Table 1.

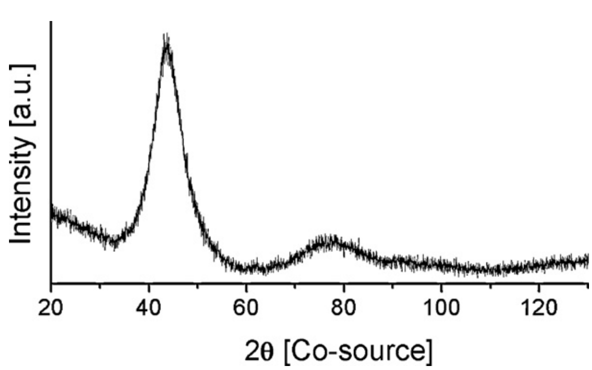

Fig. 5. X-ray diffraction (Co K_α) pattern of cast tube segment, showing amorphous structure. Samples were prepared with a water-cooled aluminium oxide impregnated cut-off wheel. Cut-off wheel residue was removed with 1 μm diamond paste polishing.

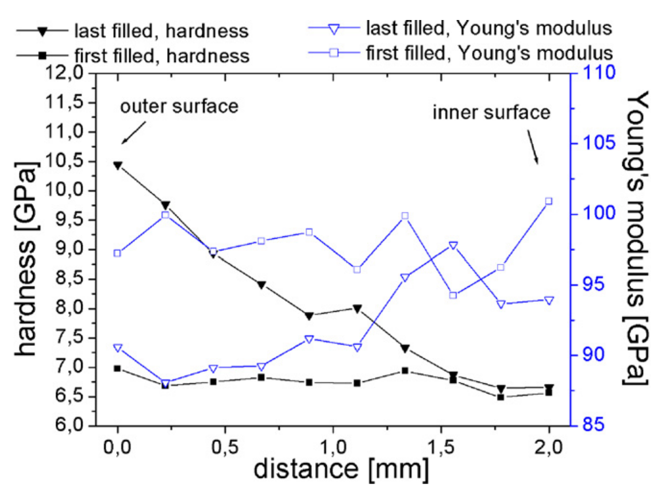

Fig. 7. Young's modulus and hardness profiles as a funtion of the distance from the outer surface to the inner surface for the as-cast partially crystallized ring. Results for both the first-filled and the last-filled sections are shown. The large hardness on the cross-section near the outer surface of the last-filled section indicates composite structure.

Table 1
Hardness and Young modulus at initial unloading, with their standard deviations, as measured by instrumented indentation.

	Hardness (GPa)	Modulus (GPa)
Tilt-cast side	6.88 (0.24)	98.6 (2.4)
Suction-cast side	6.99 (0.4)	99.4 (3.8)

4. Conclusions

A glassy $Zr_{55}Cu_{30}Al_{10}Ni_5$ (in at.%) ring with outer diameter of 25 mm and 1.8 mm thickness was produced, using a custom-built combined arc-melting tilt-casting furnace. Both tilt and suction casting were used to ensure mold filling. Tilt casting was found to fill one side of the tube mold first, with the rest of the tube circumference filled subsequently by suction casting. Different sections of the as-cast ring were investigated by X-ray diffraction (XRD), differential scanning calorimetry (DSC), and instrumented indentation to ensure glassy microstructure.

Optimized casting parameters are required to fully realize the potential of directly manufacturing complex shapes out of high-purity bulk metallic glasses by tilt casting.

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