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ScienceDirect

Acta Materialia 62 (2014) 49–57

Acta MATERIALIA

www.elsevier.com/locate/actamat

Joining of bulk metallic glasses in air

Wen Chen [a], Ze Liu [b], Jan Schroers [a,b,*]

[a] Department of Mechanical Engineering and Materials Science, Yale University, New Haven, CT 06511, USA
[b] Center for Research on Interface Structures and Phenomena, Yale University, New Haven, CT 06511, USA

Received 15 July 2013; received in revised form 26 August 2013; accepted 26 August 2013
Available online 19 October 2013

Abstract

We present a thermoplastic deforming method to join metallic glasses in air. Mechanistically during straining of the interface the oxide layer breaks and pristine alloy flows towards the interface and forms a metallic bond. To demonstrate the effectiveness of this method we chose reactive $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ as an example bulk metallic glass system. A model is introduced which quantitatively predicts the bonding strength solely from the shear strength of the metallic glass, the initial surface roughness, and the applied strain. The ability to join even reactive metallic glasses in air on a timescale of the order of milliseconds to seconds at low pressure and temperature with predictable joint strength suggest a highly practical and economic method to join metallic glasses.
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Keywords: Joining; Thermoplastic forming; Bulk metallic glass

1. Introduction

Among the attributes of a material that determine its usage is its ability to be joined to itself or other materials [1]. In principle, a strong physical bond of two surfaces requires atomic contact and surfaces free of contaminants. For polymeric materials such as thermoplastics, fusion bonding has been successfully developed to produce close to parent joint strengths through thermoplastic deforming or melting of the interface such that immediate molecular contact can be achieved [2–4]. For metals, establishing the requirements of atomic contact and a contamination-free surface in practice is challenging. The high surface energy of metals attracts many of the contaminants in the processing environment. One of these elements is oxygen, which leads to the formation of a typically rigid oxide layer. Oxide layers act as a diffusion barrier and in general render metallic bonding difficult. However, even in the absence of an oxide layer the requirement of an atomic contact over macroscopic areas is for most practical cases highly challenging for conventional metals due to their crystalline structure, imperfections within this structure, and temperature-dependent strength, which render them either too rigid or they melt.

Bulk metallic glasses (BMGs) are amorphous in nature. They exhibit a fundamentally different temperature-dependent strength which decreases rapidly but continuously to a low level, representing a soft readily deformable state [5,6]. As a consequence of this thermoplastic-like softening and due to the amorphous structure, BMGs can be readily formed even on the atomic scale [7], with broad inter-atomic contact over large macroscopic areas. However, they still oxidize or attract other contaminants to their surface.

The metastable nature of BMGs against structural relaxation and crystallization rules out most techniques that are used to join crystalline materials [8,9]. The metallic glass community has therefore invested significant effort in developing joining methods. BMG joining methods have been developed based on two different principles. One utilizes the supercooled liquid state, which is present in the

* Corresponding author at: Department of Mechanical Engineering and Materials Science, Yale University, New Haven, CT 06511, USA. Tel.: +1 203 432 4346; fax: +1 203 432 6775.
E-mail address: jan.schroers@yale.edu (J. Schroers).

1359-6454/$36.00 Published by Elsevier Ltd. on behalf of Acta Materialia Inc.
http://dx.doi.org/10.1016/j.actamat.2013.08.053

temperature region between the glass transition temperature, T_g, into the crystallization temperature, T_x [10]. A common theme among such methods is that they rely on long-range diffusion bonding [11,12]. Avoiding crystallization has been the main challenge since the diffusion kinetics and crystallization kinetics are similar [11]. Different in principle are the joining methods where the BMG is melted. These include friction welding [13,14], laser welding [15–18], chemical reactive layer joining [19,20] and electron beam welding [21,22]. Here the requirement of fast cooling to prevent crystallization is the main challenge. In joining methods based on melting a larger region than the intended interface is affected. Therefore, a broader area is heated to a high temperature above the melting temperature, T_m, and hence rapid cooling becomes challenging [18,20,22]. As a result, structural relaxation or crystallization often occurs. To reduce oxidation, joining of metallic glasses based on either of the two principles is typically conducted in high vacuum or in an inert environment, which is undesirable as a practical method.

Here we introduce a joining method in which we thermoplastically deform the metallic glass in the supercooled liquid region at the interface. The resulting interface strain breaks the oxide layer and creates cavities that are filled by pristine BMG, which form a metallic bond on the joined interface. As such, the presented method does not rely on long-range diffusion and as a consequence crystallization can be readily avoided because of the short timescale of milliseconds to seconds to fill the cavities and for formation of a metallurgical bond. The interface strain defines the fraction of pristine material on the interface, which dictates the overall bond strength.

2. Experimental procedure

To demonstrate the effectiveness of the introduced method we used $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ as an example of a reactive BMG. Master alloy ingots of $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ were prepared by arc melting a mixture of the pure elements in an argon atmosphere. The amorphous state was achieved by water quenching in a 10 mm (inner) diameter quartz tube. The amorphous nature of the BMGs was confirmed by X-ray diffraction (XRD). The onset glass transition temperature and crystallization temperature for this BMG are 583 K and 738 K, respectively, measured at a standard heating rate of 20 K min^{-1} by differential scanning calorimetry (DSC).

Two BMG circular disks of approximately 1 mm thickness and 10 mm diameter were cut from the BMG rod for joining. The disks were polished so that they were parallel and smooth using a 1 μm diamond suspension, followed by ultrasonic cleaning in acetone, isopropanol and deionized water. Joining of the two BMG disks was carried out in an Instron 5569 test machine. Two parallel loading plates were attached to the Instron machine for thermoplastic compression of the BMG disks. The temperature in the two plates was controlled by proportional integral derivative (PID) controlled cartridge heaters. Uniaxial compression of two $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ disks against each other at 700 K in the supercooled liquid region under a constant strain rate of $\dot{\varepsilon} = 5 \times 10^{-2}\ s^{-1}$ results in joining. The viscosity of $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ at this temperature is $\eta \approx 10^6$ Pa s, calculated using the Vogel–Fulcher–Tammann (VFT) equation [23]. The flow stress σ was estimated to be 0.15 MPa under the assumption of Newtonian flow, $\sigma = 3\eta\dot{\varepsilon}$. Different thermoplastic joining strains of $\varepsilon = 0.1$, 0.2, 0.3, 0.45 were realized to examine the role of joining strain on the joining strength. After joining of the two BMG disks a standard lap shear joint specimen was cut from the joined geometry (Fig. 1). Quasi-static shear tests were conducted on the lap specimen to determine the joint shear strength using an Instron 5543 tensile test machine at a strain rate of $10^{-4}\ s^{-1}$. For each set of parameters three shear tests were performed to quantify scatter. After shear failure the fractured lap area was imaged using a Keyence VHX-500F digital microscope and subsequently analyzed using ImageJ analysis software. Scanning electron microscopy (SEM) was employed to examine the joined interface and the surface fracture morphology after shear failure. Nanoindentation was used to determine the hardness in the vicinity of the interface. More than 10 in-line indents, spaced 20 μm apart were performed across the joined interface with a CSM instrumental nanohardness tester. We also investigated the influence of initial surface roughness on the joining strength. Therefore, the initial surfaces of the BMG disks were ground to four different roughnesses using 320 (200 μm), 800 (25 μm), 2500 (10 μm) SiC sanding paper, and 1 μm diamond paste. To study the feasibility of the thermoplastic joining approach in dissimilar BMGs, $Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$ was used and joined to $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$. T_g and T_x for $Zr_{44}Ti_{11}Cu_{10}Ni_{10}Be_{25}$ BMG are 623 K, 744 K respectively, which are 40 K and 6 K higher than the values for $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ [24]. The joining processing conditions of the dissimilar BMGs were 1 mm initial thick, 10 mm diameter circular disk-shaped specimens, surface polished to 1 μm roughness, joining temperature $T = 710$ K, joining strain rate $\dot{\varepsilon} = 5 \times 10^{-2}\ s^{-1}$, joining strain $\varepsilon = 0.4$.

3. Results and discussion

3.1. Effect of thermoplastic joining strain on the joining process

We joined a pair of $Zr_{35}Ti_{30}Cu_{7.5}Be_{27.5}$ BMG disks with different strain values ($\varepsilon = 0.1$, 0.2, 0.3, 0.45) and subsequently investigated fractographs of the interface after shear tests (Fig. 2). The fracture surfaces at different joining strains exhibit a combination of distinct smooth and veined regions. Such vein-like patterns are generally considered to be related to a high temperature rise due to local melting which originates from pronounced elastic energy release in instantaneous fracture [25–27]. The vein pattern reflects a strong metallic bond formed during joining and typically