



Designing Bulk Metallic Glass Composites with Enhanced Formability and Plasticity

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[Manuscript received February 14, 2014, in revised form March 10, 2014, Available online 16 May 2014]

To address the main stumbling-block of bulk metallic glasses (BMGs), i.e., room temperature brittleness, designing BMG matrix composites has been attracted extensive attention. Up to date, BMG composites in various alloy systems have been successfully developed by forming crystalline phases embedded in the amorphous matrix through either *ex-situ* or *in-situ* methods. In this paper, a brief review of our recent work in this topic will be presented and the novel approaches to improving composite formability and mechanical properties will also be highlighted. The main purpose of this manuscript is not to offer a comprehensive review of all the BMG composites, but instead focuses will be placed on illustrating recently developed advanced BMG composites including Fe-based BMG composite with no metalloids, Al-based BMG composite and BMG composites reinforced by the TRIP (transformation-induced plasticity) effects. The basic ideas and related mechanisms underlying the development of these novel BMG composites will be discussed.

KEY WORDS: Bulk metallic glasses; Designing; Formability; Plasticity

1. Introduction

Bulk metallic glasses (BMGs) have shown a unique combination of mechanical, chemical and physical properties^[1–5], but their room temperature brittleness and strain softening behavior have been one of the stumbling blocks to real structural applications^[6,7]. To answer this challenge, the concept of developing glassy composite microstructures by forming crystalline phases embedded in the glassy matrix through either *in-situ* or *ex-situ* methods have been developed^[8–21]. Initially, BMG composites were formed mainly due to partial crystallization of fully glassy precursors via annealing experiments. In the late 1990s, it was recognized that the brittle failure of BMGs could be mitigated with additions of crystalline phases in the BMG matrix. Gradually, formation of appropriate crystalline phases in BMG matrices has become a toughening strategy. By adjusting processing parameters and/or alloy compositions, various crystalline phases could be either *in-situ* or *ex-situ* formed inside BMGs, and large compressive plasticity was successfully achieved^[18–21]. The basic idea underlying this strategy is to manipulate formation of shear bands (i.e., nucleation and propagation of shear bands)

during deformation by utilizing the inclusions as reinforcing media.

Compared with the amorphous matrix, most reinforced crystalline inclusions were ductile with a lower elastic modulus and hardness. As a result, these reinforcing phases were designed to yield before the amorphous matrix and thus bear more plastic deformation^[18–21]. Nevertheless, a few BMG composites with brittle intermetallic compounds were also found to be capable of improving the plasticity^[22]. Early work was mostly focused on enhancing compressive plasticity of BMG materials^[20–22]. In recent years, by properly controlling the preparation routes and alloy compositions, the microstructural length scale (i.e., the dendrite spacing of the primary phase) was adjusted to match the mechanical length scale (i.e., the plastic shielding of an opening crack tip), large tensile ductility and superior fracture toughness have been achieved in a series of Zr- and Ti-based BMG composites^[23–26]. In this approach, the precipitated dendrites are usually thermodynamic stable phase, which endows the composite technically more processable, e.g., the characteristics of the crystalline inclusions can be controlled by processing techniques such as “semi-solid” formation and Bridgman solidification^[26–28].

In-situ formed BMG composites usually have good crystal-amorphous matrix interface and a relative simple fabrication process, but compromises among glass-formation ability (GFA), composition and cooling rates have to be made. In contrast, features of crystalline inclusions in *ex-situ* formed BMG composites, such as distribution and volume fraction, can be easily

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<http://dx.doi.org/10.1016/j.jmst.2014.03.028>

adjusted. However, the fabrication processes are complex and crystal-amorphous matrix interface is always the weak zone during deformation. In the late 1990s, various attempts were conducted to develop *ex-situ* formed BMG composites by infiltration casting with crystalline wires or powders to create two-phase mixtures^[10,11]. For instance, additions of W fibers/balls, porous Ti or particles of Ti, Mo and SiC into Zr-based and Mg-based BMGs could lead to remarkable increment in the compressive plasticity^[44–47], and the improvement was attributed to the slowdown of shear band propagation due to the *ex-situ* added reinforcements. In these reported approaches, nevertheless, the added second phases are usually distributed discontinuously with significant inhomogeneity in the BMG matrix, which greatly reduce the reinforcing effectiveness^[48]. It is important to point out that tensile ductility in these *ex-situ* BMG composites was still absent.

The concept of “transformation-induced plasticity” (TRIP) has been proven to be effective in enhancing tensile ductility and toughness in crystalline materials, such as steels and ceramics^[29,30]. In recent years, to further improve tensile ductility and work-hardening capability of BMGs, TRIP effect was also introduced and was confirmed to be a novel way to improve both tensile ductility and work-hardening ability in BMGs^[31–40]. Up to date, several TRIP-reinforced BMG composites systems have been developed, mostly in CuZr-based BMG^[35–40] and Ti-based BMG^[41–43]. Formation, mechanical behavior and microstructure evolution during deformation in this type of BMG composites have been preliminarily studied^[35–43], but the deformation mechanism, especially the details about contribution of martensitic transformation to the macroscopic properties, remain unclear. Nevertheless, this TRIP concept did prompt extensive attention in developing novel BMG composites with advanced properties, such as Ti- and Fe-based BMGs^[41–43].

In this paper, several special approaches developed recently in our group for designing advanced BMG composites with either enhanced formability or mechanical properties were summarized. The intention is to highlight basic ideas and related mechanism underlying development of these novel BMG composites, including Fe-based BMG composite with no metalloids, Al-based BMG composite and TRIP-reinforced BMG composites.

2. *In-situ* Formed BMG Composites

2.1. Large-sized BMG composites fabricated by phase separation

Phase separation has been found in metallic glasses and could be used to improve the deformation plasticity^[49–51]. Recently, we used the concept of phase separation to design BMG composites^[52].

Firstly, a known BMG with a good GFA, e.g., element *A*-based $A_xB_yC_zD_uE_v$ ($x + y + z + u + v = 100$ at.%), was selected as the starting composition, and then an element *M* having a positive enthalpy of mixing and a miscibility gap with the main component *A*, was chosen and added. As a result, two liquids, i.e., the *M*-rich region and *M*-depleted region, will form at high temperatures. Due to the phase separation, the *M*-depleted region would have a composition close to the starting *A*-based alloy, which has a good GFA. During solidification, it can be thus anticipated that crystalline phases will be precipitated out of the *M*-rich liquid, while the *M*-depleted liquid will vitrify. As such, an *M*-based BMG composite consisting of the *M*-rich crystalline

phases embedded in the *A*-based BMG matrix can be fabricated. The total volume fraction of the BMG matrix can be adjusted by the amount of the *M* addition^[52].

This approach can be widely used to design special BMG composites based on currently known BMG systems and thus optimize overall properties of BMG materials.

2.1.1. Development of centimeter sized BMG composites with high Fe concentration.

It was widely recognized that the metalloid elements (i.e., B, C, Si and P) are key glass-forming constituents in Fe-based alloy systems^[53–55], and a good Fe-based glass-former often contains a metalloid content of 20 at.% or above^[53–55]. Nevertheless, it was found that the high content of the metalloid elements usually bring in side effects on the soft magnetic properties and plastic deformation at room temperature in these Fe-based BMGs^[56,57]. Based on the concept of phase separation, we have successfully developed a series of BMG matrix composites with high Fe concentration but without any metalloid elements. The Fe element which has a positive mixing enthalpy with La and Ce, was added in the BMG composition $\text{La}_{32.5}\text{Ce}_{32.5}\text{Co}_{15}\text{Al}_{10}\text{Cu}_{10}$, as shown in Fig. 1(a).

Due to the repulsion between the Fe and (La, Cu), phase separation occurred and the melt initially separated into two liquids, i.e., the Fe-rich and the Fe-depleted. During cooling, Fe-rich phases, i.e., CeFe_2 and $\text{Ce}(\text{Fe}, \text{Co})_2$ were firstly formed and the residual composition was adjusted near to our starting composition, i.e., the LaCe-based bulk glass-former, and finally solidified into glass phase. Therefore, a composite structure with a large percentage of Fe was fabricated (Fig. 1(b)).

The current BMG matrix composites show no significant improvement in plastic deformation, which is probably attributable to the brittle primary phases and the weak interfaces between the primary phases and the glassy matrix. Nevertheless, this work provides a useful basis for developing large-sized BMG composites with a large formability and deformability.

2.1.2. Centimeter sized BMG composites with a high Al content.

Unlike that of the other metal-based alloy systems such as Zr-, Ti-, and Cu-based BMGs, the maximal attainable thickness of Al-based metallic glasses still cannot exceed 1 mm^[58–61]. The formation of Al-based metallic glasses or the corresponding glass matrix composites is limited to thin ribbons which seriously restrict the utilization of their high specific strength and low density^[58–61].

Based on the aforementioned concept, we developed a series of Al-(Co,Cu)-(La,Ce)-Fe BMG matrix composites which contain a high Al content from 35 to 60 at.%^[62]. It was found that additions of Fe, which has a positive heat of mixing with the main constitute elements La and Ce, enhanced the volume fraction of glass matrix by suppressing the precipitation of the Al(La,Ce) phase. The strength of these glassy composites reaches up to 1.2 GPa which is twice that of conventional Al-based alloys. These composites possessing extraordinarily large formability with a critical size over 15 mm were produced by common copper mold casting (as shown in Fig. 2). For the alloy containing 30 at.% Al, the BMG composites mainly consisted of the $\text{Al}_2(\text{La,Ce})$ phase and glass matrix, and the volume fraction of the glass matrix is about 80%. For the alloy containing 30–50 at.% Al, the BMG composites mainly consisted of the $\text{Al}_2(\text{La,Ce})$ phase, the Al(La,Ce) phase and glass matrix, and the volume fraction of glass matrix is about 30%–60%.

Proper addition of the Fe element effectively suppressed the precipitation of the Al(La,Ce) phase and consequently enhanced

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