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Compressive deformation of in situ formed bulk metallic glass composites

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Abstract

A bulk metallic glass matrix composite with dendritic second phase precipitates was investigated using neutron diffraction and self-consistent modeling (SCM) to ascertain its deformation mechanisms. The compressive behavior of both the composite and the second phase (in its monolithic form) were investigated. The diffraction data were compared to the predictions of a new SCM resulting in good agreement. For the first time, this model considered both amorphous and crystalline phases and allowed the calculation of single crystal elastic constants from polycrystalline diffraction data. It was shown that the ductile second phase yielded first upon loading, and this was followed by multiple shear band formation in the matrix, a process which enhanced the ductility of the composite.

Keywords: Metallic glass; Composites; Self-consistent modeling; Mechanical properties; Neutron diffraction

1. Introduction

Bulk metallic glasses (BMGs) have recently attracted attention as emerging structural materials due to their high elastic strain limit (about 2%), high strength (around 2 GPa) and good fracture toughness (~20 MPa $m^{1/2}$) [1–3]. However, most BMGs experience sudden failure during unconstrained loading at room temperature, which severely limits their use in load bearing applications. This lack of ductility in BMGs has been addressed by the development of several BMG matrix composites with significantly improved damage tolerance [4–12]. Among the most promising of these composites are those processed in situ when a dendritic second phase precipitates during casting [13–15]. This phase has a body-centered cubic (bcc) crystal structure, consists of primarily Zr and Ti and hence is referred to as the "β phase" since it is reminiscent of the β allotrope of both Ti and Zr. Upon cooling from the high temperature melt, the initial alloy undergoes partial crystallization by nucleation and dendritic growth of the β phase. The remaining liquid subsequently freezes to an amorphous solid producing a two phase microstructure containing β phase dendrites in a glassy matrix. The overall composition of the composite is $Zr_{56.2}Ti_{13.8}Nb_{5.0}Cu_{6.9}Ni_{5.6}Be_{12.5}$, while the compositions of the matrix and β phase are $Zr_{44.7}Ti_{12.2}Nb_{2.7}Cu_{10.5}Ni_{9.1}Be_{20.8}$ and $Zr_{71}Ti_{16.3}Nb_{10}Cu_{1.8}Ni_{0.9}$, respectively [15].

The dendritic structure of the β phase has been shown to inhibit the formation of macroscopic shear bands in the matrix which cause sudden failure in monolithic BMGs. The mechanical properties of the BMG/β phase composites have previously been characterized via macroscopic measurements [13–15]. It was found that the β phase leads to the formation of multiple shear bands in the matrix with a similar spacing to that of the secondary dendrites. However, neither the underlying deformation mechanisms nor

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the load sharing in the composite could be determined in these studies. There was some speculation that plastic deformation via dislocation slip, twinning or a stress-induced phase transformation in the β phase might play a role. Since the β phase composites offer an attractive venue to manipulate microstructure and optimize the mechanical properties of BMG matrix composites, it is important to understand their effective deformation mechanisms.

In the present study, neutron diffraction (ND) was used, aided by self-consistent modeling, to investigate the in situ deformation of a BMG/β phase composite and a β phase monolith during compressive loading. ND allows for in situ bulk measurements of internal strains in crystalline materials, and is ideal for composite systems as it is phase specific. Although the amorphous nature of BMG precludes its strain analysis by diffraction (except in special cases [16]), the crystalline β phase could be used as an internal "strain gauge". Then by combining the diffraction data for the β phase with model predictions of the composite behavior, it was possible to infer the in situ behavior of the BMG matrix. Moreover, the neutron diffraction data could identify the nature of the deformation mechanism in these composites.

2. Experimental procedure

A composite with 40 vol.% β phase was processed as described in [13,17]. In addition, a monolithic β phase sample with the same chemical composition found in the dendrites was prepared [17]. In situ compression testing was performed on the specimens while ND data were collected in the neutron powder diffractometer (NPD) at the Lujan Neutron Science Center. All specimens were cylindrical with a 6 mm diameter and 14.4 mm length (aspect ratio = 2.4). The setup of NPD allowed for the collection of diffraction patterns in the longitudinal and transverse directions simultaneously [18]. Using the time-of-flight technique, diffraction patterns within a d spacing range of 0.5–4 Å were collected and the Rietveld method [19–21] was employed to determine an average lattice strain in the β phase based on changes in its lattice parameter. The reported lattice strains were calculated relative to the lattice parameter at a nominal −5 MPa compression needed to hold the sample in the load frame.

3. Results and analysis

3.1. Neutron diffraction experiments

Throughout the study (in both the monolithic and composite forms) the β phase was successfully described with the space group *Im3m* (bcc). The weighted fitting residuals, R_{wp}, for all Rietveld refinements were around 0.05–0.07 suggesting a good match between the data and the Rietveld model. This result eliminates the possibility of a stress-induced phase transformation in this phase (at least within the ~3–5 wt.% detection limit of ND).

The macroscopic longitudinal deformation of both samples was monitored by an extensometer during the ND experiments (see Figs. 1(a) and 2(a)). The stiffness of the monolithic β phase sample obtained from the extensometer data was around 70 GPa, comparable to the literature value of 63 GPa [15]. The macroscopic stiffness of the composite was 74 GPa, again close to the literature value (79 GPa in Ref. [15]). The deviations are attributed to sample and extensometer mounting and sample bending during loading, since as described below, the Young's modulus and Poisson's ratio of the β phase calculated from the bulk-averaged neutron data were within 7% of the literature values.

3.2. Lattice strains and elastic anisotropy

The neutron diffraction data were analyzed using both the whole-pattern Rietveld refinement method [19–21] and single peak fits. The former yields an average strain value for a given phase whereas the latter provides

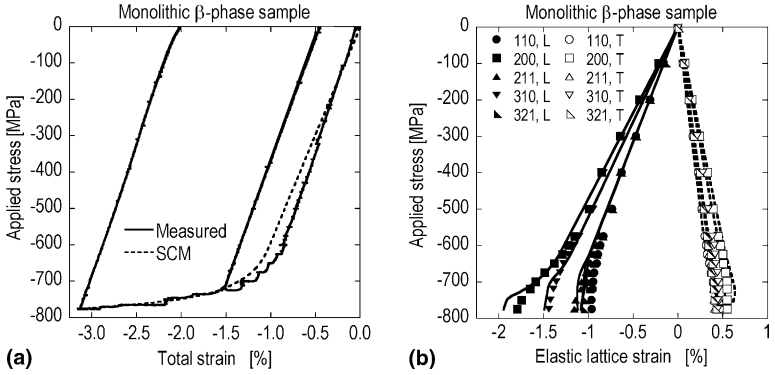

Fig. 1. Measured and calculated response of the monolithic β phase sample to applied compressive stress: (a) macroscopic strain along the loading axis; and (b) lattice plane specific elastic strain (symbols designate neutron data while lines are SCM predictions; here "L" and "T" indicate longitudinal and transverse directions, respectively). The difference between measured and calculated macroscopic stiffness shown in (a) is likely due to extensometer problems. This claim is supported by the fact that the diffraction data in the elastic region is in good agreement with the model calculations as shown in (b).

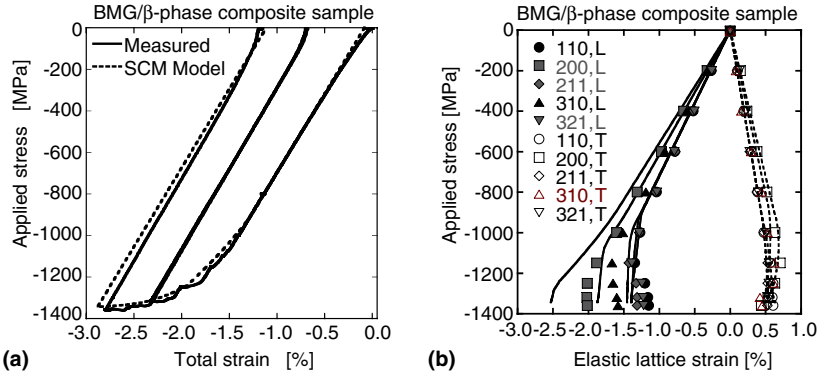

Fig. 2. (a) Comparison of measured (by extensometer) and calculated (SCM) macroscopic stress–strain curve for the composite sample. (b) Comparison of measured (symbols) and calculated (lines) lattice specific stress–strain curves for the β phase in the composite sample ("L" and "T" indicate longitudinal and transverse directions, respectively).

information about lattice plane specific elastic and plastic behavior and is useful in self-consistent model calculations.

Self-consistent models (SCM) [22,23] can predict the lattice plane specific Young's modulus and Poisson's ratio, also known as the diffraction elastic constants (DECs), with good accuracy [22,24,25]. These calculations require as input only the single crystal stiffnesses of the material. Within the SCM scheme the polycrystal is regarded as an agglomerate of ellipsoidal grains whose properties are determined by the single crystal properties (elastic and plastic) of the material. Interactions between grains are modeled using the Eshelby theory [26] where each grain is embedded in a matrix (or "equivalent medium") with the average properties of the polycrystal. Direct comparison of SCM predictions with diffraction experiments is possible via weighted averages of elastic strains in grains that are oriented with given lattice plane normals parallel to the scattering vector used in the experiment. In the present work a "reverse" approach was followed, namely the SCM was used for the first time in combination with a least squares fitting program to determine the single crystal stiffnesses of the β phase based on its measured DECs. This approach is similar to that of Gnaupel-Herold et al. [27] except that the SCM enables one to take into account the effects of the orientations and the finite size of the detectors in diffraction experiments. This is especially critical when the material is highly anisotropic, as is demonstrated below for the β phase.

The input data for the calculation of the single crystal stiffnesses of the β phase were the slopes of linear fits to the elastic region of the loading curves (up to about −600 MPa) in Fig. 1(b). The results of the calculation show that the β phase has a relatively low Young's modulus (59 GPa), but large elastic anisotropy ($2C_{44}/(C_{11} - C_{12}) = 3.0$ where $C_{11} = 90$, $C_{12} = 68$, and $C_{44} = 33$ GPa are single crystal stiffness components) leading to elastic constants that vary significantly with crystallographic orientation. This can also be deduced from the spread of slopes in the elastic portions of the loading data in Fig. 1(b).

3.3. Self-consistent modeling of plastic deformation

To understand the in situ plastic deformation of the composite, the SCM calculations were extended into the plastic regime. The β phase dendrites were approximated as spheres and a new self-consistent polycrystal deformation model was developed to describe the mechanical behavior of the β phase/BMG composites. A recent study [28] confirmed that the shape of β phase dendrites does not influence the overall SCM predictions and that a sphere is a good approximation of a dendrite.

In addition to the "reverse" calculation of the single crystal elastic constants described above, the other new addition to the SCM was the inclusion of a second phase to represent the BMG matrix. Here, the self-consistent polycrystal deformation model of Turner and Tome [23] was improved so that the deformations of both the β phase and the BMG matrix were accounted for. The basic approach was to use the measured macroscopic stress–strain curve of the composite to refine its yielding and hardening behavior. To further validate the model, its lattice plane specific predictions were then compared to diffraction data from the β phase inside the composite [28].

First, the elastic–plastic deformation of the monolithic β phase was evaluated. Reasonable fits to both the macroscopic strains and the lattice plane specific diffraction data were obtained (Fig. 1). Overall, the monolithic β phase was found to have a uniaxial yield stress of −610 MPa. This corresponds to an initial critical resolved shear stress of −300 MPa, which is the threshold shear stress used in the crystal-plasticity-based SCM. Its strain hardening coefficient was found to be about zero [28].

In the composite the metallic glass matrix was modeled as an isotropic continuum, i.e., without the elastic and plastic anisotropy found in the crystalline β phase. For the BMG, the von Mises (or J_2) yield criterion [29] was assumed, which was shown by Lowhaphandu et al. [30,31] to be a good approximation of the plastic behavior of most BMG alloys. The details of this improvement of the SCM are described in [28].

The β phase was modeled as a set of 10 000 single crystals with different orientations and the inherent single crystal stiffnesses and slip systems of the bcc crystal structure. The BMG was introduced into the model as an isotropic single grain weighted with the appropriate ratio of the volume fractions of the matrix and β phase (60% and 40%, respectively). In this formulation, the BMG still interacted with the average composite matrix (also known as the "equivalent medium") as did the β phase grains.

The results of the SCM calculations for the composite sample are shown in Fig. 2. The macroscopic predictions of the model were fitted to the measured composite stress–strain curve reasonably well (Fig. 2(a)) using the initial BMG yield stress, the initial critical resolved shear stress for the β phase, and the hardening behavior of the BMG as variables. The single crystal elastic constants for the β phase used in the composite calculations were those of the monolithic β phase. The Young's modulus and Poisson's ratio for the BMG matrix were taken from Ref. [15]. The β phase was treated as non-hardening (or perfectly plastic, similar to the case for the monolithic β phase). This is consistent with the observation that the stress vs. elastic (lattice) strain curves for the β phase in the composite become vertical after yielding (Fig. 2(b)). The reader should note that another mechanism that would lead to such behavior in the β phase is its complete debonding from the BMG matrix. So far, no large scale debonding at β phase/matrix interfaces has been observed [15].

The initial critical resolved shear stress for the β phase was estimated as −325 MPa and the initial von Mises critical shear stress of the BMG was found to be −800 MPa. These values are somewhat different than those for the monoliths (−300 MPa for the monolithic β phase and a literature value of about $-1600/\sqrt{3} = -924$ MPa for the matrix [15]). It is possible that thermal residual stresses developed in the composite during processing altered the "apparent" yield points of each phase. If true, this would suggest an initial tensile longitudinal residual stress in the β phase and a compressive stress in the BMG.

The hardening behavior of the BMG was modeled with a Voce-type exponentially decreasing hardening function (often used in the traditional SCM [23]). The hardening modulus of the BMG was found to change from an initial value of 52 GPa at an initial critical shear stress of −800 MPa to a final value of 1 GPa over an 80 MPa range. The reader should note that all the SCM parameters were determined by trial-and-error calculations which yielded an error bar of less than 5%.

To further validate the model, its lattice plane dependent estimates were compared with the diffraction data (Fig. 2(b)). The model predicts the elastic region with good accuracy. The predictions are also satisfactory in the plastic region except that beyond −1000 MPa a larger discrepancy develops between the model and the data. The SCM estimates also compare well to the transverse strain data. Overall, the agreement between the model predictions and the neutron data is satisfactory and comparable to SCM predictions for other materials [22].

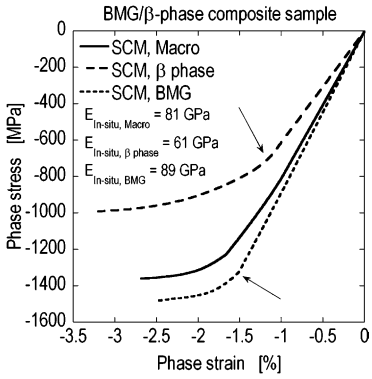

Fig. 3. Calculated in situ behavior (phase stress vs. phase strain) for the composite. The arrows indicate the yield points in the β phase (about −650 MPa) and the BMG matrix (around −1350 MPa). The solid line ("macro") designates the composite behavior. The in situ Young's moduli for each phase are also shown.

Fig. 3 shows the calculated in situ behavior of each phase in the composite as well as the composite average. The calculated in situ stiffnesses for the β phase and the matrix are 61 and 89 GPa, respectively. These values compare very well to those reported in the literature [15]: 63 GPa for the β phase and 89 GPa for the BMG monolith. Similarly, the composite modulus predicted by the SCM model (81 GPa) is comparable to the value measured by the extensometer and that obtained in [15] (∼79 GPa).

The current results clearly show that it is the β phase that yields first during the loading of a BMG/β phase composite. A possible scenario that follows is that the load transfer to the BMG matrix after the yielding of the β phase and/or the stress concentrations generated at the intersection of slip bands in the β phase and the matrix/particle interface induce multiple shear bands in the BMG matrix. Such shear bands have indeed been observed in β phase/BMG composites subject to plastic strain [13–15]. The multiple shear bands in the BMG enhance its ductility and damage tolerance as a single, catastrophic shear band is avoided. It should be noted that contrary to its monolithic behavior, the BMG does appear to strain harden in the composite (see Fig. 3), which is likely related to generation of multiple shear bands in it. In the present study, calculations (not reported here) showed that agreement between the self-consistent model and experimental data was not possible without including strain hardening in the BMG. The details of the micromechanics of multiple shear band generation in β phase/BMG composites, however, are still unclear and subject to current investigations.

4. Summary

The compressive deformation of a new class of BMG matrix composites (with in situ formed dendritic precipitates called the β phase) was investigated with ND and SCM for the first time. Previous studies [15] had proposed

several deformation mechanisms for the β phase including stress-induced phase transformations, twinning and dislocation slip. The ND data conclusively showed that no detectable phase transformations were present as the β phase remained bcc during its deformation in both monolithic and composite forms. Furthermore, the low degree of texture development observed [28] suggested that twinning was not a dominant deformation mechanism. This left dislocation slip as the only active deformation mode in the β phase. This conclusion was also supported by the fact that the SCM calculations, that considered only dislocation slip in the β phase, showed reasonable agreement with the lattice plane specific diffraction data.

The successful fitting of model predictions with diffraction data allowed the deduction of the mechanical properties of the β phase, both as a monolith and inside a metallic glass matrix. It was shown that the β phase is highly anisotropic in the elastic regime (with an anisotropy ratio reaching 3.0), has a uniaxial yield point of about −600 MPa and plastically deforms by dislocation slip. The β phase was seen to largely retain these properties inside a metallic glass matrix. The SCM predictions also yielded insight into the load sharing behavior of the two phases in the composite. It was demonstrated that, upon loading, the β phase yields first (around −600 to −700 MPa), then starts transferring load to the matrix. The BMG matrix enters the "plastic" regime around −1400 MPa by presumably initiating multiple shear bands. It is speculated that the plastic deformation of the dendritic β phase somehow triggers shear banding in the matrix. The reader should be cautioned that the properties of the β phase were recently found to be very sensitive to processing conditions [17] and what is described in the present study applies only to as-cast specimens.

This study also developed a new self-consistent model that includes two phases where one (i.e., the BMG matrix) is described using a continuum-based J_2 flow theory and the other (i.e., the β phase) by the crystal-plasticity theory. In addition, the "traditional" single phase SCM was used with a new approach to determine the single crystal elastic constants of the β phase using the diffraction data obtained from a polycrystalline monolithic β phase sample.

Acknowledgments

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