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Diffraction strain measurements in a partially crystallized bulk metallic glass composite containing ductile particles

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Abstract

In situ diffraction experiments were performed with high-energy synchrotron X-rays to examine load partitioning and high-stress relaxation during uniaxial compression of a bulk metallic glass composite containing both ductile tantalum particles and crystallized matrix material. The tantalum particles yielded at an applied stress of −800 MPa, while the matrix precipitates remained elastic up to the maximum applied stress of −1250 MPa. The von Mises effective stress in the tantalum particles at yielding was 1500 MPa, well in excess of typical tantalum yield stresses, which is attributed to a combination of solid-solution strengthening and the inhibition of dislocation motion in the 1–2 μm particles. A series of constant crosshead-position measurements made at −1250 MPa suggested the possibility of room-temperature matrix relaxation under high applied loads.

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1. Introduction

Bulk metallic glass (BMG) composites exhibit excellent strength and some ductility due to the arresting of shear bands by the reinforcing phase [1–3]. There are, however, important differences in their mechanical behavior as compared to conventional metal matrix composites (MMCs) consisting of a metallic, ductile matrix containing stiffer but brittle reinforcement, e.g. Al/SiC. The main difference is that the matrix of BMG composites is brittle while that of most MMCs is ductile. This leads to different situations upon cooling from the processing temperature, where tensile thermal mismatch strains are developed in the matrix of both types of composites. These strains are easily relaxed by matrix plasticity in MMCs but build up elastically in the BMG matrix, thus decreasing the applied stress at which shear bands are formed upon subsequent mechanical loading. Second, upon mechanical loading, the matrix of both BMG composites and MMCs transfers some of its load to the stiffer reinforcement. In BMG-composites, however, it is

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the reinforcing metallic second phase that first exceeds its yield stress during loading, followed by brittle fracture of the matrix. In conventional MMCs, matrix plasticity typically occurs before brittle reinforcement fracture [4]. The load transfer evolution between the matrix and reinforcement after yielding is thus fundamentally different in the two types of composites.

BMGs are capable of partial crystallization, with a resulting structure that also exhibits improved mechanical properties due to the arresting of shear bands by the precipitated crystallites [5]. The crystallized matrix material often has mechanical properties similar to those of the parent matrix, e.g. high strength and relatively low stiffness [6,7], and can therefore be used as a diffraction 'marker' allowing for indirect estimation of the stress state in the matrix. The purpose of the present work is to measure by synchrotron X-ray diffraction the bulk lattice strains in both the metallic reinforcement and the crystallized matrix material of a BMG composite subjected to uniaxial compressive loading both below and above the limit where plastic deformation of the reinforcement occurs.

2. Experimental procedures

2.1. Sample preparation and characterization

A BMG with the commercial designation Vitreloy 106 (V106) and a nominal composition of $Zr_{57}Nb_5Al_{10}Cu_{15.4}Ni_{12.6}$ was used as the matrix material. The reinforcement consisted of tantalum particles with nominal diameters of less than 2.0 μm and a nominal volume fraction of 5%. The composite also contained a small amount of crystallized matrix, created in situ during fabrication by controlled cooling. The composite was fabricated in a process similar to that described in Ref. [1]. This method involves the mixing of metal particles into a molten BMG alloy using induction melting in a Cu crucible within a Ti-gettered atmosphere. Since V106 is not a very strong glass former, some partial crystallization (devitrification) can occur in the matrix during cooling. A parallelepiped compression sample was machined with dimensions $2.97 \times 1.47 \times 1.47$ mm for diffraction measurements during mechanical testing. Scanning electron microscopy (SEM) was performed to characterize particle size, volume fraction, and distribution using digital image analysis techniques on a polished section of the composite.

2.2. Diffraction experiment

Diffraction measurements were performed at the DuPont-Northwestern-Dow Collaborative Access Team (DND-CAT) at Sector 5 of the Advanced Photon Source (Argonne National Laboratory). In situ uniaxial compression testing was performed using a tabletop mechanical testing machine. The general set-up for these experiments has been described in detail elsewhere [8]. The stress on the composite was varied between 0 and −1250 MPa in steps of 50–250 MPa, and was held constant during the 600-s X-ray exposures. At −1250 MPa, the load frame was maintained at a constant crosshead position during four sequential exposures. The sample was irradiated with a monochromatic 65 keV ($\lambda = 0.019$ nm) X-ray beam with a square cross-sectional area of 0.5 mm × 0.5 mm. The Debye–Scherrer diffraction cones from the two crystalline phases present in the diffraction volume of ~0.37 mm^3 were recorded using a CCD camera. The CCD camera was situated at a distance of 652 mm from the sample and had a 132 mm diameter detector with 16 bit intensity readings over an orthogonal array of 64.4 μm × 64.4 μm pixels. A stress-free iron powder standard was attached to the sample for calibration purposes.

3. Results

An SEM micrograph of the composite is shown in Fig. 1, clearly showing the smaller Ta particles and the larger crystalline precipitates. The Ta particles are both smaller and more rounded than the crystallized matrix precipitates, with some agglomeration visible. The Ta particles range in size between ~0.5 and 2 μm, and exhibit a volume fraction of ~6%. The other crystalline phase is likely one of the many intermetallic phases that

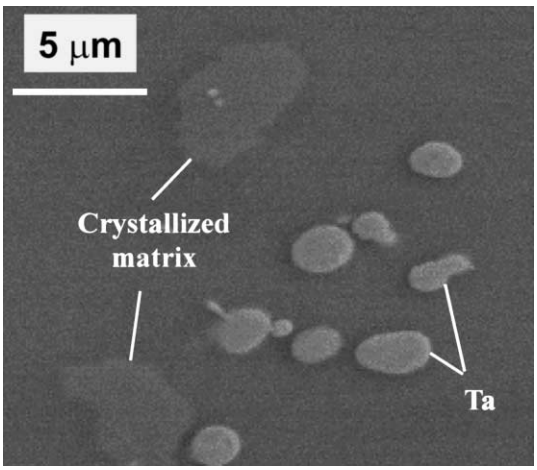

Fig. 1. SEM micrograph of the V106 BMG composite.

can form when BMG alloys devitrify [5,6], but was not crystallographically identified. This phase is 2–10 μm in size, and is present in smaller quantities, on the order of 1–2 vol.%. The macroscopic stress–strain curve during compressive loading, calculated from the deflection-adjusted crosshead displacement, is shown in Fig. 2. The applied stress vs. elastic lattice strain responses of both the Ta particles and the crystallized matrix are shown in Fig. 3(a) and (b), neglecting residual strains. The time evolution of the macroscopic sample stress and the crystallized matrix lattice strains during the 45 min hold at maximum load are shown in Fig. 4.

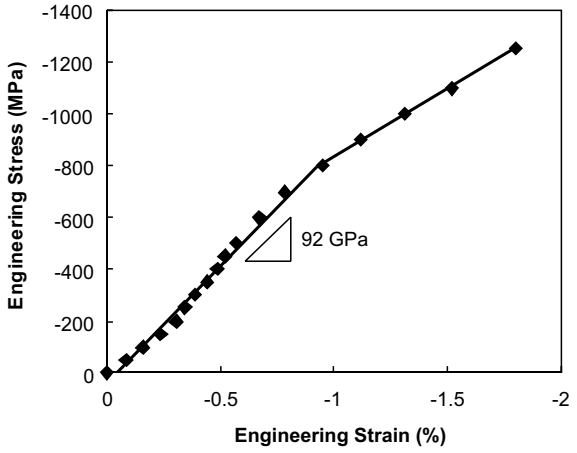

Fig. 2. Macroscopic stress—strain curve of the V106 BMG composite. Lines are to guide the eye.

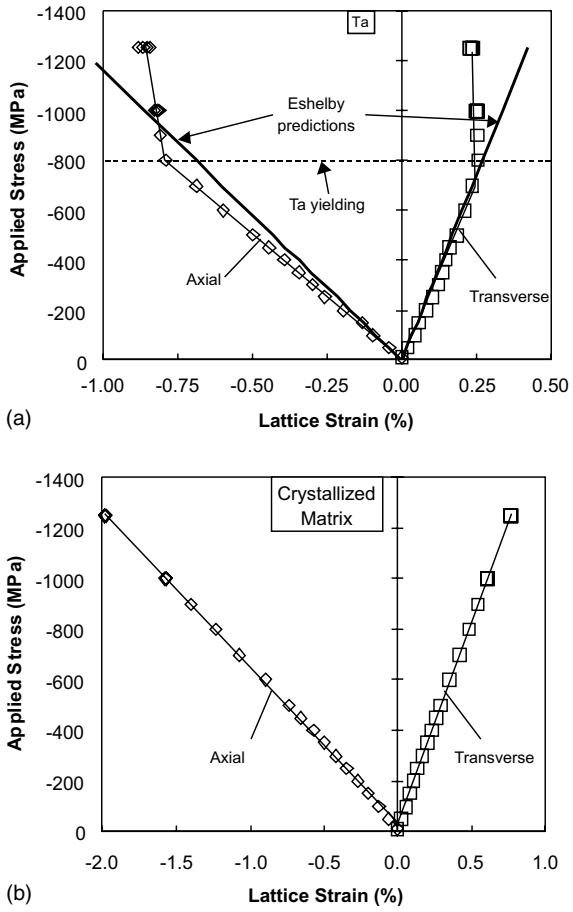

Fig. 3. (a) Applied stress vs. elastic lattice strain for the Ta particles. Strain error bars are ±0.025%. Lines through strain data are to guide the eye. (b) Applied stress vs. elastic lattice strain for the crystallized matrix precipitates. Strain error bars are ±0.005%. Lines are to guide the eye.

4. Discussion

4.1. Macroscopic composite behavior

Fig. 2 shows the macroscopic loading curve of the composite. The slope in the elastic region up to a stress of −800 MPa is 92 GPa, a slight increase above the Young's modulus of 84.7 GPa for the pure V106 matrix [1]. The Young's modulus pre-

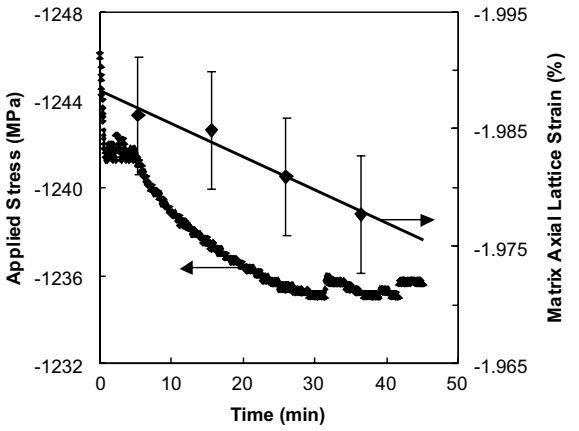

Fig. 4. Time dependence of macroscopic stress and crystallized matrix axial lattice strain at constant crosshead position. Line is to guide the eye.

dicted by the Eshelby equivalent inclusion method [4], assuming 94 vol.% V106 and 6 vol.% Ta, is 89 GPa, a satisfactory agreement given experimental errors and the presence of the crystallized matrix. The inflection in the macroscopic stress–strain curve beyond a stress of −800 MPa is attributed to yielding of the tantalum particles.

4.2. Tantalum lattice strains

Fig. 3(a) shows the applied stress vs. lattice strain curves for the Ta particles in both the axial and transverse directions, i.e. parallel and perpendicular to the loading axis, as measured using the {1 1 0} reflection. Also shown are the Eshelby elastic strain predictions, which are in reasonable agreement with the measured strains in the elastic range. As the measured strain curves exhibit somewhat lower slopes than predicted, more load transfer to the Ta phase is occurring than expected. This may be explained by the presence of crystallized matrix material with a stiffness lower than the amorphous V106 matrix. This would reduce the effective modulus of the matrix below 84.7 GPa, resulting in an increase in stiffness mismatch with, and load transfer to, the Ta particles.

The lattice strain curves in Fig. 3(a) remain linear to an applied stress of −800 MPa, above which there is a sharp upward bend in the curves, indicative of reduced load transfer to the particles. Such an inflection is expected when particles yield and begin to flow plastically in a matrix that remains elastic. What is unexpected in this case is the very high stress at which the inflection occurs, which corresponds to a von Mises effective stress of ~1500 MPa in the Ta particles (calculated from the measured lattice strains using the assumption of isotropic behavior). This value is well in excess of the yield stresses observed in commercially pure Ta, i.e. 165–220 MPa [9]. We attribute this large increase in strength to two effects: the very small size of the particles (<2 μm) limiting dislocation motion [10], and solid-solution strengthening caused by the presence of matrix elements within the particles [11]. Energy dispersive spectroscopy (EDS) measurements of the elements within the Ta particles indicated the presence of copper, nickel, and zirconium, although quantitative measurements were not made.

4.3. Lattice strains and stress relaxation in crystallized matrix

Fig. 3(b) shows the lattice strain response for the crystallized matrix precipitates, in both the axial and transverse directions. The strain response is linear at all applied stresses, indicative of elastic behavior without any yielding, as is frequently seen in intermetallics with complex structures. The slope of the applied stress vs. lattice strain curve in the axial direction is 63 GPa. This value is somewhat higher than the Young's modulus which would be measured in the bulk, due to load transfer to the slightly stiffer matrix and the Ta particles, but is in rough agreement with measured modulus values for material crystallized from similar BMGs [6].

Fig. 4 shows the macroscopic stress response and crystallized matrix lattice strain response during the hold at constant crosshead position at a nominal stress of −1250 MPa. After an initial near-instantaneous drop of 4 MPa as the load frame settled, the applied stress on the composite gradually decreased a further 7 MPa over a period of 45 min. The effect of this decrease in the composite stress is seen in the axial and transverse crystallized matrix strains, which relax in a manner consistent with the decreasing composite stress.

We believe that this decay of the composite stress at constant crosshead position may be due to relaxation of the partially amorphous matrix, although we recognize that relaxation of the load frame itself cannot be ruled out as a contributing factor. Ex situ testing of a steel sample did not show any relaxation effects, but further experiments with strain gauges are needed to obtain definitive results.

5. Conclusions

To the best of our knowledge, this is the first time that in situ diffraction strain measurements were performed on a BMG composite containing both ductile reinforcing particles and crystallized matrix material during uniaxial mechanical loading, thus allowing independent assessment of the stresses in the stiff reinforcement and the compliant matrix. Up to an applied stress of −800 MPa, the measured Young's modulus of the composite and elastic lattice strains in the tantalum particles were in agreement with elastic Eshelby predictions. At −800 MPa, the tantalum particles exhibited yielding at a very high von Mises stress of 1500 MPa, which was attributed to both limited dislocation motion and solid-solution strengthening. Lattice strains measured in the crystallized matrix material show elastic behavior at all applied stresses and suggest possible relaxation of the BMG matrix at high applied loads.

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