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Effect of reinforcement size and orientation on the thermal expansion behavior of metallic glass reinforced metal matrix composites produced by gas pressure infiltration

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ABSTRACT

Many efforts were made during the last years to utilize the superior properties of metallic glasses as reinforcements in metal matrix composites and several investigations were carried out to characterize composites' mechanical properties. Since there is a complete lack of knowledge about their thermal properties, this work focuses on the thermal expansion behavior of metallic glass reinforced metal matrix composites produced via gas pressure infiltration. The aluminum alloy AlSi12 was used as matrix material. Metallic glass $Ni_{60}Nb_{20}Ta_{20}$ flakes with different size ranges were taken as reinforcement and volume fractions in the range of 29–44% were studied. X-ray micro computed tomography (μCT) measurements were performed to investigate the reinforcement 3D-structure within the composite. The thermal expansion behavior depending on flake orientation within the composite was investigated by dilatometric measurements. Four thermal cycles were carried out between room temperature and 500 °C at a constant heating and cooling rate of 5 °C/min. Results show that thermal strain rather depends on flake size and orientation than on volume fraction. Further, the composites exhibit a distinct anisotropic behavior for the thermal expansion coefficient (CTE) due to a layered flake structure within the composite. Finally determined CTEs were compared with several thermo-mechanical models to study the underlying mechanisms of the composite behavior.

1. Introduction

Metal matrix composites (MMCs) offer the advantage of adaptability of desired material properties for specific applications. Therefore, they are applied e.g. in electronic applications instead of single alloys when high thermal conductivity and low thermal expansion are required [1,2]. Although MMCs offer better high temperature properties than the unreinforced alloy, they show a complex behavior when heated due to different thermal and mechanical properties of the single components [3], like Young's moduli and thermal expansion coefficients (CTE). Materials normally used as reinforcements, like ceramics, usually exhibit higher Young's moduli and lower coefficients of thermal expansion (CTE) than the commonly used matrix materials. This leads to formation of internal stresses due to the mismatch in thermal expansion and elastic properties of the components. The lower CTE of the reinforcement phase further causes a reduced thermal expansion of the composite [4] as desired for electronic applications. Further, the thermal behavior of composites also depends on reinforcement volume fraction, the arrangement of the reinforcement within the composite and on defects, like microscopic pores or cracks, originating during composite processing [5]. As an example: Particle reinforced MMCs exhibit nearly isotropic behavior with lower CTEs than the unreinforced matrix [6], while MMCs with fibrous reinforcements show accumulated macroscopic plastic deformation [7] with significant anisotropy depending on fiber orientation [8–11]. Hence, comprehensive investigations on understanding the numerous influencing factors on the thermal expansion behavior of different composites are essential for an extension of the application range.

Investigations on the thermal expansion behavior of composites based on AlSi-alloys with reinforcement of SiC-particles and various reinforcement volume contents were performed by Ref. [6]. They revealed that the thermal expansion of composites with isolated particles can be described by the thermal expansion behavior of the AlSi-matrix [6]. The CTE(T)-curve of these composites exhibits the same shape as for the single AlSi-alloy with the same decrease in CTE at high temperatures due to increasing solubility of silicon in aluminum with increasing temperature [12]. The increase of reinforcement volume fraction just leads to lower levels of CTE-values [6,13]. Their investigations on a composite consisting of an infiltrated SiC sintered preform further showed that the composite's CTE at high temperatures

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matches the CTE of bulk SiC due to the rigidity of the preform [6].

Over the last years, several studies dealt with the mechanical properties of MMCs with metallic glass reinforcements and showed their high potential concerning enhanced Young's modulus and strength [14–18]. To date no investigations were performed on the thermal properties of these composites although the difference in thermal expansion coefficients and correlated internal stresses are also present in MMCs reinforced with metallic glass. Therefore, it is inevitable to characterize their thermal properties in detail to obtain a comprehensive view on materials processing-structure-property-relations to develop new areas of application. In this study, the thermal expansion behavior of several metallic glass flake reinforced composites with different size ranges produced by gas pressure infiltration is investigated. Thermal expansion characterization is carried out with a special focus on the influence of flake size and flake orientation. The composites will be examined by means of dilatometry during four thermal cycles between room temperature and 500 °C. CTEs determined by these measurements will be further compared with several thermo-mechanical models.

2. Experimental procedure

2.1. Material

As matrix material, the unrefined aluminum alloy AlSi12 (EN AC-44200) was utilized. Metallic glass $Ni_{60}Nb_{20}Ta_{20}$ with a crystallization temperature of T_x = 723 °C [19] was used as reinforcement in the studied composites. The metallic glass was produced by Fraunhofer Institute IFAM (Dresden, Germany) as melt spun ribbons with a mean thickness of 50 µm. The reinforcement flakes were prepared by ball milling these ribbons. The milled ribbons were sieved to separate different flake size ranges. The different size ranges are listed in Table 1.

The composites investigated in this study were fabricated by conventional gas pressure infiltration of the respective flake size and aspect ratio ranges (cf. Table 1). At the beginning of the infiltration process, the processing chamber was evacuated to 0.08 mbar and purged with argon to remove remaining oxygen. The processing chamber was heated with constant heating rate of 5.5 °C/min to the maximum processing temperature of 660 °C. This maximum temperature was held for 2 h during the melting of the matrix raw material and homogenization of the melt. Infiltration was achieved by argon with a pressure of 40 bar. This pressure was held during cooling down the processing chamber with constant cooling rate of 5.5 K/min. Previous investigations including X-ray diffraction measurements proved that it is possible to produce composites using these processing parameters without crystallization of the metallic glass since the maximum processing temperature is lower than its crystallization temperature [15]. For a detailed processing technique description, processing parameters and infiltration device, we refer to Refs. [15] and [18].

Investigated flake size ranges and aspect ratios, measured density and calculated average volume content of reinforcements within the composite are given in Table 1. Since smaller particles lead to smaller inter-particle distances [20], increasing volume fractions of metallic glass within the composites can be realized with decreasing flake sizes.

Table 1
Flake size and aspect ratio ranges used in this study and resulting average composite density and reinforcement volume fraction.

Composite	Flake size range (µm)	Flake aspect ratio	Density (g/cm^3)	Reinforcement volume fraction (%)
A	100–200	2–4	6.03 ± 0.07	43.1 ± 0.9
B	200–600	4–12	5.59 ± 0.13	37.6 ± 1.7
C	600–2000	12–40	4.93 ± 0.09	29.1 ± 0.9

It is not possible to adjust reinforcement volume fractions to a defined level due to processing restrictions.

2.2. Structural analysis via X-ray micro computed tomography (µCT)

As shown in [15] for large flake sizes, metallic glass flakes within the composites are supposed to be oriented perpendicular to the infiltration direction of the melt into the mold during processing due to the plate-like shape of the flakes and the high aspect ratios. To prove this assumption for the composites of this study, 3D X-ray micro computed tomography (µCT) investigations were carried out on selected samples. For this purpose, an Yxlon Precision computer tomograph with a flat panel detector from Perkin Elmer and a tungsten target were used. Acceleration voltage was fixed to 180 kV and the target current was 0.02 mA. The scan consisted of 2400 projections during a 360° sample rotation along the vertical axis with an integration time of 700 ms per projection. Scan resolution was 4.31 µm/voxel. The image data was rendered using the image processing program Avizo.

Orientation of the flakes was analyzed using the aligned and cropped µCT data. Therefore, orientation of the normal vector in each voxel (c.f. Fig. 4(a)) was calculated using a method based on the structure tensor [21]. This method was earlier implemented in the open source project *Composight* [22] for fibrous materials [23] and was modified for application with planar structures. The algorithm is mostly equal to that for fibers, but the direction of a normal vector of a plane is given by the largest eigenvalue of the structure tensor and its corresponding eigenvector instead of the lowest eigenvalue as for fibers. Results of this orientation analysis are given as orientation histograms showing the number of voxels in a µCT image with an absolute deviation angle from a defined axis.

2.3. Dilatometer measurements

Investigations on the thermal expansion behavior of the metallic glass reinforced composites were carried out in a common push-rod dilatometer type DIL 805A/D from Bähr-Thermoanalyse GmbH (Hüllhorst, Germany). The device consists of a fixed push-rod and a variable rod which is connected to a linear variable differential transformer (LVDT) for measuring sample dilatation. Heating is realized by an induction coil. A separate perforated coil for gas quenching is used in parallel. Further information about the dilatometer setup are given in Ref. [24]. In this investigation, parallelepiped samples of each composite with dimensions of 7 × 4 x 4 mm^3 were taken from the infiltrated material by electrical discharge machining. Matrix material samples with the same temperature history as the composites were taken as reference. Edges of the samples were deburred to ensure adequate contact of the sample with the push-rods inside the dilatometer. Since processing restrictions and flake aspect ratios are expected to lead to a layered flake arrangement within the composite with flake orientation perpendicular to the infiltration direction (see Sections 2.2 and 3.1), samples were taken from the infiltrated material with different orientations concerning the infiltration direction. Fig. 1 illustrates the layered reinforcement structure within the composite including exemplary samples and testing directions. As depicted, samples were studied in longitudinal direction along flake orientation (along y-axis) and in transverse direction perpendicular to flake orientation (along z-axis).

The temperature-dependent elongation of the samples was measured along the major axis of each sample (see Fig. 1). Four thermal cycles between room temperature and 500 °C with constant heating and cooling rate of 5 °C/min were recorded for each sample. The thermal expansion was measured during heating and cooling. The experiments were performed in an inert helium atmosphere to avoid oxidation of the composites. All experimental data was corrected by a reference measurement with a platinum sample to eliminate any thermal length change effects of the testing device as required by Ref. [25].