DAMAGE EVOLUTION AND ANISOTROPY IN FREEZE CAST METAL/CERAMIC COMPOSITES: AN IN-SITU SEM ANALYSIS

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ABSTRACT

Innovative metal/ceramic composites produced by melt infiltration of innovative ceramic preforms prepared by a freeze-casting technique are examined in this study. These composites exhibit a characteristic hierarchical structure: On a mesoscopic length scale, lamellar domains with sizes up to several millimetres are observed. These domains are composed of alternating ceramic and metallic lamellae with thicknesses ranging from 20 to 100 µm. The aim of the present study is to investigate the domain level elastic and elastic-plastic flow behavior of the composite processed under different conditions. Elastic properties are determined with the help of a special ultrasonic method while detail in-situ SEM analysis is carried out to thoroughly investigate the damage evolution under compressive load. Results show that the individual domains show pronounced anisotropy in their mechanical properties. The composite is stiffest and strongest along directions parallel to the freezing direction. When loaded along freezing direction, it behaves as a brittle solid, showing limited or no plasticity, while when loaded perpendicular to the freezing direction, the behavior is controlled by the metallic component, showing extensive plasticity.

1. INTRODUCTION

Metal matrix composites (MMC) are technically important because of their high specific stiffness and strength, high wear and fatigue resistance and enhanced high temperature properties etc [1, 2]. Because of their potential applications, MMCs with different properties have been developed in last few decades. Good reviews of the different routes for MMC fabrication are presented in [3] and [4]. Continuous research is however going on to fabricate composites having novel property profiles and more efficient and economic processing routes. A new possibility has recently been opened by the availability of ceramic preforms processed by freeze-casting of ceramic suspensions. Details about the freeze casting process can be found in Refs. [5, 6, 7]. Ceramic preforms produced by freezing of water-based suspensions (slurries) have a typical hierarchical lamellar domain structure. The size and internal structure of these domains are controlled by the freeze casting parameters [5, 8]. Preforms produced this way have excellent permeability for liquids and gases along with acceptable mechanical strengths and they are suitable for the fabrication of metal/ceramic composites by infiltration of liquid metal [5, 9]. This way it is possible to fabricate composites with ceramic contents in the range of about 30 to 70 vol%. This intermediate range is of particular interest since conventional particle- or fiber-reinforced composites typically contain either a relatively low (5–30 vol. %, e.g. [4]) or a fairly high (50–80 vol. %, e.g. [10, 11]) reinforcement content due to processing constraints.

The structure of the composite materials produced by infiltration of liquid metal in freeze-cast ceramic preforms has resemblance with that of lamellar two phase alloys (e.g. γ – TiAl alloys). In these alloys it is observed that the domain level mechanics control their mechanical properties [12]. The aim of the present study is to investigate the domain level elastic and elastic-plastic flow behavior of the composite material processed under different conditions. A first study has already shown that in these composites, the individual domains show pronounced anisotropy in their elastic properties [13]. In-situ SEM analyses under compressive load were additionally performed to directly investigate the elastic-plastic flow behavior and damage evolution in single domain samples having different orientations.

2. EXPERIMENTAL PROCEDURE

2.1 Specimen material

Alumina preforms with porosities of about 56 vol. % were produced at Institut für Keramik im Maschinenbau (IKM) at Universität Karlsruhe, Karlsruhe, Germany, via freeze-casting of a ceramic suspension and subsequent sintering. Details about the freeze-casting process and the process parameters can be found in a previous publication [13]. Preforms with nominal dimensions of 10 × 44 × 66 mm³ were infiltrated with a eutectic aluminium–silicon alloy (Al–12Si) at the Casting Technology Center at Aalen University of Applied Sciences, Aalen, Germany, using a squeeze casting technique. Before infiltration the preforms were preheated to 800°C and the press was heated up to 400°C. After squeeze casting, the infiltrated samples were heated to 450°C, held at that temperature for 2 hours and then subsequently furnace cooled. To investigate the influence of interfacial properties between the ceramic and metallic component on the mechanical behavior of the composite material, some preforms were coated with Cu before melt infiltration. Commercially available coating solutions were used. Before applying the coating, the substrate surface was activated with Pb/Sn solution containing hydrochloric acid. For coating, the preforms were clamped to a mounting plate made of teflon and the Cu solution was manually pressed into the porous preform with a stamp under appropriate conditions. Some of the Cu coated preforms were further heated to 1150°C to allow Cu to completely oxidise to Cu_2O. Finally the Cu and Cu_2O-coated ceramic preforms were infiltrated with Al-12Si melt by squeeze casting as discussed before.

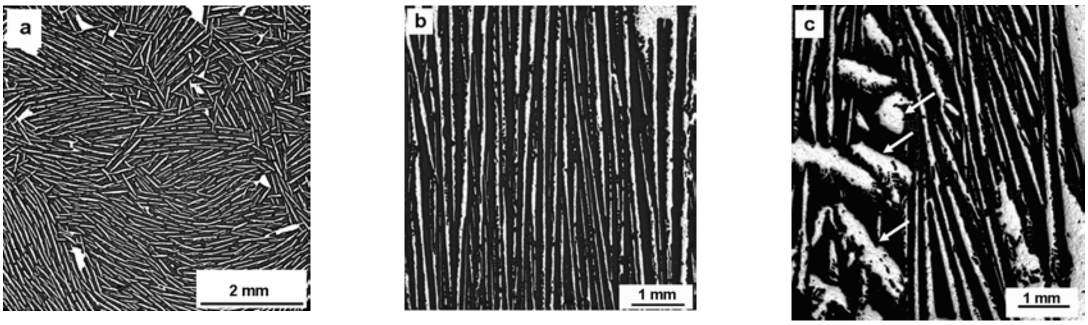

Figure 1: Microstructures of the three orthogonal faces of the composite; (a) face perpendicular to freezing direction and (b - c) faces parallel to the freezing direction

Fig. 1 shows typical microstructure on the three orthogonal faces of the composite. In these light-optical micrographs, the ceramic component appears dark while the metallic alloy appears brighter. Fig. 1a clearly shows the domain structure on the face perpendicular to the freezing direction, with individual domains composed of alternating metallic and ceramic lamellae. Fig. 1b and 1c show the microstructures of the faces parallel to the freezing-casting direction. It can be seen that the lamellae are predominantly oriented parallel to the freezing direction and stretch over the whole thickness of the material. The partly irregular regions marked with arrows in Fig. 1c, indicate that the lamellae are neither perfectly planar nor perfectly aligned parallel to the macroscopic direction of heat removal. For elastic and elastic-plastic analysis of individual domains, small rectangular parallelepiped samples with dimensions between 1.8 to 2.6 mm were produced from the composite plates via wire cutting using 220 µm thick, diamond-coated steel wire. The edges of the final parallelepipeds were always aligned parallel to the edges of the composite plates and they had arbitrary domain orientations. As is apparent from Fig. 2, the lamellae in such samples are fairly parallel over the whole sample and their orientation can be described by the angle α (angle between lamellae and specimen axis 2).

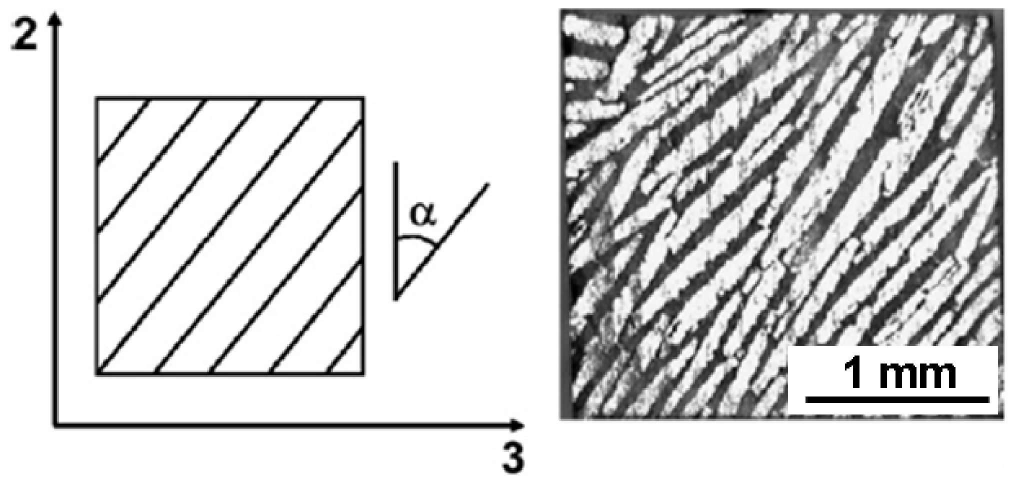

Figure 2: Definition of the angle α, marking the orientation of a single domain composite specimen with respect to direction 2 of the specimen coordinate system [13]

2.2 Determination of longitudinal elastic properties by phase comparison

The longitudinal elastic constants of the single domain samples were determined by measuring the velocity of continuous, sinusoidal, elastic waves. This was done by a special phase comparison method. Detail theoretical background of this method can be found in [14] while thorough information about the experimental procedure is presented in Ref. [13].

2.3 Determination of elastic-plastic flow behavior and damage mechanism by in-situ compression test

The elastic-plastic flow behavior and damage mechanism of the single domain samples were determined under compressive load. The compression tests were carried out using a miniature mechanical testing machine manufactured by Kammrath & Weiss GmbH, Dortmund, Germany. The photograph in Fig. 3 shows a single domain sample placed between the punches in the miniature test setup. The compressive load was applied with the help of two flat punches made of hardened steel. 20 µm thick commercially available Al foil was used between the samples and the punches to minimise the sample friction. The total strain was measured with the help of an in-built LVDT (Linear Variable Differential Transformer). All experiments were carried out at a fixed crosshead velocity of 2 μms^{-1}. In-situ analyses was carried out in a scanning electron microscope (SEM) of type Zeiss EVO 50 to observe the damage evolution in the material when loaded. This was done by carrying out the compression test while the miniature setup along with the sample was placed inside the microscope. Damage evolution was directly observed by stopping the test at different loads to take SEM micrographs.

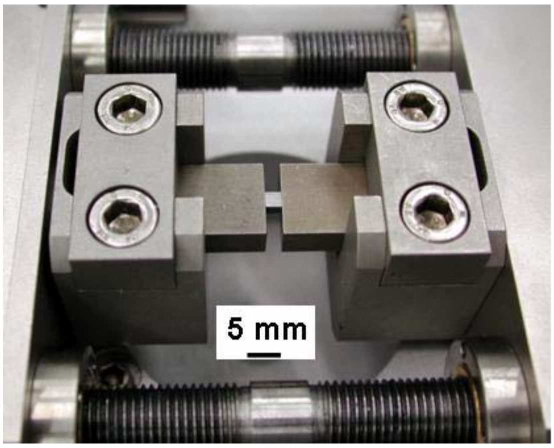

Figure 3: Single domain sample placed between two flat punches in the miniature mechanical test device

3. RESULTS AND DISCUSSION

Figure 4 shows the longitudinal elastic constant of the single domain samples (ρV^2 in GPa, where ρ is the mass density and V is the elastic wave velocity) plotted versus the angle β (where $\beta = \alpha$ for $V = V_2$ and $\beta = 90 - \alpha$ for $V = V_3$).

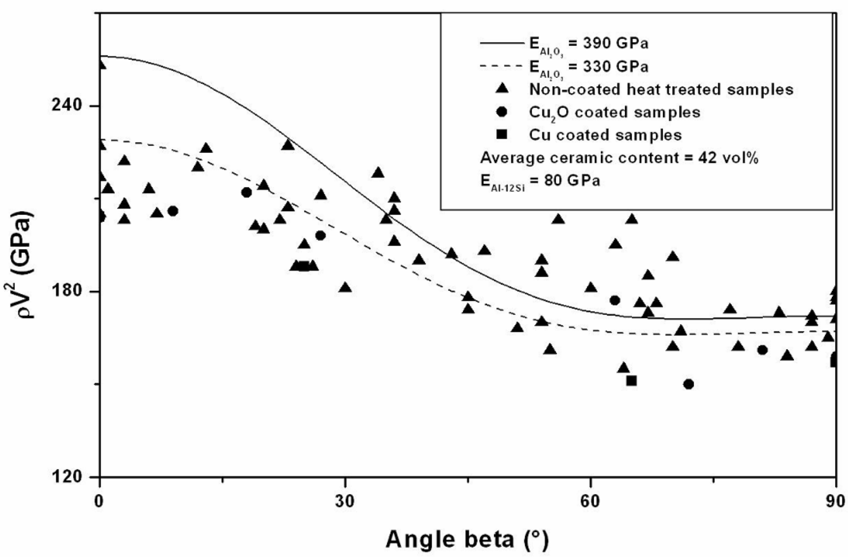

Figure 4: Effect of domain orientation on the quantity ρV^2 for single domain samples

A first account of this domain level elastic anisotropy can be found in [13]. The substantial scatter in the results for individual samples is due to the small probed volume and a very small sample size. The ordinate values obtained from two extremes of the plot (corresponding to $\beta = 0°$ and $\beta = 90°$) are the elastic constants C'_{33} and C'_{22} of the lamellar domains in the domain coordinate system, where direction $2'$ is perpendicular to the lamellar plane and direction $3'$ is parallel to it. C'_{33} is about 215 GPa and C'_{22} is about 165 GPa. The third elastic constant, C'_{11} (along the freezing direction), is simply obtained from the average velocities V_1 of the single-domain samples. The result for this elastic constant is C'_{11} = 227 GPa and as expected this is close to the C'_{33} in Fig. 4 corresponding to $\beta = 0°$. The two lines show the correlation of the experimental results with the theoretical model for 3D laminates compiled by Torquato [15] and using the transformations rules summarised by Berthelot [2]. For these calculations, the average ceramic content in the composite samples was calculated to be 42 vol % (calculated from the mass-dimension measurement and assuming no porosity) and the Young's modulus of Al-12Si was measured to be 80 GPa. The Young's modulus of the ceramic preform used was unknown and a value of 390 GPa was taken from handbook data for bulk alumina [16]. The resulting match between the theory and experimental results is very poor (solid line in Fig. 4) at low domain orientations. A better match is obtained when a value of 330 GPa is taken as the Young's modulus of alumina (dashed line in Fig. 4), probably because of the fact that the presence of pores and other structural irregularities in the ceramic preform reduces its stiffness significantly from that of the bulk value.

Figure 5 shows stress-strain plots of single domain samples with the load applied along the freezing direction. For comparison, data for a sample without any coating and one

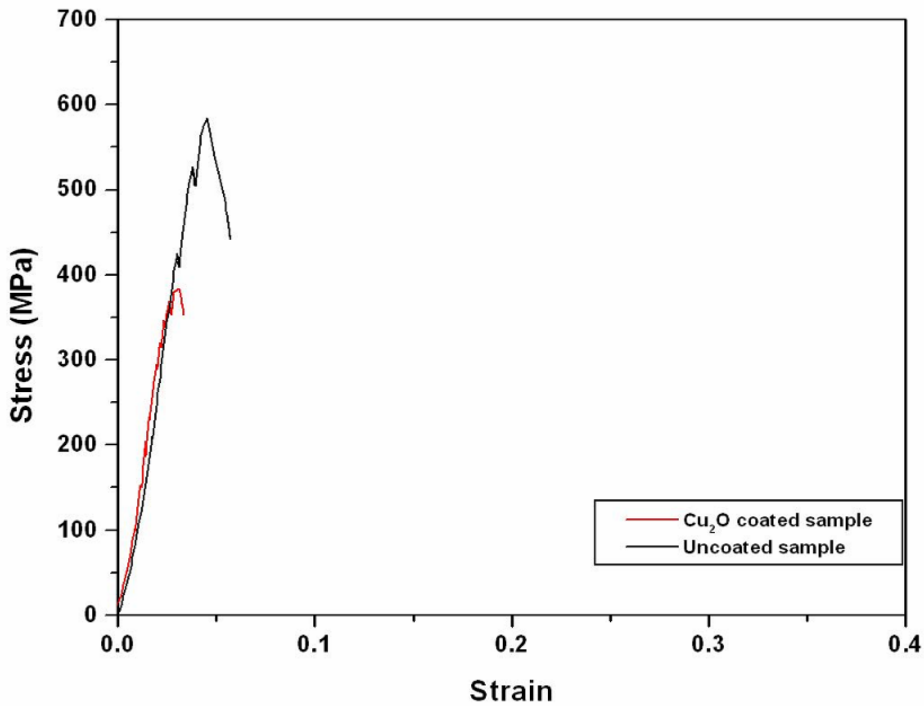

Figure 5: Stress-strain curve for single domain samples compressed along the freezing direction

Cu_2O coated sample are shown. The drops in the stress-strain plot correspond to the already mentioned "break points" for in-situ SEM analysis. In both the samples, the failure takes place in a brittle manner, with a sudden release of the stored energy. The compressive strength of the Cu_2O coated sample is significantly less than the uncoated sample. Figure 6 shows the failure mode for the uncoated samples loaded along the freezing direction. The direction of load application is visualized by the two arrows

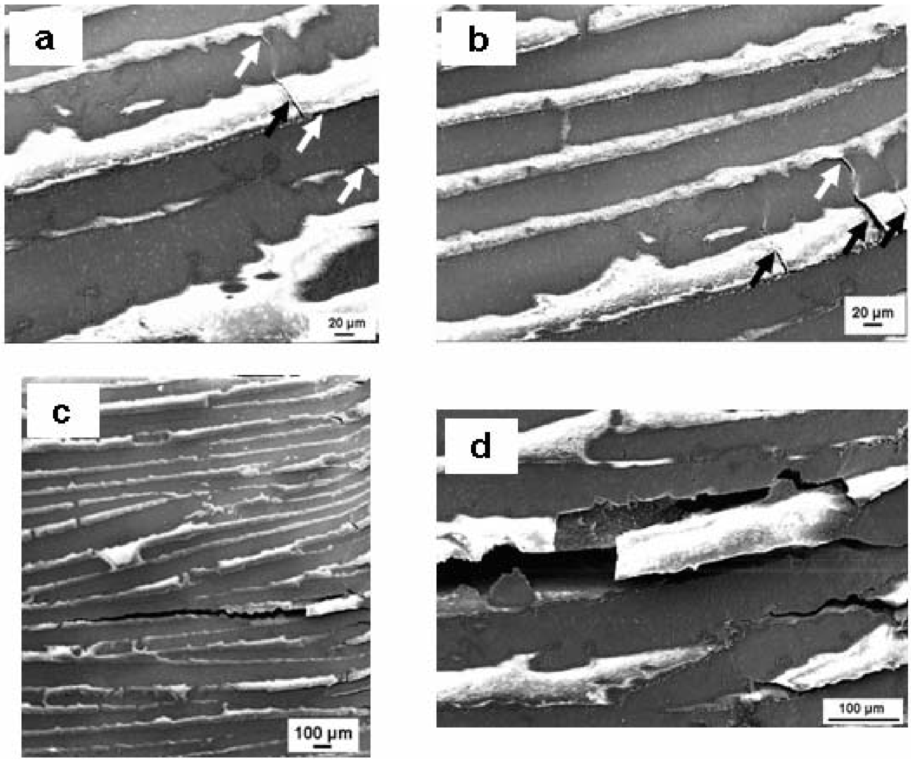

Figure 6: SEM micrographs showing the damage evolution in one uncoated single domain sample at different loads when compressive loaded along the freezing direction (= horizontal direction); (a) initial transverse cracking within the ceramic lamellae, (b) generation of more transverse cracks within ceramic lamellae at higher load and propagation of such cracks into the metallic alloy, (c) propagation of one longitudinal crack through one ceramic lamella and hence causing catastrophic failure of the sample and (d) enlarged view of the region close to the main crack showing ceramic crushing and longitudinal splitting of the metallic lamella

in Fig. 6. In these electron micrographs, the ceramic component appears bright because of the charging of the isolating ceramic regions while the metallic regions are darker. The failure mechanism is complex and several different steps are involved depending upon the individual properties of the ceramic and the metallic alloy and the interface. Moreover, because of the heterogeneous structure of the ceramic preform and thus the final composite, the structural variation from sample to sample is enormous, resulting in gross variation of the mechanical properties. Cracking begins within the ceramic lamellae as transverse cracks and then propagate into the metallic alloy (marked by arrows in Fig. 6a). Localised debonding at the metal-ceramic interfaces is also observed. With further loading more transverse cracks generate within the ceramic lamellae. Because of the ceramic cracking and subsequent load redistribution, the localised stress in the metallic alloy increases, which causes the transverse cracks in them to propagate further (shown by arrows in Fig. 6b). With further loading, one large crack propagates longitudinally through one ceramic lamella and the sample fails catastrophically (Fig. 6c). The damage development is not homogeneous throughout the sample, suggesting that because of the structural heterogeneities in the freeze-cast preform the stress concentration in a localised region causes the ceramic lamellae in that region to fail. Figure 6d shows a micrograph taken from the region in the vicinity of the main crack.

Apart from gross ceramic crushing, longitudinal splitting is observed within the metallic lamellae, suggesting that the debonding shear stress (τ_d) is greater than the ultimate shear stress of the metallic alloy (τ_{mu}). To have a closer look into the interface region of the uncoated sample, one sample damaged under compressive load was torn apart and detail SEM analysis was performed. Figure 7 shows the corresponding micrograph. The top region of the micrograph shows a broken ceramic lamella with most of the grains showing the features of intergranular crack propagation. The lower region of the micrograph shows honeycomb-shaped metallic residue on the surface of this ceramic lamella. This indicates very good bonding between the two components, substantiating the already made statement that the interfacial shear strength is higher than that of the metallic alloy.

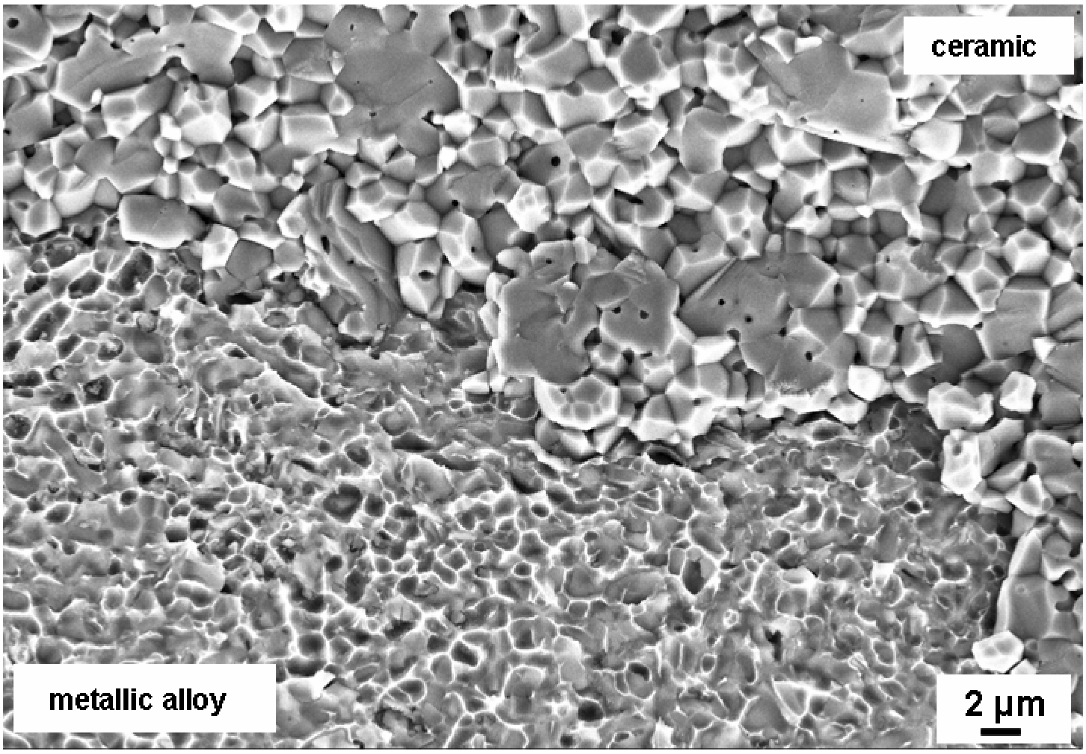

Figure 7: SEM micrograph showing the interfacial region

Spowart and Dève [17] have summarised the different failure modes of fiber reinforced MMCs loaded under compression along the fiber direction. Fiber kinking (in case of poor fiber alignment or high fiber strength) and fiber crushing (for well aligned fibres or low fiber strength) have been identified as the dominating modes. No trace of ceramic kinking could be observed in the material under study. Hence, ceramic crushing, due to the inherently low strength of the ceramic preforms after freeze casting seems to be the dominant failure mode when loaded along the freezing direction. A coating of Cu_2O on the ceramic probably weakens the inherently strong interface, hence reducing the interfacial shear strength. That would explain the lower compressive strength observed in these samples.

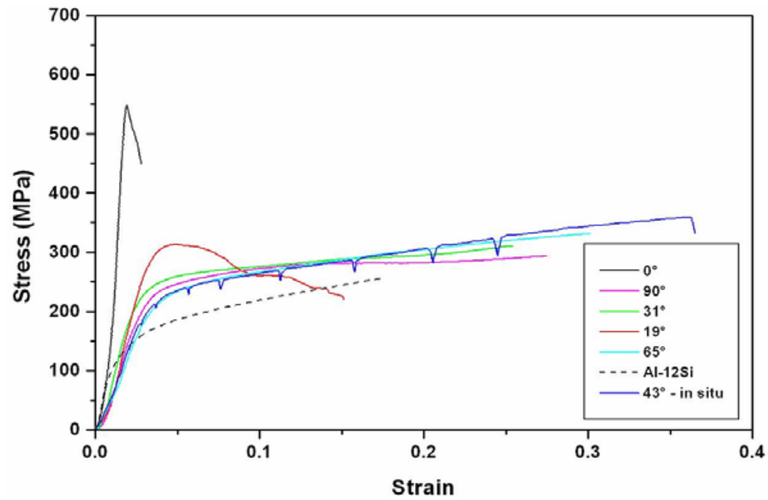

Figure 8: Compressive stress-strain plot for single domain samples compressed transverse to the freezing direction: effect of domain orientation

Figure 8 shows the compressive stress-strain plots for single domain samples having different domain orientations and compressed along directions orthogonal to the freezing direction. To compare with the behavior of the unreinforced metallic alloy, the compressive stress-strain curve for Al-12Si has also been added in the same figure.

When loaded along 0° to domain orientation, the composite behaves like a brittle solid, which is similar to that along the freezing direction. The material is also strongest along this direction. As the domain orientation increases, the metallic alloy plays a more dominant role and at orientations more than 30°, the behavior becomes fully metallic alloy controlled. For domain orientations within 30° - 90°, there is no perceptible change in the compressive stress-strain behavior. Because of the presence of the rigid ceramic lamellae, in the samples undergoing extensive plasticity the strength increases by a factor of about 1.3 with respect to the metallic alloy. The sample having about 43° domain orientation was compressed in-situ to observe the damage mechanism. The corresponding micrographs are shown in Fig. 9.

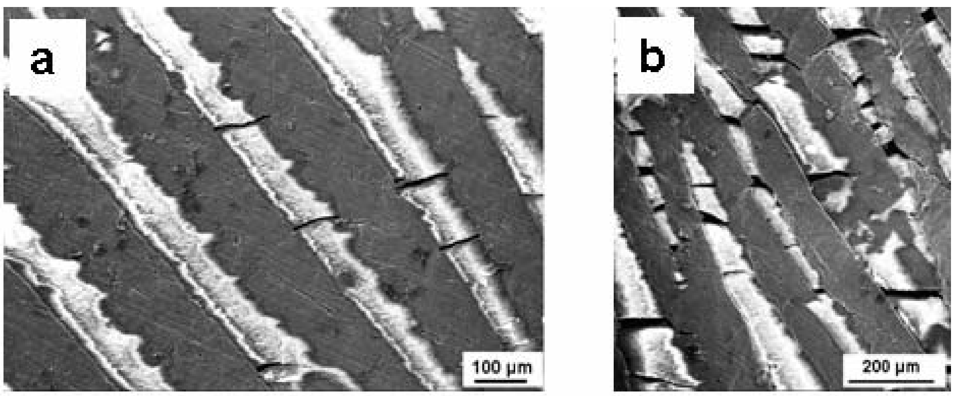

Figure 9: SEM micrographs showing the damage evolution when compressed at 43° to the domain orientation in a single domain uncoated sample (Compressive loading in horizontal direction); (a) initial transverse cracking within the ceramic lamellae, (b) extensive transverse cracking of the ceramic lamellae and ductile cracking of the metallic alloy at a much higher total strain

As shown in Fig 9a, damage evolves by transverse cracking within the ceramic lamellae due to the elastic-plastic mismatch of the plastically flowing metallic alloy and the rigid ceramic lamellae. Although, multiple transverse cracks are observed, they do not cause the material to fail. At higher strains, extensive transverse cracking of the ceramic lamellae and ductile cracking of the metallic alloy can be seen (Fig. 9b).

According to Spowart and Dève [17] the primary factors controlling the transverse compressive strength of fiber reinforced MMCs are the ceramic volume fraction and distribution of the fibres relative to the loading direction. According to these authors, failure takes place by the matrix ductile failure, while for composites having strong interface the fibres and the interface remain unaffected. Depending upon the direction of loading and provided the fiber content is above a critical minimum value, the factor of strengthening lies between 1.15 and 3. Effective strengthening requires the ceramic fibres to strongly interrupt the matrix shear bands. On the contrary, in the interpenetrating composite studied here, there seems to be no effect of domain orientation on transverse compressive strength at domain orientations more than about 30°. This may be justified by the fact that plastic flow of the matrix causes the ceramic lamellae to crack transversely and hence their ability to interrupt the matrix shear bands and simultaneously strengthening the composite decreases.

4. CONCLUSIONS

Innovative metal matrix composites (MMC) produced by melt infiltration of ceramic preforms prepared by a freeze-casting technique were examined. These composites exhibit lamellar domains with sizes up to several millimetres, composed of alternating ceramic and metallic lamellae with thicknesses ranging from 20 to 100 µm. Elastic and in-situ elastic-plastic analysis of single domain samples were carried out. Results show that the individual domains exhibit a pronounced anisotropy in their mechanical properties. The highest stiffness is observed in the direction parallel to the preform freezing direction, the lowest in the direction perpendicular to it. Compressive mechanical tests of single domain samples showed that the domains are strongest along directions parallel to freezing direction and fails in a brittle manner by ceramic crushing. At intermediate to high domain orientations the behavior is controlled by the metallic alloy. Coating of the ceramic preform with Cu_2O before melt infiltration weakens the metal-ceramic interfaces and reduces the compressive strength along the freezing direction. The pronounced elastic and elastic-plastic anisotropy is of interest for further studies to investigate the behavior of samples having more than one domain and to investigate the internal load transfer from the compliant metallic alloy to the rigid ceramic.

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