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Strong and tough metal/ceramic micro-laminates

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ABSTRACT

There is a growing interest in the development of composites with complex structures designed to generate enhanced mechanical properties. The challenge is how to implement these structures in practical materials with the required degree of control. Here we show how freeze casting of ceramic preforms combined with metal infiltration can be used to fabricate Al_2O_3/Al-4wt% Mg micro-laminated composites. By manipulating the solid content of the suspension and the morphology of the ceramic particles (from platelets to round particles) it is possible to access a range of structures with layer thickness varying between 1 and 30 μm and metallic contents between 66 and 86 vol%. The mechanical response of the materials is characterized by combining bending tests with observation of crack propagation in two and three dimensions using different imaging techniques. These composites are able to combine high strength and toughness. They exhibit a rising R-curve behaviour although different structures generate different toughening mechanisms. Composites fabricated with Al_2O_3 particles exhibit the highest fracture resistance approaching 60 MPa $m^{1/2}$, while laminates prepared from Al_2O_3 platelets exhibit higher strengths (above 700 MPa) while retaining fracture resistance up to ~40 MPa $m^{1/2}$. The results provide new insights on the effect of structure on the mechanical properties in metal-ceramic composites as well as on the design of appropriate testing procedures.

1. Introduction

The advance of many technological fields, from healthcare to aeronautics, relies on the development of new materials with unconventional combinations of mechanical properties. Among these, ceramic-based composites are often chosen for their better corrosion and wear resistance, enhanced chemical and thermal stability, high flexural strength and hardness that make them suitable for demanding applications. However, the performance of ceramics is often limited by their brittle behaviour. The achievement of both strength and toughness is an essential requirement for many structural materials but these properties are often mutually exclusive [1]. Strong and hard materials with limited deformation such as ceramics tend to be brittle but toughness can be generated by extrinsic mechanisms that, acting behind the crack tip, shield locally the stress, generating fracture resistance. Several strategies have been engineered to maximize the toughness of monolithic ceramics such as crack deflection, grain or fibre bridging [2]. However, the values achieved so far are much lower than those of metals. Metal matrix composites (MMCs) are candidates to overcome this problem [3]. Among the different types of MMCs so far developed, laminates exhibit some of the most efficient toughening strategies [4–7]. The metallic ductile phase, after initial crack propagation in the ceramic layers, tends to remain intact and bridges the opening crack increasing the crack-propagation resistance of the material.

Metal matrix composites have been fabricated from mixtures of ceramic and metallic powders. However, this approach hampers structural control. Good powder dispersion is not always easy to achieve and aggregation is often observed [8]. Infiltration of porous ceramic preforms with a liquid metal is an attractive alternative to produce interpenetrated composites with a wide variety architectures and metal contents [9]. Infiltration usually requires the application of pressure – e.g. squeeze casting, gas pressure infiltration and high-pressure die casting infiltration – what may lead to failure of the ceramic preforms during fabrication. Therefore

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these conventional techniques are not appropriate for highly porous preforms [10]. Pressureless infiltration, such as in the Lanxide process [11], is an attractive alternative due to its cost effectiveness and near-net-shape capability [12]. However, it is usually accompanied by reactions at the metal-ceramic interface and the types of alloys that can be used are limited. One of better known systems, as reported by Aghjanian et al. [13], is the spontaneous infiltration of an alumina preforms by Al-4wt% Mg (Al-4Mg) alloys. Aluminium alloys are also attractive materials to produce MMCs due to their low density, good corrosion resistance, high thermal and electrical conductivity and high ductility.

In order to optimize the mechanical performance of a composite, it is extremely important to control accurately its structure at multiple length scales [14]. This kind of control is well exploited by Nature in the production of damage tolerant materials that are at the same time strong and tough [15]. The design of natural structures such as bone, seashells or teeth, is based on highly hierarchical architectures where an inorganic phase is cleverly interlayered with an organic component that acts as compliant phase. These composites show characteristic structural features at multiple length scales, from molecular to macroscopic dimensions. In this way, natural materials can generate both intrinsic and extrinsic toughening mechanisms within the same structure. In the last decades many researchers have attempted to replicate this type of architectural organization in artificial materials, as corroborated by the large number of publications in the field of bio-inspiration [16–19]. Some of these publications deal with the development of ceramic-based composites that use metals as the "soft" phase. However, man-made materials are still far from fully replicate the complex and intricate hierarchical structure of natural ones [15]. In general, conventional ceramic processing techniques (slip/tape-casting, hot pressing, chemical vapour deposition etc.) cannot generate the complex hierarchical organization at many different length scales necessary to simultaneously enhance strength and toughness. In response to this challenge, processing techniques such as freeze casting (ice-templating) [16,17,19,20], or more recently magnetically assisted slip casting [21] have been developed. Because these are a new class of composites, with unique combinations of mechanical properties, a deep understanding of the relationships between structure and mechanical response encompassing the influence of structural parameters acting at multiple length scales is still needed.

In this study we analyse the mechanical performance of micro-laminated alumina-aluminium composites fabricated through the pressureless infiltration of freeze casted alumina preforms. Traditionally, metal-ceramic layered composites combining different materials, e.g. Nb/Nb_3Al [7], Al/Al_2O_3, Ni/Al_2O_3 or Cu/Al_2O_3, have been produced by hot pressing [5,6,22], This approach can be used to build materials with layer thickness ranging from 50 to 500 μm. More recently layered composites have been fabricated through the infiltration of freeze-casted ceramic preforms [16,23–26]. However, the process usually requires the use of pressure. In this work we use pressureless infiltration of alumina preforms with Al-4Mg to fabricate bulk ceramic-metal layered composites. This approach allows the infiltration of highly porous preforms (>80%) with ceramic layer thickness down to 4-5 μm that cannot be easily achieved by other means. In order to manipulate the composite structure, here we use two different alumina particle morphologies to fabricate the preforms: sub-micron "round" particles or platelets [27]. By controlling the freeze casting conditions we are able to prepare layered preforms with different residual porosity and microstructural features. Mechanical testing is combined with two (optical and scanning electron microscopy) and three (X-ray tomography) dimensional characterization techniques to develop an understanding of the effect of structure in the development of strength and toughness.

2. Materials and methods

2.1. Sample preparation

Freeze casting of water-based alumina particles and platelets/particles suspensions was used to produce lamellar scaffolds as reported in other studies [16,17,27–29]. The alumina particles used were Baikalox SMA6 (Baikowski, France), with a nominal particle size of 0.3 μm, a specific surface area of 7 m^2/g and no more than 70 ppm of impurities. Suspensions with solid contents of 9 and 20 vol% (30 and 50 wt%) were prepared. The alumina platelets (Alusion™, Antaria Limited, Bentley, Western Australia) had a diameter and a thickness of 5–10 μm and 300-500 nm, respectively. To improve sintering between the platelets, water based suspensions with a total solid loading of 9 vol% (30 wt%) were produced using a bimodal distribution of platelets and particles in a ratio of 70:30 in weight. In both types of suspensions Dolapix type CA (Zschimmer and Schwarz GmbH and Co., Germany) was used as dispersant, polyvinyl alcohol (PVA) 22000 (VWR, Belgium) as binder and sucrose type Anala R Normapur (VWR, Belgium) as lamellae shaping additive. Sucrose was added to the suspension in order to induce the formation of micro-roughness on the lamellae walls and small bridges between them that play both a critical role on the shear at the ceramic-metal interface [30]. Slurries containing only alumina particles were ball milled overnight with alumina milling media. Before freezing, the slurries were de-aired at least for 1hr to remove entrapped air bubbles. To avoid any damage to the platelets only the particles used as sintering aid were ball milled overnight. After removing the milling balls, the alumina platelets were added and the suspension was sonicated for at least 30 min and mixed in a turbula mixer overnight. Afterwards, a degassing step was used in both cases.

After de-gassing, all the suspensions were directionally frozen at a controlled rate to promote ice crystal growth. The suspensions were placed in a Teflon® mould on top of a copper cold finger whose temperature was decreased at a controlled rate of 5 or 15 °C/min. When platelets were used the cold finger cooling rate was 5 °C/min to induce better platelet alignment within the lamellae [27,31]. To better align the ice crystals during their growth and therefore the ceramic lamellae in the final microstructure, a patterned cold finger was used. The patterning consisted of insulated tape covering the cold finger in which parallel scratch with a separation of ~1 mm were produced with a scalpel to ensure more controlled nucleation sites for the ice crystals [30]. The frozen samples were then freeze-dried (Freezone 4.5, Lobconco, USA) and the green bodies sintered in a conventional furnace (Ultratherm, Pyrotherm Ltd, UK) in air. The temperature was raised at 1 °C/min up to 1000 °C and held for 1 h to burn out of the binder. Afterwards the temperature was increased at 5 °C/min to 1550 °C and held for 2 h.

The porous ceramics were infiltrated with molten Al-4Mg alloy (5083 Al-alloy, Smithmetals, UK) using a pressureless process. Infiltration was carried out in a tubular furnace (Lenthon, UK). The sintered alumina scaffolds were placed on top of aluminium alloy pieces inside an alumina crucible. The tube was flown with nitrogen for 1 h to reduce the oxygen in the system. Several aluminium alloy pieces were usually placed right before the sample to be infiltrated, as external getter to reduce the concentration of oxygen inside the tube [13]. The furnace was heated at 35 °C/min to 400 °C where the temperature was held for 30 min. The temperature was then increased up to 950 °C for 180 min to complete the infiltration.

2.2. Microstructural characterization

The microstructures of the materials were analysed via