



Developing strength and toughness in bio-inspired silicon carbide hybrid materials containing a compliant phase



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ABSTRACT

Freeze casting has proven to be a versatile processing route to fabricate bio-inspired (“nacre-like”) hybrid composites that exhibit unique combinations of strength and toughness (damage-tolerance). To date, however, the effects of small changes in the architecture of such composites on their mechanical properties have been poorly investigated. Here we examine the influence of microstructural features such as ceramic/polymer ratio, layer thickness, and presence of bridges between ceramic lamellae, on the mechanical performance of the resulting composites. To this end, we compare the flexural strength and resistance to failure of a suite of silicon carbide/polymethyl methacrylate (SiC/PMMA) layered composites made by polymer infiltration of freeze-cast SiC scaffolds with various architectures. Our composite structures all show an increasing fracture resistance with crack extension (rising R-curve behavior) due to extrinsic toughening mechanisms such as uncracked-ligament bridging, inelastic deformation of ductile layers, lamellae pull out and ceramic bridge fracture. We show that a fine tuning of the composite architecture can lead to SiC/PMMA samples with a dendritic morphology, which exhibit the best strength and toughness. Specifically, the presence of ceramic bridges connecting the lamellae is seen to provide a strengthening effect similar to the mineral bridges between aragonite platelets in nacre, where they prevent debonding and limit platelet sliding; additionally, the fracture of these bridges between lamellae during crack extension is a potent toughening mechanism, thereby conferring optimal damage tolerance to the material.

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1. Introduction

The mechanical properties of structural materials, particularly in the quest for tougher and stronger (damage-tolerant) materials, have been an important topic of research for many decades [1]. Although ceramics are often ideal candidate materials when high specific strength, stiffness and creep resistance are required, their inherent brittleness and flaw sensitivity severely limit their potential use for many structural applications.

Natural materials such as nacre, bone or dentin, are biological composites in which highly brittle minerals (hydroxyapatite, aragonite, etc.) and soft biopolymers (collagen, chitin, cellulose, lignin, etc.), are combined in complex hierarchical structures. Despite containing significant amounts of the brittle component, the mechanical properties of these materials, in particular their fracture toughness, are quite exceptional and invariably exceed those of the individual constituents [2]. The excellent fracture resistance

of biological composites, however, does not necessarily result from conventional measures of the toughness, such as the *crack-initiation toughness* K_{Ic} , but rather from the fact that these materials can sustain subcritical cracking without catastrophic failure, the latter being a function of their *crack-growth toughness* which can be assessed in terms of so-called rising crack-resistance curve (R-curve) behavior. Such fracture resistance results from a combination of toughening mechanisms that occur both ahead of the crack tip, i.e., intrinsic mechanisms largely associated with plasticity, and those occurring primarily in the wake of a propagating crack, i.e., extrinsic mechanisms associated with crack-tip shielding¹ [3].

¹ Intrinsic toughening mechanisms operate ahead of the crack tip to generate resistance to microstructural damage, primarily through plastic deformation which provides a means of blunting the crack tip through the formation of “plastic” zones. Extrinsic toughening mechanisms, conversely, operate primarily in the wake of the crack tip to inhibit cracking by “shielding” the crack from the applied driving force. Whereas intrinsic toughening is effective in inhibiting both the initiation and growth of cracks, extrinsic mechanisms, e.g., crack bridging and crack deflection, are only effective in inhibiting crack growth [1,3].

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Nacre, the ‘gold standard’ for biomimicry with both high strength and toughness, has been the source of inspiration for many synthetic hybrid materials that aspire to combine strength with deformability [4–9]. Comprising ~95 vol.% of aragonite (CaCO_3) platelets surrounded by thin layer of proteins, nacre displays a fracture toughness that is at least one order of magnitude higher than that of either the mineral or the bio-polymers.² This result is achieved through a precise architecture that resembles that of a brick wall, and the clever design of the interface. Specifically, hard aragonite platelets as the “bricks” provide for strength and the organic layer as the “mortar” enables *limited* inelastic deformation and thus energy dissipation, yielding a composite, when hydrated, that is remarkably strong (~130 MPa) and tough (work of fracture ~1 kJ m⁻²) [10]. Nacre’s characteristic brick-and-mortar architecture and the associated toughening mechanisms have been widely studied [10–13] to the extent that the design of nacre-inspired composites is now arguably guided by theoretical models predicting the mechanical response of ideal structures [14–17].

However, enticing as these natural composites are, the development of a new class of damage-tolerant, nature-inspired materials is still surprisingly limited to but a few examples [5–9]; despite some notable results, this field is far from maturity. Although the concepts underlying the design of nacre-inspired materials are quite straightforward [15], the development of such composites is constrained by our ability to make them. Conventional processing techniques, e.g., tape casting and hot pressing [18,19], are unable to replicate the hierarchy observed in natural composites such that toughening is generated mainly from crack deflection rather than concomitant mechanisms acting at multiple length-scales. “Bottom-up” approaches, such as layer-by-layer deposition [20,21], self-assembly [22,23], etc., are often not suitable to fabricate bulk materials and are generally limited to thin films of moderate practical interest. Alternatively, freeze casting (ice-templating) [24] offers the unique opportunity to employ a “top-down” approach to mimic the hierarchical structure of nacre; indeed, to date this processing technique has yielded several bulk hybrid materials with very unique properties [6,8,9,25].

This technique has been exploited to produce scaffolds with complex porosity [24,26–30], and has proven to be effective for the fabrication of nacre-like composites based on alumina [3,6,8,24,25]. By unidirectional freezing of a ceramic suspension, a solid body made of alternate layers of ice and ceramic particles is produced. Upon sublimation of the ice and sintering of the green ceramic body, a scaffold with unidirectional pores is obtained that can be infiltrated with a polymer or a metal. Control of the freezing conditions, suspension characteristics and additives is paramount to tailor the features of the scaffolds, such as size and morphology of pores, thickness and roughness of ceramic walls, ceramic fraction, etc. [24,29,31,32]. By careful control of the processing parameters, lamellar composites containing up to 45 vol.% of ceramic were first fabricated [24]. These composites showed similarities to nacre but with a lamellar structure with the characteristic surface roughness of the ceramic layers [24]. Shortly afterwards, the same procedure was modified to increase the ceramic fraction up to 80 vol.% and to produce a composite that exhibited a “nacre-like” brick-and-mortar structure resembling many features of the inorganic platelets in the bio-composite (including the presence of ceramic bridges) [6]. Starting from ice-templated structures, both lamellar and brick-and-mortar composites consisting of alumina and polymethyl methacrylate (PMMA) were produced that displayed exceptional toughness and strength. Combining a

ceramic, which provides for strength, with a relatively soft polymer acting as a “compliant” or “lubricant” phase led to final materials that exhibited a rising R-curve behavior with fracture toughness up to 300 times higher, in energy terms, than that of the main constituents [6,25]. However, systematic investigations of the effects of structural parameters, such as lamellae thickness, presence of ceramic bridges, ceramic fraction, etc., on the mechanical response of these hybrid composites has to date been largely overlooked. Launey et al. reported that varying the thickness of the ceramic layer in lamellar Al_2O_3 -PMMA composites had an almost negligible effect on the flexural strength [25], but their study was focused on a comparison between lamellar and brick-and-mortar composites, rather than the investigation of the relationship between structural features of the composites obtained from freeze-cast structures and their mechanical performance.

Based on observations that the structures obtained by freeze casting can change considerably by varying the processing parameters [29], the objective of the current study was specifically to investigate the influence that these changes can have on the mechanical response of ceramic/polymer lamellar composites. As silicon carbide (SiC) had never been examined in this context, we used freeze casting to fabricate SiC scaffolds with characteristic pores morphologies, either lamellar or dendritic. These scaffolds were infiltrated with PMMA generating composites that vary in ceramic content (40–60 vol.%), lamellae thickness (5–35 μm) and the number of ceramic bridges between the individual lamellae. The mechanical behavior of these composites is described in terms of flexural strength and resistance to failure using micro-notched samples loaded in three-point bending. The results obtained show a clear dependence of the damage tolerance of these materials on the ceramic content of the individual composites and a strong correlation with the structure achieved by varying processing routines.

2. Experimental procedures

2.1. Processing

Porous silicon carbide scaffolds were prepared by freeze casting, following the procedure described in detail in our previous study [29]. We prepared suspensions containing 17–30 vol.% of SiC powders with sintering aids (Al_2O_3 and Y_2O_3) dispersed in deionized water, and added 5 wt.% polyethylene glycol (PEG) as a binder. The suspensions were electrostatically stabilized by adding ammonium hydroxide (NH_4OH) to change the pH to 10 and by ball milling for 40 h. Ice-templated scaffolds were produced by freezing the SiC suspensions at cooling rates between -1 and -15 °C min⁻¹. After water sublimation, the scaffolds were first treated in air at 450 °C for 4 h and then sintered in inert atmosphere at 1900 °C for 30 min. Details on the effects of the processing parameters on the ceramic fraction and the architecture of the scaffolds are reported elsewhere [29]. Based on that study, we selected a combination of parameters to produce scaffolds with three different ceramic contents: 40, 50 and 60 vol.%. For samples containing 50 vol.% ceramic, we produced scaffolds with either lamellar or dendritic morphology.

The SiC scaffolds were infiltrated with PMMA in order to fabricate composites and assess the influence of the different architectures on the mechanical properties. The polymer infiltration was carried out in two steps, preceded by grafting of the ceramic surface. For this purpose, the ceramic scaffolds were immersed in 50 vol.% solution of 3-(trimethoxysilyl) propyl methacrylate (γ -MPS) in acetone for 12 h. After drying, the grafted scaffolds were immersed in a solution of methyl methacrylate (MMA) in toluene (toluene:MMA 2:1 by weight) for 2 h at 70 °C to perform free

² Expressing the toughness in energy terms, i.e., in terms of the critical strain-energy release rate, G_{IC} , the toughness of nacre is some three orders of magnitude higher than the toughness of its constituent phases.

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