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Processing of Al$_2$O$_3$/Y-TZP laminates from water-based cast tapes

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Abstract

Laminated structures have been investigated due to their capability for the reinforcement of ceramics. Crack deflexion and bifurcation, surface strengthening and threshold strength are the mechanisms associated to the fracture of laminated ceramic. In all the cases, a precise control of the thickness and composition of the layers is necessary. In this sense, colloidal processing techniques have proved their adequacy for the fabrication of designed layered structures. This paper deals with the fabrication of layered ceramics by stacking water-based cast tapes at room temperature and using low pressures. In order to control the pressing procedure, the engineering strain–stress curves recorded during the stacking of the tapes were analyzed. Afterwards, the sintering conditions have been optimized by adjusting the green density of the tapes to avoid differential sintering and the associated cracks. Monolithic and layered materials free of cracks have been fabricated using optimized processing conditions.
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1. Introduction

Severe environments imposed by new technologies demand the fabrication of materials with better properties and more tolerance restrictions, in which reliability must be ensured. Consequently, ceramics have been proposed as either substitutes for currently used materials (e.g. metals and plastics) or as complements to existing materials (i.e. in the form of composites), due to their favorable properties such as high temperature hardness and strength, and good thermal and chemical stability. However, the intrinsic brittleness of ceramic materials has forced to look for new designs and processing routes to improve their mechanical behavior while maintaining the low cost and low environmental impact.

One such method for improving the mechanical behavior of ceramics has been the reduction of the defects in the ceramic body through colloidal filtration and processing techniques [1]. Another method has been the creation of multi-phase composite architectures composed of ceramic matrices reinforced by the addition of particulate, fiber, and/or laminar secondary phases. The use of laminar reinforcement has been identified as a simple and inexpensive method of achieving much of the mechanical property benefit of fiber reinforcement without the associated processing complexity.

The use of laminar reinforcements is not, however, a completely original idea. For million of years, living creatures have designed protective laminar structures with optimized mechanical properties utilizing the rather limited resources provided by their surroundings [2]. Such is the case of the mollusk shell, in which a laminar structure composed of alternating brittle but strong aragonite layers and ductile but weak organic polymer layers provides a protective shell with toughness and strength roughly 10 times that of a corresponding aragonite single-crystal [3]. Mimicking this concept, Pizyk and Aksay fabricated several laminar metal–ceramic and ceramic–polymer composites that showed improved mechanical properties compared to their corresponding monolithic materials [4].

Layered ceramics were initially developed in the 1960s as a result of the necessity of these structures for the packaging of microelectronics. As a consequence of this demand, different methods to obtain ceramic multilayers with controlled thicknesses, as well as handling and lamination possibilities, were studied [5,6]. These methods were based on the tape casting technique, in which multilayer structures were obtained by the stacking of green tapes followed by subsequent consolidation by either the application of pressure at a suitable temperature, or by roll-to-roll compaction.

The preparation of tape-cast layered ceramics for structural applications was first reported by Mistler [7]. He described an

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experimental device designed for the fabrication of green tapes, which cast multiple layers simultaneously, thus eliminating the need for subsequent consolidation operations. Using this device he prepared a trilayered alumina/alumina ceramic in which only the two outer layers were doped with talc and the inner layer with MgO. This distribution of dopant resulted in a stress free laminate with a coarse-grained, high toughness core layer surrounded by fine-grained, high strength outer layer. Mechanical testing revealed that the fracture strength of this laminar material was increased relatively to that of the monoliths with the same composition as that of the constituent layers. Russo et al. [8] used a similar microstructural engineering concept to tape cast a trilayered material where the outer, thick layers were made up of a homogeneous alumina/aluminum titanate particulate composite, while the inner layer was made of a heterogeneous composite of the same composition. It was observed that, at an optimum surface layer thickness (around 100 μm), the outer layer strength dominated fracture at small flaw sizes, while the strength of the inner layer dominated at large flaw sizes.

In pioneering work performed by Clegg et al. [9], it was shown that a layered structure composed of strong layers separated by weak interlayers, formed by a dough rolling technique, could improve mechanical behavior significantly by causing the material to fail in a graceful, step-wise manner. The authors showed that by intercalating thin layers of graphite between thicker layers of SiC, deflection of the crack by the graphite interlayers caused the load–displacement curve to display such behavior. The work of fracture of these laminates was two orders of magnitude higher than that of monolithic SiC samples and the apparent toughness was increased from 3.6 MPa m$^{1/2}$ in the monolithic to 17.7 MPa m$^{1/2}$ in the laminate. It was also reported that the toughening effect of the graphite layers reached a maximum at layer thicknesses of 3 μm. Step-wise, graceful failure has also been produced in alumina/mullite laminates made by co-sintering of extruded alumina sheets of 1 mm thickness joined with kaolin and resin tapes [10]. The fracture behavior showed once again the importance of the relative thickness of the layers and the microstructure of the thin internal layer. Step-wise load/displacement behavior resulting from crack deflection by the thin layers was observed for mullite layers with thicknesses in excess of 100 μm, while the crack passed undeflected through mullite layers with thicknesses less than 100 μm, resulting in non-graceful, catastrophic failure. This series of results shows that a combination of ceramic layers in a structure is capable to result in crack deflection and step-wise fracture, even when the combined layers have the same composition but different microstructure [11–13]. He and Hutchinson [14] settled the energy criteria for the possible propagating path of a crack in a biphasic material where it could either deflect or penetrate through the interface. The conditions for each possible case were studied in terms of elastic modulus and fracture energy. Their analysis concluded that the impinging crack is likely to be deflected at the interface if

$$\frac{G_{ic}}{G_c} < \frac{G_d}{G_p} \qquad (1)$$

being G_{ic} the toughness of the interface, G_c the toughness of material were the crack penetrate, while G_d and G_p are the energy release rate of the deflecting and penetrating crack, respectively.

Most of the laminates reported in the above paragraphs base the reinforcement in the deflection criteria of He and Hutchinson. However, the residual stresses constitute a particularly significant feature associated with laminated ceramics and play a very important role during fracture of ceramics. These stresses are developed as a consequence of the thermal strain mismatch between layers of different composition that occurs during cooling from sintering. This strain mismatch can be intentionally designed through the use of constituent materials with differing thermal expansion coefficients (CTE) [15–18], or through the incorporation of materials that undergo volume-displacing phase transformations [19–24]. Despite the differing mechanisms associated with these two processes, both generate a differential strain between the layers, $\Delta\varepsilon$, that results in the development of residual stresses.

The level and sign of the expected residual stresses can be evaluated using the simplified model of a symmetric plate constituted by alternated layers with a biaxial distribution of stresses constant across the layers [25]. The level of the stresses in the center of the layers that constitute the layered structure are given by

$$\sigma_a = -\frac{\Delta\varepsilon E_a'}{1 + (E_a' n_a t_a / E_a' n_b t_b)} \qquad (2)$$

$$\sigma_b = -\sigma_a \frac{n_a t_a}{n_b t_b} \qquad (3)$$

where the subscript 'a' and 'b' refers to the alternating layers, n_i and t_i are the number and thickness of the layers of each composition, respectively, and E_i' is given by

$$E_i' = \frac{E_i}{1 - \nu_i} \qquad (4)$$

where E_i and ν_i are the Young's modulus and the Poisson ratio of the layers. The strain mismatch, $\Delta\varepsilon$, is calculated as follows

$$\Delta\varepsilon = \int_{T_r}^{T_s} (\alpha_b - \alpha_a)dT + \Delta\varepsilon_t \qquad (5)$$

where, α_i are the coefficients of thermal expansion of the layers, T_s is the temperature above which mass transport occurs in the material, and T_r the room temperature, and $\Delta\varepsilon_t$ is the strain mismatch between the layers due to processes different from thermal expansion, such as phase transformation.

The fracture behavior phenomena associated to the residual stresses of laminates are bifurcation [15,19,20] surface strengthening [16,21] and threshold strength [17,22]. All these mechanisms are associated to the development of residual compressive/tensile stresses in alternated layers. Usually, it is desired the relative thickness of the compressed