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Ceramic laminates with improved mechanical reliability by tailoring the porosity of the constituting layers

Vincenzo M. Sglavo[a,b,*], Niccolò Bellettati[a,*]

[a] Department of Industrial Engineering, University of Trento, Via Sommarive 9, Trento, Italy
[b] INSTM Research Unit, Via G. Giusti 9, 50123 Firenze, Italy

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ABSTRACT

Two different ceramic laminates composed of porous alumina and alumina/zirconia layers were designed and produced in the present work. The two symmetrical architectures were selected whose fundamental difference is the presence on the surface of a porous layer in the first and a compact alumina/zirconia composite layer in the second. The residual stress profile and corresponding fracture toughness were tailored to promote the stable growth of surface defects prior to final failure to increase the mechanical reliability of the material. The laminates were realized by stacking together different green laminae (containing specific pore former content) in a specific order, thermo-compression and co-sintering. The results point out an important reduction of the strength scatter and a clear insensitivity to surface damage. It is also shown that the mechanical performances are strictly related to the specific architecture of the laminate, this allowing to tailor a priori the mechanical performances of the composite.

1. Introduction

Several strategies have been advanced in the last decades to increase the fracture toughness and overcome the problems associated to the limited mechanical reliability of ceramics. Important advancements have been obtained through the discovery of some fundamental toughening mechanisms associated to microcracking, martensitic phase transformations, grain bridging effects or reinforcements due to the presence of whiskers and fibres [1,2]. Flaw tolerant materials have been also produced in laminated structures where porous/weak interfaces or layers under strong compression promote delamination, crack deflection or even fracture arrest [3–23].

More recently, Sglavo et al. have shown that ceramic laminates [24–28], as well as specifically tempered glasses [29–33], with a residual stress profile with a maximum compression at a certain depth from the surface posed under tension can arrest crack propagation or force it to occur in a stable way before final catastrophic failure. Using this approach it is possible to design and produce a ceramic material characterized by higher fracture toughness and limited strength scatter, thus improving enormously the flaw tolerance and mechanical reliability.

The design procedure is based on the fundamental idea that residual stresses can be developed in a layered ceramic after sintering due to differences in thermal expansion coefficient, stiffness and possible phase transformations. If these latter are absent, under the hypothesis of perfect adhesion among the different layers, in a planar symmetrical laminate each layer (i) is subjected to a biaxial residual stress equal to [24,26]:

$$\sigma_{res,i} = E_i^* \left(\bar{\alpha} - \alpha_i \right) \Delta T \tag{1}$$

where:

$$E_i^* = E_i / \left(1 - \nu_i \right) \tag{2a}$$

$$\bar{\alpha} = \frac{\sum_{i=1}^{n} E_i^* t_i \alpha_i}{\sum_{i=1}^{n} E_i^* t_i} \tag{2b}$$

E_i, ν_i, α_i and t_i being the elastic modulus, Poisson's ratio, thermal expansion coefficient and thickness of the specific ith ($1 < i < n$) lamina, respectively, and ΔT the difference between the "stress free temperature" (always lower than sintering temperature) [8] and room temperature.

In this way, a residual stress with step profile along the laminate thickness is obtained.

At this point the concept of "apparent fracture toughness" can be introduced [26,30] which results from the combination of the residual stress effect with the fracture toughness of the material constituting each layer; therefore, the possibility for a crack

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* Corresponding author.
E-mail addresses: vincenzo.sglavo@unitn.it, vincenzo@sglavo.it (V.M. Sglavo).

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to propagate can be identified by comparing the external applied stress intensity factor (K) with such apparent fracture toughness. A general equation, which defines the apparent fracture toughness in the i^{th} layer in the depth interval $[x_{i-1}, x_i]$ (x_i being the starting depth of i_{th} layer) has been proposed [24–26,30]:

$$T = K_{C,i} - \sum_{j=1}^{i} \left\{ 2Y \left(\frac{c}{\pi} \right)^{0.5} \Delta\sigma_{res,i} \left[\frac{\pi}{2} - arcsin \left(\frac{x_{j-1}}{c} \right) \right] \right\} \qquad (3)$$

where $K_{C,i}$ is the fracture toughness of the specific i^{th} layer, c the crack length, $\Delta\sigma_{res,i}$ the stress difference between layer i and layer (i-1); $Y \approx 1.12$ for an edge crack in a semi-infinite body [2]. One can easily observe that T increases with depth as long as $\Delta\sigma_{res,i}$ is negative, i.e. when compressive stresses become more intense entering within the material. Therefore, it is possible to tailor the apparent fracture toughness by changing the residual stress profile, i.e. the composition of the layers, their thickness and stacking order; the possibility occurs to design an apparent fracture toughness, which can promote the stable growth of surface defects (under the condition $dT/dc < dK/dc$) under external loading before final catastrophic failure, thus increasing the mechanical reliability of the material [30].

The reported theory has been reduced to practice on several dense ceramic laminates composed by alumina/mullite, alumina/zirconia, alumina/mullite/zirconia and alumina/silicon carbide [24–28]; all materials have shown peculiar crack propagation, high strength and insensitivity to surface defects.

The inspiration of the present work aroused from the observation that, according to Eq. (1), the residual stress can be varied by changing not only the thermal expansion coefficient but also the elastic modulus, which, if the composition is constant, depends on the material porosity. The present experimental work can be considered within the more general research activity aiming to realize ceramic laminates where the entire surface architecture is tailored to achieve specific mechanical performances like threshold stress or insensitivity to macroscopic surface flaws; it must be pointed out that this is rather different than the method typically used in ceramic laminates where two different layers are alternated in the entire thickness. In the current approach, the surface layers' composition is changed uniformly for reducing possible processing defects (like delaminations, tunnelling or edge cracks) arising when very high residual stresses are generated [34–36], *i.e.* when very different materials are combined. By using such methodology, layers of different porosity are not used to promote cracks deflections [6,7]; instead, the attempt has been made to design and produce ceramic laminates whose mechanical performance depends on two different porous architectures and surface structure.

2. Experimental procedure

Sub-micron α-alumina (CT3000SG, Almatis – $D_{50} = 0.5$ μm) powder was used as primary starting material. Rice starch (Sigma-Aldrich, 85654 Fluka Biochemika) was chosen as pore former. Yttria (8 mol%) stabilized zirconia powder (TZ-8YS, Tosoh – $D_{50} = 0.6$ μm) was used to produce, together with alumina, dense composite layers with variable thermal expansion coefficient. Fully stabilized cubic zirconia powder was specifically chosen to avoid any phase transformation upon sintering and/or cooling or successive mechanical testing.

The green alumina layers were obtained by tape casting water-based slurries, prepared by a two-stage milling/mixing procedure. The suspensions were produced by using NH_4-PMA (Darvan C-N, R.T. Vanderbilt) as dispersant and acrylic emulsions (B1014, Duramax) as binder. Lower-T_g acrylic emulsion (B-1000, Duramax) was also added to the suspensions as plasticizer. Finally, 10 min before casting, three drops of 10 wt% wetting agent water solution (NH_4-lauryl sulphate, 09887, Fluka Chemie) were added to improve the tape spread onto the casting substrate. The alumina slurry was prepared by mixing the powder with deionized water and the dispersant (0.4 mg/m^2 of active matter per unit area of alumina powder) with alumina balls in a plastic jar for 24 h. Then, rice starch was added to the suspension [37] for obtaining the required porosity after sintering and the produced slurry was mixed for additional 2 h. The suspension was filtered and de-aired for 30 min using a low-vacuum Venturi pump to remove air bubbles entrapped during the mixing stage. Finally, binder and plasticizer were added and the suspension was slowly mixed for 30 min to reach good homogeneity, taking care to avoid the formation of new bubbles. Tape casting was carried out using a double doctor-blade assembly (DDB-1-6, 6 in wide, Richard E. Mistler, USA) with speed of 0.75 m/min and blade height ranging between 100 μm and 250 μm. The used substrate was a composite film (PET12/Al7/LDPE60, BP Europack, Vicenza, Italy) fixed on a rigid float glass plate, with the tape casted onto the polyethylene hydrophobic surface side. The relative humidity of the over-standing environment was set to 80% in order to control the drying speed of the tape for at least 24 h. Green composite layers were produced in the same way by mixing alumina and zirconia powders in a weight ratio of 1/1.036; this allowed to realize a composite containing 40 vol% zirconia after sintering. Disks of 30 mm nominal diameter were the cut from the tapes by a hollow punch; then, they were opportunely stacked to produce the designed laminate architecture.

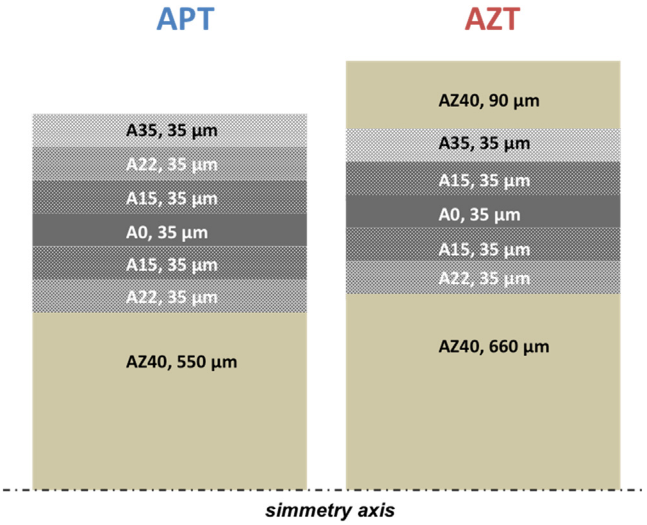

Fig. 1. Nominal architecture of the two considered engineered laminates: the nominal thickness (dimensions are not in scale) of each layer is indicated.

Two different laminates were considered in the present work, whose architecture is schematically shown in Fig. 1. In the current approach [26,30] the surface layers' architecture is tailored for obtaining specific mechanical performances. The two architectures considered here were selected among others with two different strategies. In the first case (laminate "APT"), the surface layer is the most porous and consequently the most compliant; then, the porosity of the layers decreases in the successive internal layers down to the fourth one, which corresponds to dense alumina. This sequence makes the material "resistance and stiffness" increasing with depth, down to about 150 μm. In the second case (laminate "AZT"), a compact alumina/zirconia composite surface layer is used, maintaining substantially the same sequence of porous layers below. In both cases, the residual biaxial stress in each layer arises fundamentally from the different elastic modulus determined by the variable porosity and the presence of dense alumina–zirconia

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