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.

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
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 tri layered alumina/alumina ceramic in which only the two outer layers were doped with tale 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 monolithics with the same composition as that of the constituent layers. Russo et al. [8] used a similar microstructural engineering concept to tape cast a tri layered material where the outer, thick layers were made up of a homogeneous alumina/alumina 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 behavior to display such behavior. The work of fracture 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 of these laminates was two orders of magnitude higher than that of monolithic 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, Δε, 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

\[ \sigma_a = \frac{\Delta \epsilon E_a^i}{1 + (E_a^i n_a t_a / E_b n_b t_b)} \]

\[ \sigma_b = -\frac{\sigma_a n_a t_a}{n_b t_b} \]

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 \( E_i^i \) is given by

\[ E_i^i = \frac{E_i}{1 - \nu_i} \]

where \( E_i \) and \( \nu_i \) are the Young’s modulus and the Poisson ratio of the layers. The strain mismatch, Δε, is calculated as follows

\[ \Delta \epsilon = \int_{T_c}^{T} (\alpha_b - \alpha_a) dT + \Delta \epsilon_t \]

where, \( \alpha_i \) are the coefficients of thermal expansion of the layers, \( T_c \) is the temperature above which mass transport occurs in the material, and \( T_r \) the room temperature, and \( \Delta \epsilon_t \) 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...
layer to be as thin as possible in order to minimize the residual tensile stresses developed in the adjacent layer.

The control of the residual stresses and, consequently, the reinforcing mechanism, is achieved by a strict control of the composition and thickness of the layers that have to be designed for an optimum behavior. In this sense, the fabrication of multilayer ceramics by colloidal processing techniques has been widely used due its versatility and reliability. These methods have the advantages that allow to strictly control the composition and, depending on the technique, the thickness of the layers. The colloidal processing techniques described for the fabrication of laminated ceramic include tape-casting [6,11,18,24], centrifugal casting [26,27], sequential slip-casting [17,28–30], electrophoretic deposition (EPD) [31,32], and others (e.g. [33–35]). All of them are based on the preparation of stable slurries with specific compositions that are piled up by adding a layer to a previously formed one. Stable slurries that ensure a homogenous and well-dispersed composition are obtained by controlling the interparticle potentials developed within the liquid media [36–38]. The thickness is controlled by controlling the processing parameter associated to the technique (casting time [30,31], blades gap [6], amount of slurry [26], etc.). Finally, the green layers that form the laminate are co-sintered.

Table 1 illustrates the versatility of three of the above-mentioned colloidal processing methods for the fabrication of structural laminated ceramics. Studies dealing with the processing and behavior of laminar ceramics for non-structural applications (e.g. microelectronics packaging) are beyond the scope of this work.

Tape casting is one of the methods that more extensively has been used for producing multilayer ceramics. It is very attractive due to its suitability for mass production and its design ability for different layered structures by varying the individual layer composition and thickness as well as the stacking sequence [5,24,47]. Usually, the tapes are fabricated with thermoplastic binders and plasticizers in an organic media and pressed at a temperature close to the melting temperature (20–120 °C) of the tape additives. Nowadays, the tendency in production methods is to use water-based formulations, for economic reasons as well as in order to avoid environmental and safety problems derived from the use of organics [48–50]. Unfortunately, the use of water-based systems for tape casting makes tapes to be more prone to cracking during drying, because of the evaporation of water is slower than that of organics. In order to overcome this problem, the optimization of the slurry in terms of high solid content is required. High solids content reduces the amount of water to be evaporated and, consequently the tendency of the tape to cracking [48]. With water-based tapes, the studies related to the lamination process are not as extensive as in the case of organic-based tapes. Some works describing the fabrication of compacts from lamination of water-based tapes can be found in the literature [51,52]. In these cases, the interfaces should be completely removed by the applied pressure [53].

In this paper, a method to obtain multilayer ceramics from water-based green tapes using a gluing agent for stacking and pressing at room temperature is described. The criteria for selecting the mechanical conditions to ensure the joining between tapes are presented. After this the green density of the layers is adjusted in order to avoid cracks associated with the differential sintering between the layers. This method of adjusting the green density of the layers has been reported to ensure the fabrication of metal–ceramic graded materials with very different sintering behavior [54,55]. Following an optimized procedure, an alumina–alumina/zirconia crack free multilayer ceramic has been obtained.

2. Experimental

2.1. Preparation of tapes

Tapes were cast from stable slurries of high purity α-Al2O3 and Y-TZP powders in deionized water as dispersing media. Table 2 shows the main characteristics (particle size, specific surface area and density) of the starting powders used.

Slurries were prepared by mixing the powders with the water containing a 0.8 wt%, referred to solids, of a polyelectrolyte (Dolapix CE 64, Zschimmer and Schwarz, Germany) used as dispersant. After mixing, the slurries were ball milled during 4 h in alumina jars using alumina balls.

Table 2

<table>
<thead>
<tr>
<th>Powder (manufacturer)</th>
<th>dₐ₀ (µm)</th>
<th>SSA (g m⁻²)</th>
<th>ρ (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>α-Al₂O₃ (Condea HPA 0.5, USA)</td>
<td>0.3</td>
<td>9.5</td>
<td>3.88</td>
</tr>
<tr>
<td>ZrO₂ (3 mol% Y₂O₃) (TZYS, TOSOH, Japan)</td>
<td>0.45</td>
<td>6.7</td>
<td>6.04</td>
</tr>
</tbody>
</table>
Two compositions were formulated containing 95 vol% of \(\alpha\)-Al\(_2\)O\(_3\) and 5 vol% of Y-TZP (named A-5YTZP) and 60 vol% of \(\alpha\)-Al\(_2\)O\(_3\) and 40 vol% of Y-TZP (named A-40YTZP). For each composition, two slurries with different solid content loading (solid content of 47 and 50 vol%) were prepared.

As tape casting additive a water-based polymeric emulsion (Mowilith DM 765 E Celanese, Spain), with a \(T_g\) of \(-6^\circ\)C and solid content 50 vol\%, particle size 0.05–0.15 \(\mu\)m was added in a concentration of 5 wt% referred to solids. Tapes were cast using a moving carrier with a blades gap of 500 \(\mu\)m. Full details of slurry preparation and tape casting procedure are given elsewhere\[53,56\].

After casting the green ceramic tapes were dried in air for 24 h, to further drying at 60 \(^\circ\)C for 48 h. The final thickness of the green tapes obtained varied between 480 and 520 \(\mu\)m.

### 2.2. Lamination of tapes

Round shaped tapes (diameters \(\varnothing\) 26 mm and \(\varnothing\) 60 mm) were used to avoid heterogeneous stress distribution within the specimens during pressing \[56\]. Green densities of the single tapes were determined using the geometrical method. Reported values are the average of those obtained for four discs of 26 mm diameter processed under nominally identical conditions.

Laminated and monolithic compacts with seven layers were obtained by piling up tapes with a gluing agent between them and further pressing. Before lamination some of the tapes were subjected to ‘wet’ pretreatment that consists of dipping the ceramic tape in distilled water during 1 min. The gluing agent consisted in a 5 wt% dilution of the binder used in the formulation of the starting slurries (Mowilith DM 765 E). Full details of the gluing agent selection can be found elsewhere \[56\].

Symmetrical laminated samples were stacked in a way to keep the A-5YTZP composition tapes as outer layers. Obtained green laminates were cold uniaxially pressed using a universal testing machine (Microtest SA, Spain) with steel compression plates. In order to avoid friction with the plates, the stacked pieces were placed between two sheets of polypropylene film. The pressure was applied using a load frame displacement rate of 0.05 mm/min. The load and the displacement of the load frame were recorded during the pressing process and engineering stress–apparent strain curves were calculated assuming uniaxial compression using the dimensions of the pieces. The safe pressure interval was determined by analyzing the curves corresponding to nominally identical samples, adjusted to third degree polynomials, and the average of the curves was used to represent the behavior of the samples under the pressure. The curves corresponding to two individual (\(\varnothing\) 26 mm) samples of each composition and pre-treatment were recorded for samples pressed up to the maximum stress values (\(\approx\)90 MPa). The corresponding stress–strain behavior was represented by the average of the corresponding polynomial fits and error bars indicated the maximum and minimum values. For lower pressures (up to 20 MPa) five samples were analyzed, in this cases error bars corresponded to the standard deviations. Green densities of obtained specimens were determined by the Archimedean method in mercury, using five dry pieces fabricated under nominally identical conditions, and compared to those of the dry green tapes. Reported values are the average of the five values and errors are the standard deviations. Relative green densities were calculated as percent of the calculated theoretical density for each composition, using 3.99 g/cm\(^3\) for \(\alpha\)-Al\(_2\)O\(_3\) (ASTM 42-1468) and 6.10 g/cm\(^3\) for Y-TZP (ASTM 83-113).

<table>
<thead>
<tr>
<th>Sample</th>
<th>Starting solid loading (vol%)</th>
<th>Green density (th.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A-5YTZP(1)</td>
<td>50</td>
<td>59.1 ± 0.1</td>
</tr>
<tr>
<td>A-5YTZP(2)</td>
<td>47</td>
<td>56.2 ± 0.1</td>
</tr>
<tr>
<td>A-40YTZP(1)</td>
<td>50</td>
<td>55.1 ± 0.1</td>
</tr>
<tr>
<td>A-40YTZP(2)</td>
<td>47</td>
<td>53.5 ± 0.1</td>
</tr>
</tbody>
</table>

Fig. 1. Characteristic behavior of dry and wet samples made of stacked A-5YTZP(1) tapes during pressing. (a) Average polynomial curves for each treatment (error bars represent the upper and lower curves). (b) Derivative of the average polynomial curves at low pressures.
2.3. Sintering and characterization

Large green samples (Ø 60 mm) were machined into bars (approximately 50 mm × 7 mm × 4.1 mm) and the surfaces were smoothed with sandpaper. Binder burn out and sintering were performed in single thermal treatment cycle. The binder burn out was carried out with heating at the rate of 1 °C/min up to 600 °C, with a dwell time of 30 min. Right after this the sintering was carried out by increasing the temperature with heating rate of 5 °C/min up to 1550 °C with a dwell time of 2 h. The dilatometer curves were recorded using green monolithic samples (5 mm × 5 mm × 4 mm) in a dilatometer with alumina support (Setaram, Setsys-16/17, France) and corrected for alumina expansion. Density values of sintered pieces were determined by the Archimedes method in distilled water and relative densities were calculated as described for green densities.

Cross-sections of the sintered samples were polished using diamond paste down to 1 μm. Microstructural observations were carried out using a reflected light optical microscopy (Carl-Zeiss H-P1, Germany) and scanning electron microscopy (Zeiss DSM-950, Germany). Samples for tunnel type crack observation were chemically etched with HF (40 vol%) for 15 min (20 °C).

3. Results and discussion

3.1. Stacking of green tapes

Two series of tapes of each composition were cast using different solid loading in the starting slurries. Table 3 shows how, for each composition, the green density increases with the solid loading in the slurry. The slurry with the largest alumina content presented better compaction capability.

In order to identify the defects associated with the stacking procedure, with no interferences of sintering related effects, the stacking of green tapes of the same composition was first investigated. Once optimized, the same method was applied to fabricate the laminate.

The engineering stress–strain curves corresponding to the compression of six wet (solid line) and six dry (dotted line) A-5YTZP stacked tapes is shown in Fig. 1. It is clearly observed the extreme differences between the behavior of wet and dry samples. Fig. 1a shows that the stacked wet...
tapes sustained higher pressures than the dry ones and also maintain an increasing slope for all the pressures. From about 30 MPa, increasing strains did not lead to stress increase in the dry samples, and variability increased. This is the typical behavior for a fractured sample where the stress is released by the opening of cracks. The maximum allowed pressure for working without cracking can be evaluated by analyzing the maximum in the derivative of the stress–strain curves in the low pressure regime (Fig. 1b), which will correspond to the inflexion point for the stress–strain behavior. From figure, the strain limit is 0.07, which corresponds to a pressure of 13 MPa.

On the other hand, the wet sample does not present a maximum in the derivative curve, meaning that no fracture nor important structural changes happened during the pressing process within the range studied for these samples (up to 90 MPa).

These changes revealed in the strain–stress curves can be clearly observed in the microscopical analysis after pressing. Fig. 2a shows clearly the macroscopic failure of a sample obtained using dry tapes after pressed at 30 MPa. This failure is a consequence of the low deformation capability of dry samples and did not occur in samples obtained using wet tapes even after pressed at 90 MPa (Fig. 2b). Fig. 2c and d show a micrograph for the cross-section of samples obtained with dry and wet tapes pressed below the joining pressure (10 MPa). In this picture, the fact that dry tapes developed cracks across the tapes before joining to other tapes can be also clearly observed. On the other hand, wet tapes did not develop these cracks perpendicular to the layers direction. In these samples, the lack of complete joining was revealed by the presence of linear arrangements of pores along the interfaces (Fig. 2d). From these complementary observations (strain–stress curves and microscopy after pressing) it can be determined from a structural point of view that water-based green tapes can be safely pressed and joined at relatively higher pressures if they are pre-treated by soaking in water before stacking and pressing.

Once established that appropriate pre-treatment of the tapes (wetting) generate a wide safe pressure interval, optimized pressure for adhesion between the layers was studied by analyzing the evolution of the green density of the compacts after pressing. In principle, optimum pressure will be the one that allows the achievement of a maximum interface contact without producing microstructural changes inside the individual layers. In order to determine this pressure, the green density of the compacts was compared with that of the single tapes. Stacked tapes shall be joined when no porosity is between layers, at this point the density of the stack of tapes should be the same that the density of a single tape. On Table 4, it is indicated the green density of A-5YTPZ tapes as a function of the single tape density.

<table>
<thead>
<tr>
<th>Pressure (MPa)</th>
<th>Density (g/cm³)</th>
<th>$p_{\text{piece/}}p_{\text{single tape}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>2.38±0.01</td>
<td>98.0±0.4</td>
</tr>
<tr>
<td>5</td>
<td>2.39±0.01</td>
<td>98.4±0.4</td>
</tr>
<tr>
<td>10</td>
<td>2.40±0.01</td>
<td>98.3±0.3</td>
</tr>
<tr>
<td>15</td>
<td>2.43±0.01</td>
<td>100.0±0.4</td>
</tr>
<tr>
<td>18</td>
<td>2.44±0.001</td>
<td>100.0±0.1</td>
</tr>
</tbody>
</table>

Density is indicated as the percentage of a simple tape density.

Table 4
Density of A-5YTPZ samples obtained by pressing stacked wet tapes at different pressures.

Fig. 3. Strain–stress behavior of A-5YTPZ(2)/A-40YTPZ(1) layered sample (diameter $\varnothing=60$ mm) up to the selected fabrication pressure (18 MPa). (a) Polynomial fit curve of experimental curves (error bars represent the standard deviation). (b) Derivative of the polynomial fit curve.
and A-40YTZP tapes with different green densities \((\rho_{A-5YTZP(1)} \neq \rho_{A-40YTZP(2)})\) and similar green densities \((\rho_{A-5YTZP(2)} = \rho_{A-40YTZP(1)})\).

Fig. 3 shows the strain–stress curves for the layered material \((\rho_{A-5YTZP(1)} s \rho_{A-40YTZP(2)})\). It can be seen that strains as high as 30% can be achieved by working with wet tapes and no inflexion point is detected indicating that the sample maintains its structural integrity in all the range of pressures. Very similar behavior was observed for the laminate made with tapes with similar green densities. For the monolithic and laminated materials described above, a pressure of 18 MPa was selected using a 5 wt% dilution of binder as a gluing agent, as it has been appointed from Table 4.

3.2. Sintering optimization of the laminate

The fabrication of multilayer ceramics from powder processing techniques involves the co-sintering of green joined layers. This is a critical point in order to obtain crack free layers. Different compositions sinter in different ways and stresses can be developed between layers if shrinkage levels and rates are very different. In this sense, the sintering behavior is very influenced by the packing in the green stage. Fig. 4 shows the linear shrinkage and the sintering rate curves recorded for the monoliths of the same compositions as those of the layers in the laminates. In Fig. 4a, curves for materials with very different green densities are plotted. It can be seen that sintering started at similar temperature for both materials, and that the dilatometric curves run close up to 1150 °C. After this temperature, they shrank in a different way giving a final shrinkage of 17% for A-5YTZP(1) and 22% for A-40YTZP(2). Also the maximum sintering rate was achieved at different temperatures (1400 °C for A-40YTZP(2) and 1460 °C for A-5YTZP(1)). On the other hand, the samples with similar green densities (Fig. 4b) showed different sintering behavior at low temperatures but finally they achieved similar shrinkages at 1350 °C, and continued together until a final shrinkage around 20% was reached. Also the maximum sintering rate was located at close temperatures (1410 °C for A-40YTZP(1) and 1425 °C for A-5YTZP(2)). The differences in the shrinkage and sintering rate of the different layers are made evident in the microstructure of the samples. Fig. 5a shows a tunneling crack developed in a A-40YTZP(2) layer during sintering of
A multilayer with $\rho_{A-5YTZP(1)} \neq \rho_{A-40YTZP(2)}$. As it can be observed in Fig. 4, this layer shrank more than A-5YTZP(1). This differential shrinkage would develop inside the layers a stresses system as the one represented in Fig. 6, where the A-40YTZP(2) layer is under tension while the A-5YTZP(1) is under compression. If tension stress overcomes the tensile fracture strength of the layer at the given temperature, a tunneling crack will be formed, as schematized in Fig. 6a. As the differential shrinkage continues, two processes would occur. First, the opening of the crack in the layer (Fig. 6b) and, second, the change in the stresses state in the adjacent layers with tension instead of compression in the crack tip (Fig. 6c). This can be enough to produce small cracks on the adjacent layers, as extensions of the main crack, which will propagate under the combination of tension and compression; thus, their path will be not straight. On the other hand, the multilayer shown in Fig. 5b will not stand sintering stresses because of the similar shrinkage levels of both layers through the whole temperature interval.

Tunneling cracks has been described to appear due to the tensile stresses that associated layers will generate when a differential strain between layers is present [28,57]. In the laminates here described differential strain can arise from two different mechanism, one the differential shrinkages and other the difference in the thermal expansion coefficients between layers. To check the fact that cracks were developed during sintering and not during cooling from the sintering temperature due to differences in the thermal expansion of the layers, a close up to the cracks was made by SEM. Fig. 7 shows the cross-section of a sample polished and chemically etched (HF 15 min). In Fig. 7a, the crack in the A-40YTZP(2) layer is presented. It can be observed how the grains located at the edge of the crack had round and smooth surfaces, indicating that crack underwent a thermal treatment after been created. If cracks would be generated during cooling, due to thermal residual stresses, sharp edges would be observed. Moreover, a low density linear zone extending into A-5YTZP(1) layers was observed (Fig. 7b). This observation is justified at the light of the tensile residual stresses, schemed in Fig. 6, that will oppose to the sintering shrinkage in a very narrow zone giving an

Fig. 5. Micrographs of the polished cross-section of A-5YTZP/A-40YTZP laminates with (a) different densities ($\rho_{A-5YTZP(1)} \neq \rho_{A-40YTZP(2)}$) and (b) similar densities ($\rho_{A-5YTZP(2)} = \rho_{A-40YTZP(1)}$).

Fig. 6. Scheme of the stresses developed during sintering into the layers. (a) The layer subjected to tensile stress generates a crack, (b) crack opening and stress state changes as the sintering goes forward and (c) tensile stresses extends crack in the adjacent layers.
aspect similar to a crack partially healed into the A-5YTZP(1) layer.

4. Conclusions

Fig. 8 is the main conclusion of this work: dense multilayer ceramics with strong joining between layers and free of defects can be obtained by stacking water-based tapes and pressureless sintering. The use of wet tapes and a glue agent, with the same composition that the tape binder, avoids defects in the stacks due to the pressure and allows the use of higher pressures than with dry tapes. By controlling the solid content of the starting slurries, it is possible to control the final green density of the tapes and consequently the shrinkage and sintering rate of the layers that compose the laminate. The differential sintering between layers develops tunneling cracks that extend into the tapes that supposedly are under compression. If tapes with the appropriate green density are selected, stresses due to sintering as well as the associated defects can be avoided.

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