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# Nano-structured Alumina-$ZrO_2$ ceramic laminates

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## ABSTRACT

In the last years many efforts have been expended to develop colloidal process that uses water instead of organic solvents in the tape casting process. In present work, alumina/zirconia laminated nanocomposites were fabricated by layer-by-layer method and using water-based tape casting process. Physical and mechanical properties as well as the fracture mode were investigated. The laminates consisted of stacked alumina and zirconia green sheets produced by thermopressing. The ceramic laminates were first heated at 450°C (organic elimination) and subsequently sintered in air at 1500°C during 1 h. The ceramic laminates showed a mechanical strength of approximately 103 MPa (AZAZA) and 44 MPa (ZAZAZ), respectively and an intergranular-transgranular mixed fracture mode.

**Keywords**: nanocomposites, laminates, tape casting .

## 1. INTRODUCTION

Nanostructured materials have received much attention in last decades, which can be attributed to the unusual physical and mechanical properties, such magneto resistance, unusual dielectric properties and high temperature mechanical properties [1-4]. The ability to sinter the powders into dense bodies and retain the grain size in the nano scale has attracted many researches in various fields from materials science to biotechnology, solar cells and in biology for tissue engineering. Tape casting is an effectively technique for making thin sheets and flat ceramic substrates and multilayer structures mainly for the electronic industry [5-7]. The characteristic uses for tape cast products were solid electrolytes for sensors and solid oxide fuel cells. Tape casting is a well-stablished method that consists in the preparation of a suspension of the ceramic powder in a solvent, with addition of a dispersant, binder and plasticizer [8-10].

The mainly factor is the control the rheological behavior of the slips, which will produce an adequate strength and flexibility to the green tapes, respectively. Recently water based tape casting process has been used, in order to avoid the toxic effects produced by organic solvents [11-12]. The use of alumina and zirconia as the constituent materials of ceramic laminates can be related to the excellent bonding between the layers in the absence of excessive diffusion between components, their good thermo-mechanical properties and their relatively ease of processing [13-14]. These characteristics make the two materials interesting candidates for the manufacture of ceramic laminates.

The literature about the fabrication and characterization of alumina-zirconia and other nanocomposites laminates are still scarce. Some recent works published in the literature reports some results obtained for composite ceramic laminates [15-20]. The laminated structure shows an improvement of the mechanical properties as compared to monolithic material and indicated that the physical, mechanical and electric properties are very sensitive to process variations. [20-23].

This increase is associated to the energy dissipation and also the crack propagating path that depends strongly on the interface property of the laminated composite material [22-24]. This study uses water instead of organic solvents in the tape casting process to produce a nanostructured laminate constituted by alumina and zirconia sheets. Physical and mechanical properties as well as the fracture mode of the laminates were investigated.

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## 2. MATERIALS AND METHODS

Commercial yttria-stabilized zirconia powder (TZ-3YE, 3 mol% $Y_2O_3$ stabilized $ZrO_2$, Tosoh, Japan) and alumina powder ($\alpha$–$Al_2O_3$ Taimei, Japan) were used in this study. Two different tapes using both the powders were separately produced by aqueous tape casting. The powder was deagglomerated in deionized water in a ball mill for 24 h with addition of a dispersant, respectively Darvan 821A in zirconia and alumina slurry. After deagglomeration, an acrylic emulsion binder (Mowilith LDM 6138, Clariant), a defoaming agent (Antifoamer A, Sigma-Aldrich), a surfactant (coconut diethanolamide, Stepan) and a plasticizer were added.

**Table 1:** Slurry composition (in wt%).

| | $ZrO_2$ | $\alpha$-$Al_2O_3$ |
|---|---|---|
| Ceramic powder | 55 | 55 |
| Binder | 25 | 25 |
| Dispersant | 1 | 1 |
| Defoaming agent | 0.5 | 0.5 |
| Surfactant | 1.5 | 1.0 |
| Plasticizer | - | 0.5 |
| Deionized water | 17 | 17 |

The slurry was mixed by ball milling for 120 min, and then cast at 25°C by a tape cast machine (CC-1200, Mistler) with moving polyethylene terephthalate carrier film coated with a fine silicon layer (Mylar G10JRM, Mistler). A casting speed of 200 mm/min was set. The gap between the blade and the carrier was adjusted manually to obtain a final tape thickness of 150 to 200 µm. The stability of the two slurries was analyzed by means of rheological characterization using a Haake Viscotester-Thermo Fischer Scientific viscosimeter with cone and plate geometry, at room temperature, and with shear stress between 0 and 800 s-1.The green tapes were dried at 25°C for 24h. Composition of alumina and zirconia slurries is showed in Table 1. The laminar composites were arranged as follows; five layers in the cast direction for each combination, where ceramic A is alumina Z is zirconia. Lamination was carried in a warm pressing between two metal plates at 60 °C, 19 MPa for 5 min. Details about the fabrication of both tapes can be founded elsewhere [9,16]. Debinding of the green laminates tapes was carried out by slow heating (0.5°C/min) up to 600°C with a dwell time of 1h. Then, the laminates were sintered with a heating rate of 5°C/min up to 1500°C with a dwell time of 1h. The microstructure of green laminates was analyzed using a high-resolution field-emission gun scanning electron microscopy FEG-SEM (Supra 35-VP, Carl Zeiss, Germany). Density of the sintered tapes was measured by Archimedes method. A mechanical testing machine (BZ 2.5/TS1T, Zwick/Roell) was used to measure the mechanical properties of the laminates with a crosshead speed of 5 mm min−1 based on the ISO 527-3 norm. For those mechanical tests, 7 rectangular laminar composites specimens (50 × 20 mm) were cut using a blade.

## 3. RESULTS AND DISCUSSION

### 3.1 Rheological charactertization

Figure 1 shows the typical rheological behavior of alumina and zirconia slurries. Both materials have shown a decrease of viscosity with the increasing shear rate, which is characteristic of a pseudoplastic behavior. At high shear rates the flakes are destroyed, causing a decrease of the viscosity, leading to a production of a homogeneous tape with smooth surface.

### 3.2 Microstructure and physical properties

Fig. 2 shows a dense ceramic laminate with strong joining and a visible interface. The laminate shows also some cracks that are characteristic of the tape process. Delamination effects can be also observed along the sample (figure 3) and is a consequence of the warm processing. Table 2 show the porosity and density values of the laminates. The Z-A-Z-A-Z laminate shows better values of porosity (13.67 %) and density (4.15 gcm-3) as compared to the A-Z-A-Z-A material, which can be associated to the better sinterability of the zirconia nanopowder.

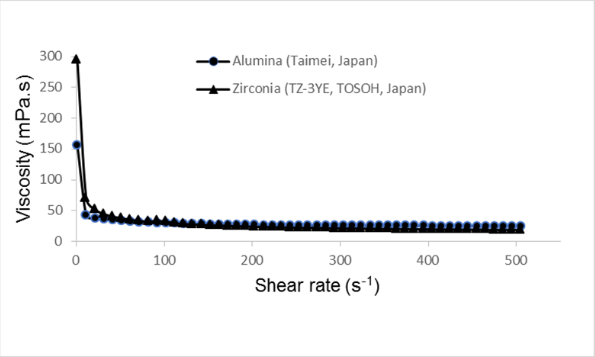

**Figure 1:** Viscosity as a function of the shear rate for the ZrO2 and Al2O3 suspensions.

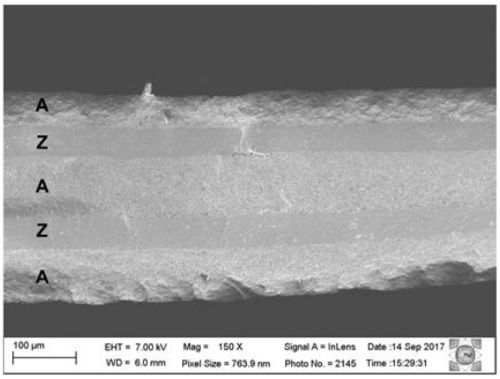

**Figure 2:** Schematic representation of the alumina (A) and zirconia (Z) layers in the respective AZAZA laminates.

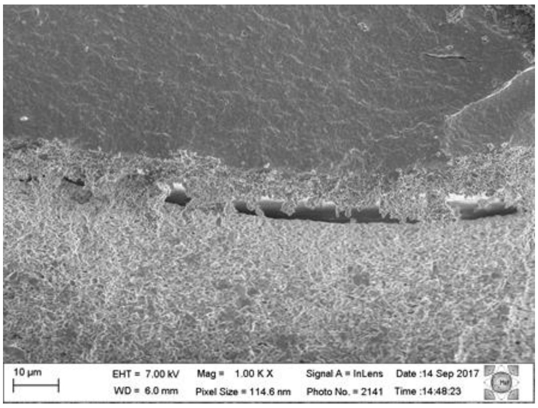

**Figure 3:** Typical micrograph showing delamination.

**Table 2:** Density and porosity mean values of the laminated samples with 5 layers after pressing and sintering.

| Condition | Density (g/cm$^3$) | Porosity (%) |
|---|---|---|
| A-Z-A-Z-A | 3,57 | 16,74 |
| Z-A-Z-A-Z | 4,15 | 13,67 |

### 3.3 Mechanical behavior

Figure 4 shows a representative stress-strain curve of the laminates tested in 3-point bending. Both samples presented the typical behavior of laminated composites, where the material deforms elastically until crack growth begins. The nanocomposites materials show several falls in loading that occur due to failure of the individual layers until a chain disruption. Each step of the load-displacement curve of the both laminated composite represents the break of one or several adjacent layers, according to other works [20-24]. The A-Z-A-Z-A laminate present a better strength value (103 MPa) as compared to ZAZAZ composite (44 MPa) and may be attributed to the higher density value and lower porosity value founded in this laminate (table 2). Large stresses are developed on cooling the laminate from the sintering temperature, due the higher coefficient of thermal expansion of the zirconia (10.5 x 10-6 C-1) as compared to alumina (7.2 x 10-6 C-1), at 40-400 °C. The existence of such stresses may be large enough to lead to failure of individual layers in the laminate contributing to the laminates strength values. This difference on expansion coefficient of zirconia may cause on cooling a tension state in zirconia and a compression state in alumina, also contributing to the strength results in the nanocomposites laminates. These results are in agreement with other works [16, 20-22]. It may be concluded that the resistance of the laminates depends strongly on the level of residual stress on its surface. The laminate with alumina layers on the surface reached higher values of mechanical strength, which can be associated to the compressive residual tension in this material and higher density values. The A-Z-A-Z-A nanocomposites laminate shows a strength value of approximately 100 MPa that is comparable with recent results reported in the literature for laminated-graded zirconia composites [22]. Studies about the use of higher temperatures and pressures during the warm pressing of the laminates are still under way in order to decrease the presence of laminate defects such as microcracks in order to improve the strength vales of the laminates.

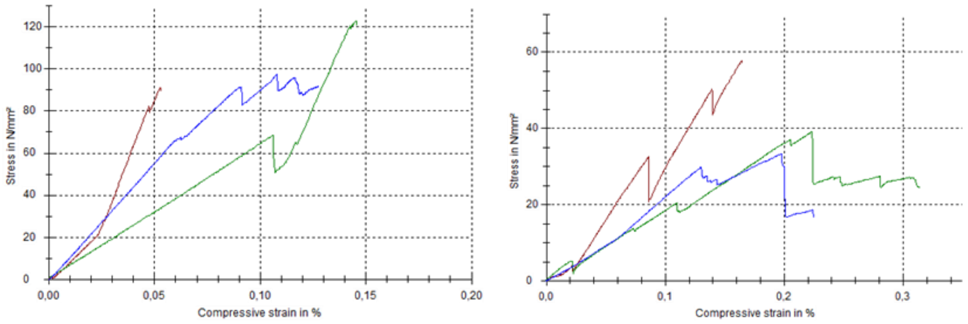

**Figure 4:** Stress-strain curves for the two conditions of laminates after 3-point flexural tests (a) A-Z-A-Z-A; (b) Z-A-Z-A-Z.

Figure 5 shows the fracture surface of the laminates after 3-point flexural testing. It is clear to see that the crack propagation develops differently in the components of the laminate. The alumina layer shows a predominant intergranular fracture mode, while the zirconia presents mainly a transgranular fracture mode.

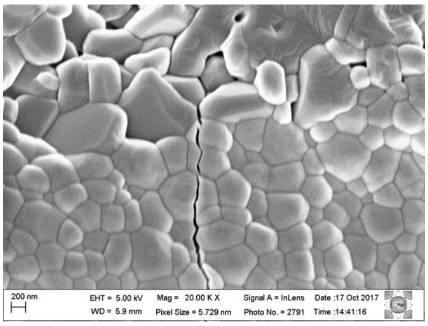

**Figure 5:** Crack propagation in Z-A-Z-A-Z and A-Z-A-Z-A laminates.

## 4. CONCLUSIONS

Alumina/zirconia laminated nanocomposites were fabricated by aqueous tape casting process. The results indicate a better mechanical performance of the AZAZA-nanostructured material, showing the nanocomposite laminate a strength value of 103 MPa. The lower strength in the Z-A-Z-A-Z laminates is caused by the presence of microcraks due the large stresses during the cooling from the sintering process. The propagation of the crack develops differently in the components of the laminate. In the zirconia layer, the crack develops mainly in a transgranular form, while in the alumina layer the predominant mechanism is the intergranular mode.

## 5. ACKNOWLEDGMENTS

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