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Neutron diffraction investigation for possible anisotropy within monolithic Al_2O_3/Y-TZP composites fabricated by stacking together cast tapes

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Abstract

The development of residual stresses in two Al_2O_3 + 5 vol.% yttria-tetragonal zirconia polycrystal (Y-TZP) ceramic composites fabricated by conventional slip casting and by joining green cast tapes was investigated. Neutron diffraction profiles revealed compressive microstresses (−200 MPa) in the Al_2O_3 matrix and tensile ones (2200 MPa) in the Y-TZP particles, irrespective of the processing route and the direction of measurement, which demonstrates the lack of residual macrostresses due to the joining procedure.

Keywords: Ceramics; Layered structures; Residual stresses; Neutron diffraction; Tape casting

1. Introduction

The production of multilayer ceramics is an area that is extensively researched. Initially developed in the 1960s as a result of the need for laminated structures for the packaging of microelectronics [1,2], the interest extended into structural applications and providing improved mechanical performance [3–6]. In particular, different laminate designs have been proposed in the system alumina–zirconia, for which several methods to obtain multilayer ceramics with controlled layer thicknesses have been developed. Most of them are based on joining green ceramic tapes fabricated by the tape casting technique [3,7–20]. In general, ceramic tapes are processed on the base of organic solvents with polymer binders and stacked together [7–11,16,18,19]. Subsequently, green state joining between the tapes is reached by applying pressure at temperatures above the glass transition point of the binder. Such methods involve the use of high contents of organic additives that may originate gradients in the green density during drying [7] and, therefore, anisotropy of the final product. The applied thermomechanical compression helps in minimizing these gradients, but cannot remove them completely. Moreover, the ceramic structures obtained via tape casting in the way described above present rather high porosity. Apart from these reasons, economic and ecological factors [8] forced the appearance of new lamination techniques for the manufacturing of laminated ceramics. These routes are based on joining procedures, which employ lower temperatures and pressures [12,15] and the fabrication of tapes from water-based suspensions with very low content of organic additions [13,14,19,20].

Several works have suggested better mechanical performance of monoliths fabricated from tapes in direction perpendicular to the constituent layers than that of the conventionally processed monoliths of the same composition [9–11]. Boch et al. [9] observed this effect for toughness values of Al_2O_3 materials and Chartier et al. [10,11]

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reported similar behaviour for toughness and flexural strength values of Al_2O_3–$mZrO_2$ specimens. In these works, relatively high temperatures (≈60–110 °C) and pressures between 15 and 64 MPa were used to join the tapes processed in organic solvents.

Preferred orientation of grains due to the tape casting procedure, investigated by X-ray diffraction, was dismissed as an explanation of such anisotropic behaviour [9]. Higher strength values have been attributed to the absence of large scale compaction defects and the greater homogeneity of the laminated monoliths [10,11]. Residual stresses due to anisotropic shrinkage of the laminate tapes or originated by the thermo-mechanical compression have been invoked as being responsible for strength and toughness increases [9–11].

Up to now, no conclusive experimental evidence of the presence or not of residual stresses has been provided. Nevertheless, the understanding of the effect of green processing on the properties of laminates is crucial to differentiate processing from compositional effects in order to develop new materials with improved properties. In this sense it is important to count with a suitable experimental technique that allows the analysis of residual stresses in the bulk of the materials, and thus is not affected by surface effects.

Neutron diffraction can be employed to measure non-destructively residual strains and stresses in the bulk of materials [21]. Residual stresses have been measured by neutron diffraction in several ceramic systems [22–25]. Wang et al. [22,23] investigated Al_2O_3/Ce-tetragonal zirconia polycrystal (TZP) composites fabricated by pressure filtration of aqueous suspensions with different volume fractions of tetragonal ZrO_2. The average residual stresses were compressive in the Al_2O_3 matrix and tensile in the Ce-TZP particles. Similar results were found by Lukas et al. [24] in functionally graded Al_2O_3/Y-TZP ceramics processed by electrophoretic deposition. In other work Todd and Derby [25] analyzed Al_2O_3/20%SiCp composites uniaxially hot pressed and found that the average residual stresses were in this case tensile in the alumina matrix and compressive in the SiC particles. Such residual stress states were always coincident with those expected from the thermal expansion mismatch between the phases. On the other hand, neutron diffraction has also been proposed as a means of establishing the lattice strain gradients for Al_2O_3 and Y-TZP in gradient materials [24]. In a similar way, the neutron diffraction can be used to determine the macro-residual stresses in laminated ceramics.

The objective of the present paper is to study the effect on the development of residual stresses in the sintered materials of a novel route for stacking, at low pressure (from 15 to 18 MPa) and room temperature, ceramic tapes fabricated by tape casting of water-based suspensions of Al_2O_3–Y-TZP mixtures [13,14]. Neutron diffraction was chosen for this purpose because of its capability to analyse stresses in the bulk of materials as discussed above. The final objective of the processing method developed is the fabrication of laminated structures with controlled macro-residual stresses originated by the thermal expansion mismatch of the different layers [26,27]. Therefore, this system is a characteristic case of the need for separating processing from compositional effects described above.

As a first approach to the problem, a single composition, Al_2O_3 + 5 vol.%Y-TZP, was investigated. Specimens fabricated by conventional slip casting were used as a reference material free from macro-residual stresses.

2. Experimental procedure

2.1. Material preparation

The materials to be studied were prepared using α-Al_2O_3 (Condea HPA 0.5, USA), with a mean particle size of 0.35 μm and a specific surface area of 9.5 m^2/g, and a *t*-ZrO_2 stabilised with 3 mol.%Y_2O_3 (TZ3YS, TOSOH, Japan), with a mean particle size of 0.4 μm and a specific surface area of 6.7 m^2/g, as the starting powders. A polyelectrolyte (Dolapix CE 64, Zschimmer and Schwarz, Germany) was used for powder dispersion. Two series of pieces with the same composition (A-5Y-TZP:Al_2O_3 with 5 vol.% of Y-TZP) were fabricated from stable slurries of the mixture of powders with 47 vol.% of solid content in deionized water. A 50 vol.% water-based latex emulsion Mowilith DM 765 E (Celanese, Spain) was used as binder (5 wt.% referred to ceramic powder).

One series was fabricated by slip casting of the deflocculated suspensions in plaster moulds and leaving them to dry for 24 h. Subsequently, the pieces were smoothed with sandpaper before sintering. The other series was fabricated by stacking tape cast tapes.

Tape casting was performed using a mobile container and fixed carrier tape casting device with Mylar® as the carrier film. After tape casting, the green ceramic tapes were dried at room temperature for 24 h and subsequently at 60 °C degrees for 48 h. The final thickness of the green tapes varied between ≈480 and 520 μm. Round shaped tapes were stacked to form a piece of 11 layers by using a gluing agent under uniaxial pressure of 18 MPa at room temperature. A detailed description of the processing steps regarding joining and pressing to obtain these specimens are given elsewhere [13,14].

Binder burn out and sintering of both kinds of pieces were performed in a single thermal treatment. The binder burn out was carried out with a heating rate of 1 °C/min up to 600 °C, followed by a dwell time of 30 min. Subsequent sintering was carried out by increasing the temperature with a heating rate of 5 °C/min up to the sintering temperature (1550 °C) with a dwell time of 2 h.

Sintered pieces were cut and ground to obtain the test samples with final geometry of $40 \times 40 \times 5\ mm^3$. In the pieces fabricated from cast tapes, the large surfaces ($40 \times 40\ mm^2$) were parallel to the surface of the stacked tapes.