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On the determination of the stress-free temperature for alumina–zirconia multilayer structures

Zdenek Chlup[a,*], Hynek Hadraba[a], Daniel Drdlik[b], Karel Maca[b], Ivo Dlouhy[a], Raul Bermejo[c]

[a]*CEITEC IPM, Institute of Physics of Materials, Academy of Sciences of the Czech Republic, 616 62 Brno, Czech Republic*
[b]*CEITEC BUT, Brno University of Technology, 616 69 Brno, Czech Republic*
[c]*Institut fur Struktur- und Funktionskeramik, Montanuniversitaet Leoben, 8700 Leoben, Austria*

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Abstract

Internal residual stresses can enhance the fracture resistance and mechanical reliability of layered ceramics. The magnitude of the stresses depends on the elastic and thermal properties of the layers and the typically assumed reference (stress-free) temperature, below which internal stresses develop. A novel combined experimental and numerical simulation approach has been developed to determine the reference temperature and experimentally proved in alumina–zirconia ceramic laminates. Dilatometric data of monolithic phases are input for the numerical simulation and experimental data on the laminate properties are used for the stress-free temperature determination. In contrast to typical assumptions, reference temperature very near the sintering temperature (i.e. approx. $T_{ref} \approx 1470$ °C) was found, which should be considered for the estimation of internal (residual) stresses in alumina/zirconia-based layered ceramics.

Keywords: A. Sintering; C. Thermal expansion; Zirconia; Alumina; Layered Ceramics

1. Introduction

The brittle fracture of ceramics is a consequence of the material defects located either within the bulk or at the surface, resulting from the processing and/or machining procedures [1–3]. Under external applied stress, the stress concentration associated with those defects is the common source of failure for ceramic components. If each defect is considered as a crack or a potential source for crack initiation, then it becomes clear that the size and type of these defects determine the mechanical strength of the material [4]. The distribution of defects within a ceramic component yields a statistically variable strength which can be described by the Weibull theory [5–7]. Since flaws are intrinsic to processing and in most cases unavoidable, the reliability of ceramic components in terms of strength is associated with such a flaw distribution. In an attempt to reduce the level of uncertainty in mechanical strength and to overcome the lack of toughness of monolithic ceramics, several processing routes have arisen in the last two decades which do not utilise the conventional "flaw elimination" approach, but rather use the implication of energy release mechanisms to obtain "flaw tolerant" (more reliable) materials, with improved fracture toughness. One strategy is to design multilayer structures combining layers of different materials. An effective way has proven to be layered ceramics with strong bond interfaces [8–16]. The strain mismatch between layers after cooling from sintering temperature (associated with differences in CTE and/or phase transformations [17–21]) can induce significant internal (residual) stresses within individual layers which can effectively change the crack path or even stop the crack during its propagation. In some cases layered ceramics can effectively act as a barrier to crack propagation, yielding a so-called "threshold strength", i.e. a stress level under which the material does not fail [10,12,22,23]. It has been recently shown that the layer thickness and exact ordering of layers (i.e. periodic or non-periodic) can be modelled by means of analytical and numerical calculations in order to obtain an optimal mechanical behaviour of the laminate in terms of strength and fracture resistance [24,25]. For tailoring

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[*] Corresponding author. Tel.: +420 532 290 335.
E-mail address: chlup@ipm.cz (Z. Chlup).
http://www.ipm.cz

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the design various methods including tape casting, slip casting and electrophoretic deposition (EPD) have been employed where a precise control of the kinetics during the deposition process is necessary to obtain accurate layer thicknesses [18,26–34].

The key feature in the design of laminates is the distribution of internal stresses in the layers, which depends on the elastic properties, thermal expansion coefficients, green densities, and volume ratio of the materials. The residual stresses in each layer may be assessed by means of experimental techniques such as X-Ray [35,36], neutron diffraction [37–40], Raman or fluorescent spectroscopy [41–43], among others. However, some limitations associated with the spatial resolution, edge effects, surface cracks, etc. may hinder the accurate stress measurement.

From the point of view of mechanical behaviour, the in-plane stresses in the bulk material (far from the free surfaces) can be calculated within each layer using the following equation:

$$\sigma_{\mathrm{res},i} = \frac{E_{\mathrm{i}}}{1-\nu_{\mathrm{i}}}(\overline{\alpha}-\alpha_{\mathrm{i}})\Delta T = \frac{E_{\mathrm{i}}}{1-\nu_{\mathrm{i}}}\Delta\varepsilon_{\mathrm{i}} \tag{1}$$

where E_{i}, ν_{i} and α_{i} are the Young's modulus, Poisson's ratio and coefficient of thermal expansion of the *i*th layer, respectively. $\Delta\varepsilon_{\mathrm{i}} = (\overline{\alpha}-\alpha_{\mathrm{i}})\Delta T$ is the mismatch strain of the *i*th layer. The coefficient $\overline{\alpha}$ is given as an averaged expansion coefficient of the thermal expansion of the laminate and depends on the materials volume fraction of the different layer types (for details see [24,25]). The temperature difference is $\Delta T = T_0 - T_{\mathrm{ref}}$, where T_0 is the room temperature and T_{ref} refers to the temperature above which the laminate is considered to be stress free.

The theoretical calculation of internal stresses in a layered structure using Eq. (1) can provide a good estimation only when exact input data are provided. The elastic properties and coefficients of thermal expansion and geometrical parameters can be obtained with high accuracy. However, the determination of the stress-free temperature above which stresses are relaxed seems to be the main course of uncertainty. Previous authors have assumed that for typical glass-phase free materials stress-free temperatures are in the range 1180–1300 °C [17,37,44]. They state that this temperature is influenced by the processing route, grain size, phase composition and/or cooling rate used after sintering [45–47]. Therefore it may be easily under or over estimated, thus resulting in inaccurate prediction of internal stresses. The present work introduces a novel approach to determine the stress-free temperature in layered ceramics based on combined numerical simulations and dilatometric measurements performed on alumina–zirconia laminates.

2. Experiments and simulations

2.1. Material of study

Materials for this study were prepared by the electrophoretic deposition technique. Two monolithic compounds (Al_2O_3 and ZrO_2) and a multilayer system combining both compounds in a layered structure were fabricated. The thickness of each layer was adjusted to approx. 50 μm, giving a volume ratio of approximately 1 between both materials see Fig. 1(a). Following powders were used: alumina (type HP-DBM, Malakoff Ind., USA) and tetragonal-zirconia stabilized with 3 mol% of Y_2O_3 (type TZ-3YSE, Tosoh, Japan) for manufacturing the monolithic Al_2O_3 (A), ZrO_2 (Z) and the alumina–zirconia laminate (L). The mean particle sizes of the alumina and zirconia powder were 470 nm and 140 nm, respectively. Isopropanol (p.a., Onex, Czech Republic) with addition of monochloroacetic acid (99%, Aldrich, Germany) were used for suspension preparation. The electrophoretic deposition was carried out in a constant current mode at 5 mA. Details about manufacturing process can be found elsewhere [28,32,33]. For all deposits three orientations were selected, where *X* was the deposition horizontal axis, *Y* was the vertical axis and *Z* was the axis in the direction of the deposition (see Fig. 1b). We caution the reader that the orientation may be important during the EPD process. The packaging may be affected by a number of factors such as (i) gravity (on the *Y*-direction), where heavier particles tend to deposit at the bottom part of the cell, (ii) shape of the powder particles, where particles have rather smaller dimension perpendicular to the electrode, (iii) electric field, which can orient particles based on their physical properties (i.e. domains oriented), and (iv) capillary forces which can affect the density of deposit in the vertical axis during withdrawing of the electrode with deposit from the suspension, etc. After the drying process, each deposit was annealed (800 °C/1 h; heating rate +2 °C/min in air) and finally cut and polished to the shape suitable for dilatometric measurements.

2.2. Dilatometer measurements

The relative length change of all specimens during sintering was monitored using a high-temperature dilatometer (L75/50, Linseis, Germany). The nominal sample geometry of $5 \times 5 \times 20\ \mathrm{mm}^3$ was used. Two different conditions covering standardly used conditions for sintering i.e. sintering temperature 1500 °C with dwell time 2 h; heating and cooling rate (5 °C/min or 10 °C in air) were applied for all materials. Two different heating and cooling rates were used to obtain an influence on the dilatometric results, if exists. Higher cooling rates were not tested because of physical limits of the furnace. Fig. 2 shows a scheme of the dilatometric set-up for both monolithic and laminate samples. The coefficients of thermal expansion were determined from the cooling part of the shrinkage curve fitted by a third order polynomial function of the form

$$\varepsilon = \frac{\Delta l}{l_0} = a_0 + a_1 T + a_2 T^2 + a_3 T^3, \tag{2}$$

where ε is the thermal strain given by the length increment Δl with respect to the initial length of sample l0. The polynomial fit provides better accuracy of measured data around the room temperature. Additionally it is possible to use instantaneous α