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Residual stress profile in ceramic laminates

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Abstract. Tape casting followed by high temperature compaction and sintering can be used for the production of ceramic laminates with a very high fracture toughness compared to their bulk counterpart. These enhanced properties can be obtained by suitable choice of the layers to be co-sintered in order to obtain a particular residual stress profile in the final component. The stress profile can be accurately measured by means of synchrotron radiation diffraction in energy-dispersive fixed-gauge-volume setup. The data for a set of alumina/zirconia/mullite laminates of ca. 1mm thickness, made of layers as thin as ca. 40μm are here presented and the results compared with theoretical predictions. In addition to the average stress profile, diffraction allowed to establish the stress in each of the different phases constituting the layers, thus showing coupling between the grains present therein.

Introduction

Traditional ceramic materials usually show brittle behaviour and low toughness. Strength values are highly scattered, as they strongly depend on the presence of defects such as cracks and scratches, introduced by the manufacturing process. Therefore, as the characteristics of such defects are heavily dependent on the conditions the material has undergone, the material has very low reliability because the fracture point is far from being clearly definite [1,2].
In order to enhance the strength of such materials, a possibility is to inhibit, limit or suitably guide crack propagation. Several techniques have been proposed to achieve this goal [3,4]. The simplest one is to create a low-energy path for crack advancement by adding soft or porous layers. There is no actual increase in strength but elongation and toughness increase significantly. Also, the material shows clear signals of imminent rupture before this happens. A smarter solution is to exploit a designed residual stress profile in the material [5,6]. If a compressive residual stress is introduced near the surface, then cracks must overcome the stored elastic energy barrier in order to propagate: the same principle is used to enhance the resistance properties of glass components (tempered glass). To achieve this, the traditional material is transformed into a *functionally graded* one. Functionally graded materials show a composition which changes along a spatial coordinates to obtain a specific property profile

(see figure 1) like, for instance, a residual stress profile. This can be obtained through co-sintering of a set of laminas showing different coefficient of thermal expansion.
Laminas are produced by dispersing a blend of ceramic powders in a binding organic phase which glues them as long as the ceramic is in the green state, and is then burnt out during sintering.

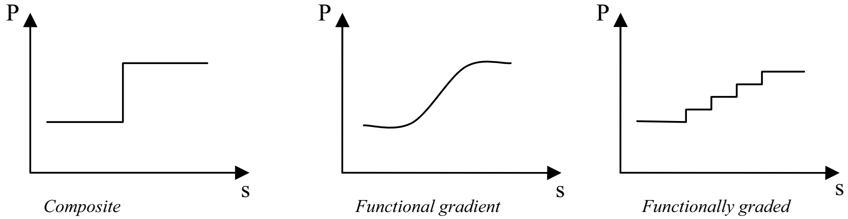

Figure 1. Distribution of a property across the sample thickness for three types of materials.

Synchrotron radiation possesses unique features for the non-destructive measurement of the residual stress in massive components. This is valid both for large (metre-sized) specimens, where thickness is the limiting factor but also for small specimens where the size of the gauge volume needs to be kept as small as possible. In the last case, the high brilliance (allowing very narrow beams to be employed), the availability of high precision positioning stages and the possibility of working with a white beam in energy-dispersive mode can, in principle, allow a 3D stress mapping with a resolution in the micrometer range. Station 16.3 at the Daresbury Laboratory (DL) possesses all the required features.
In this work a non-destructive test of the agreement between design parameters and actual status of stress in a ceramic laminate is proposed. A three-phase (alumina, zirconia, mullite) ceramic laminate was designed and produced: the residual stress was measured by means of a narrow-white-beam (10µm) through-thickness residual stress measurement on station 16.3 at the Daresbury Laboratory. As will be shown, good agreement between prediction and actual measurement is found.

Design and production of ceramic laminates

The procedure proposed by the authors of refs. [5] and [6] provides a method for calculating an optimal stress profile to be induced in the laminate material, and subsequently for determining the composition of each layer constituting it. In the case proposed here, the desired properties of each lamina were obtained by adding a suitable quantity of alumina to tetragonal zirconia (t-ZrO_2) or mullite ($3Al_2O_3{\cdot}2SiO_2$). Each mixture is identified by a name tag (AM for alumina-mullite and AZ for alumina-zirconia), followed by the weight percentage of the addition (e.g. AM40 stands for 60% alumina-40% zirconia). The specimen produced for this study, named AMZ-1, was made of 9 layers (see table 1). Further details on the materials can be found e.g. in ref. [5]. The laminates were produced from dry powders via the fast and inexpensive tape casting route. In this process, a slurry (made of the powder to be cast, a solvent, a dispersant and a binder) is cast by means of a (moving) doctor blade apparatus onto a rigid substrate, and then dried.

Table 1. AMZ1 laminate: design parameters. Layer 1 is the most external one (free surface). Just half laminate composition is proposed: the laminate is symmetrical with respect to layer 5.

Id	Type	Thickness (μm)	Starting depth (μm)	Average stress (MPa)
1	AZ30	36	0	32
2	AZ0	41	36	-336
3	AM40	93	77	-717
4	AZ0	41	170	-336
5	AZ40	522	211	178

The tape casting apparatus, in which the motion between doctor blade and substrate is obtained by moving the reservoir (moving blade), is shown schematically in figure 2. The result of this operation (green tape) is a single continuous flexible and deformable sheet that can be punched, blanked, drilled, cut and/or laminated very easily.

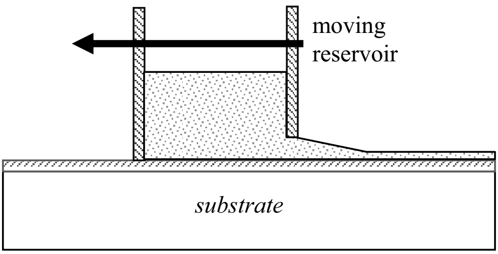

Figure 2. Doctor blade assembly (moving blade).

A set of sheets, as in table 1, was then stacked and thermo-compressed at low temperature (70°C, 30MPa for 15 minutes in present case) to give the green laminate. Specimens of nominal 60 mm x 7.5 mm x 1.5 mm size were produced in this way. The final laminate product was obtained by sintering the green in air at 1600°C for 2h. In order to allow all organic part to burn-out and to avoid problems in the subsequent sintering phase, the specimens were slowly pre heated (1°C/min) up to 600°C

White beam - synchrotron radiation stress measurement principle

The need for a stress tomography of the specimen, i.e. scanning without changing the characteristics of the gauge volume, does not allow the traditional residual stress measurement techniques to be employed in this case. In fact, in the well-known $\sin^2\psi$ [7,8] and η-rotation [7,8] techniques, the shape and therefore the penetration and/or extension of the measured volume change during the measurement, and the resulting stress value is an integral over a variable depth or a variable region inside the material.

An alternative technique can be employed, by using a white beam and an energy dispersive detector: working at low incidence angle and with a thin X-ray beam allows a large portion of the reciprocal space to be simultaneously measured on a finite and constant diamond-shaped gauge volume. Given the particular geometry of the specimen (and the presence of lamellas), three possible operation modes can be identified (see figure 3).

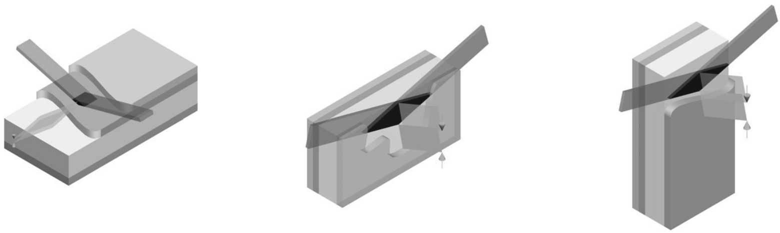

Figure 3. (a)-(c) Possible diffraction geometries for direct through-thickness stress mapping.

As the X-ray beam must enter and exit the specimen, and considering the spatial size of the latter, the most convenient geometry is the one proposed in figure 3c. It has the advantage of a limited divergence in the beam (the narrow equatorial i.e. horizontal size helps limiting the equatorial divergence whereas large intensity is available in the other direction where collimation is intrinsically better). In all cases proposed in figure 3, the specimen must be carefully aligned with the beam as to limit to the minimum the cross contamination of the signal from neighbouring laminas. The diffraction pattern is measured with the highest possible accuracy in the gauge volume for the stressed specimen and for a reference, where a zero or known stress is present: the result is obtained from the strain calculated as $\varepsilon = (d/d_0-1)$ where d and d_0 are the stressed and unstressed interplanar distances. As a multichannel energy dispersive detector is used, a careful calibration of channel versus energy (i.e. versus interplanar distance) is needed to obtain the high precision required for this kind of measurement. To this purpose, the stimulated X-ray emission spectra were measured for some elements (Cu, Ag, Mo and Tb, stimulated by a radioactive ^{241}Am source) and the corresponding peaks fitted. With a good approximation, the relationship between energy and channel was found being linear.

Residual stress results

Diffraction patterns were collected on the AMZ-1 specimen at 20μm intervals with a 10μm-wide by 1mm-high beam, using the geometry of figure 3c; further patterns were collected on single-layer stress-free AZ0, AZ30, AZ40 and AM40 specimens, used as reference. Figure 4 shows a diffraction patterns collected in the middle of the AZ30 layer: peaks corresponding to the constituting phases are clearly present. Pattern decomposition was employed to obtain information on intensity and position of each observed reflection. Intensity values were employed to identify the interfaces: the position where the intensity dropped halfway was assigned to the interface. Peak positions were used to determine the (local) strain at different positions. Stress values for each phase were calculated from the strain by using X-ray elastic constants for the given phase (obtained from literature mechanical data for the given materials [5]).

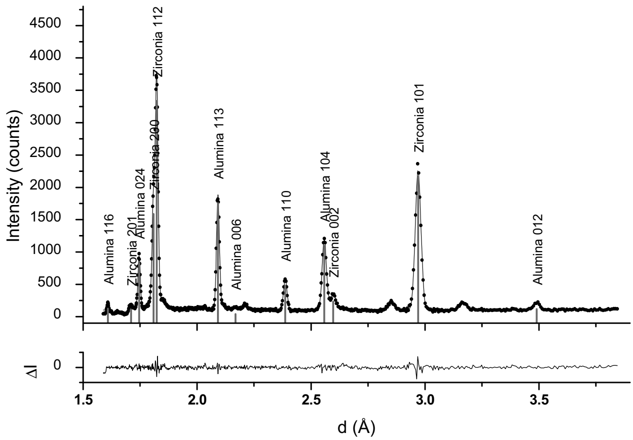

Figure 4. Diffraction pattern collected in the middle of the AZ30 layer.

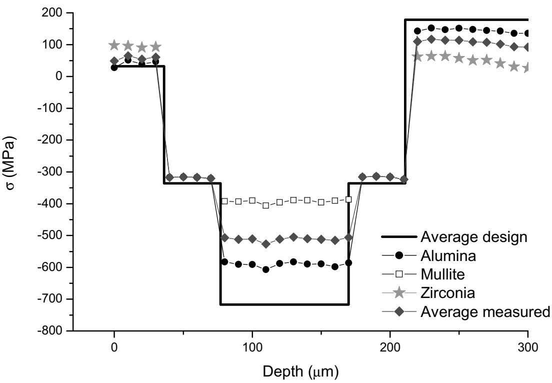

Figure 5. Stress profile for the AMZ-1 specimen: average value from design (line) and actual stress values in alumina (dots), mullite (squares) and zirconia (stars). The weighted average obtained from the single phase values is shown (diamonds). Error bars (of the order of 10%) have been omitted for clarity.

The stress in each phase of the laminate and corresponding total weighted average are shown in figure 5. The match with the predicted (design) stress is quite good, and confirms the expected sharp stress variation at the interface between neighboring layers.

Concluding remarks

Millimetre-thick ceramic laminates based on alumina with the addition of zirconia and mullite (to locally tailor the elastic and thermal expansion properties), have been designed and produced via tape casting followed by sintering. Each lamina, ca. 40 to 500 micrometer thick, was designed in order to obtain a well defined residual stress profile in the laminate, with the purpose of increasing the overall toughness of the specimen by guiding crack propagation. The residual stress profile was measured with a gauge size of about ten micrometers by using synchrotron radiation white beam. Peak intensity variations were used to determine the position of layer interfaces and peak position shifts were related to the residual stress. The result, a step-like profile with sharp variations at the interface between the laminas, agrees well with the designed stress profile, thus confirming the project expectations and the capability of this technique to map the through-thickness stress in complex, multi-layer components.

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