HIGH RELIABILITY CERAMIC LAMINATES BY DESIGN

Vincenzo M. Sglavo and Massimo Bertoldi

DIMTI, University of Trento, Via Mesiano 77, 38050 Trento (ITALY)

ABSTRACT

A design and processing approach has been identified and reduce to practice to obtain ceramic materials with high mechanical reliability, *i.e.* high failure resistance, limited strength scatter and increased damage tolerance. Different ceramic layers are stacked together to develop a specific residual stress profile after sintering. By changing the composition and the stacking order of the laminae it is possible to produce a material with predefined failure stress as it can be evaluated from the fracture toughness curve associated to the residual stresses. In addition, by tailoring the fracture toughness curve, surface defects can be forced to grow in a stable way before reaching the critical condition, thus obtaining an unique-value strength. In this way, reliable ceramic materials could be safely used in many structural applications. Laminates composed of alumina/zirconia layers designed and fabricated in this work through the proposed approach showed a "constant" strength of ≈ 600 MPa (standard deviation < 10%) even when large surface damages were produced, in very good agreement with the design value.

1 INTRODUCTION

Brittle materials, like ceramics and glasses, usually present limited mechanical reliability that represents the main reason of their scarce employ in structural applications. Many efforts have been made in the past to overcome such problems. Higher fracture toughness have been achieved through the exploitation of the reinforcing action of grain anisotropy or second phases or the promotion of crack shielding effects associated to phase-transformation or micro-cracking (B. R. Lawn [1]). As an alternative, fracture behaviour of ceramics has been improved by introducing low-energy paths for growing crack in laminated structures (J. B. Davis et al. [2], W. J. Clegg et al. [3], W. M. Kriven et al. [4], R. E. Mistler et al. [5], M. P. Harmer et al.[6]) or by introducing compressive residual stresses (C. J. Russo et al. [7], R. Latkshminarayanan et al. [8], M. P. Rao et al. [9]). Laminates presenting threshold strength have been successfully produced by Lange and co-workers (M. P. Rao et al. [9]) by alternating thin compressive layers and thicker tensile layers.

The idea that surface stresses can hinder the growth of surface cracks has been extensively exploited in the past especially on glasses (I. W. Donald [10], R. F. Bartolomew & H. M. Garfinkel [11]). Recently, Sglavo and Green have proposed that the creation of a residual stress profile in glass with a maximum compression at a certain depth from the surface can arrest surface cracks and result in higher failure stress and limited strength variability (V. M. Sglavo et al. [14], V. M. Sglavo & D. J. Green [15], D. J. Green et al [16]).

Residual stresses in ceramic materials can arise either from differences in the thermal expansion coefficient of the constituting grains or phases, from uneven sintering rates or from martensitic phase transformations associated to specific volume change. As described below, if the development of the residual stresses in ceramic multilayer is opportunely controlled, materials characterised by high fracture resistance and limited strength scatter can be designed and produced.

2 THEORY

In order to analyze the effect of residual stresses on crack propagation and failure resistance, the simple model depicted in Fig. 1 can be considered. The residual stress, which is a function of the distance from the surface only, is associated to a stress intensity factor defined as:

$$K_{res} = 2\left(\frac{c}{\pi}\right)^{0.5} \int_0^c Y\left(x/c\right)\sigma_{res}\left(x\right)dx \quad (1)$$

being Y a function of x/c reported in (D. J. Green [17]).

The generic system in Fig. 1 can be also subjected to external loads, associated to the stress intensity factor, K_{ext}. Crack propagation occurs when the sum (K_{res} + K_{ext}) equals the fracture toughness, K_C, of the material. If the residual stresses are virtually considered as a material characteristic, the apparent fracture toughness can be defined by combining K_C with the stress intensity factor associated to σ_{res}:

$$K_C^* = K_C - K_{res}. \quad (2)$$

If the simple situation shown in Fig. 2 is considered, the residual stress possessing a simple step-profile, K_C^* can be analytically calculated (Fig. 2). Assuming the presence of surface cracks smaller than a critical length, the material strength can be calculated through the graphical construction reported in Fig. 2. The addition or subtraction of step profiles like those in Fig. 2 allows the reproduction of the stress generated within a ceramic laminate constituted by laminae of different composition; then, by using the effects superposition principle, corresponding apparent fracture toughness can be evaluated. Referring to the generic profile in Fig. 1, if each step has an amplitude $\sigma_{res,j} = \sigma_{res,j\text{-}1} + \Delta\sigma_{res,j}$, the apparent fracture toughness in layer i ($x_{i\text{-}1}<x<x_i$) can be defined as:

$$K_{C,i}^* = K_{C,i} - \sum_{j=1}^{i}\left[2\psi\left(\frac{c}{\pi}\right)^{0.5}\Delta\sigma_{res,j}\left[\frac{\pi}{2} - \arcsin\left(\frac{x_{j-1}}{c}\right)\right]\right]. \quad (3)$$

where $\psi \approx 1.12$. Here the approximation is made that the elastic modulus of the different layers is constant (M. Bertoldi [18]). Nevertheless, the approximation in $K_{C,i}^*$ estimate does not exceed 10% if the Young modulus variation is less than 33% (R. J. Moon et al. [19], T. J. Chung et al. [20]).

The residual stress profile that develops within a ceramic laminate is related either to the composition/microstructure and thickness of the laminae and to their stacking order. According to the theory of composite plies (J. C. Halpin [21]), in order to maintain flatness during in-plane loading, if each layer is isotropic and the stacking order is symmetrical, the laminate remains flat upon sintering and its response to loading is similar to that of a homogeneous plate (J. C. Halpin [21]).

Regardless the physical source of residual stresses, their presence in co-sintered multilayer is related to constraining effect. When the different layers perfectly adhere each other, every lamina must deform similarly and at the same rate of the others. The difference between free deformation or free deformation rate of the single lamina with respect to the average value of the whole laminate accounts for the creation of residual stresses. With the exception of the edges, if thickness is much smaller than the other dimensions, each lamina can be considered to be in a biaxial stress state.

One fundamental task to properly design a symmetric multilayer is the estimate of the biaxial residual stresses. In the common case where stresses are developed upon cooling from differences in thermal expansion coefficients only, the residual stress in layer i (among n layers) can be written as:

$$\sigma_i = E_i^*\left(\overline{\alpha} - \alpha_i\right)\Delta T. \quad (4)$$

where a_i is the thermal expansion coefficient, $E_i^* = E_i/(1-\nu_i)$ (n_i = Poisson's ratio, E_i = Young modulus), $\Delta T = T_{SF} - T_{RT}$ (T_{SF} = stress free temperature, T_{RT} = room temperature) and $\overline{\alpha}$ is the

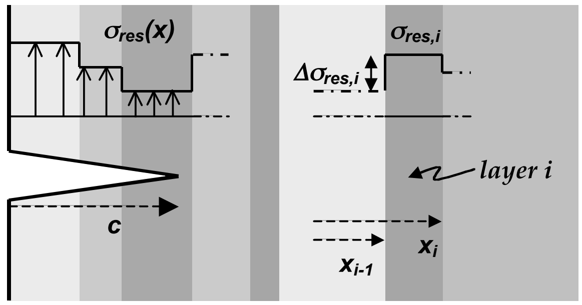

Figure 1: Schematic representation of the crack model used for the calculation of the apparent fracture toughness in a multilayered body subjected to residual stresses, $\sigma_{res}(x)$.

average thermal expansion coefficient of the whole laminate:

$$\overline{\alpha} = \sum_{1}^{n} E_i^{*} t_i \alpha_i \Big/ \sum_{1}^{n} E_i^{*} t_i \,. \tag{5}$$

t_i being the layer thickness. It has been previously shown that the stress free temperature represents the maximum temperature above which residual stresses are suddenly removed (M. P. Rao et al. [22]).

Equation (3) represents the fundamental tool for the design of a ceramic laminate with peculiar mechanical properties. Different ceramic layers can be stacked together to develop after sintering a specific residual stress profile that can be evaluated by Eq. (4). Once the associated K_C^* curve is calculated by Eq. (3), strength and fracture behaviour are defined. By changing the stacking order and composition of the laminae it is therefore possible to produce a material with predefined failure stress. In addition, if the shape of K_C^* is also tailored, surface defects can be forced to grow in a stable way before reaching the critical condition, thus obtaining an unique-value strength.

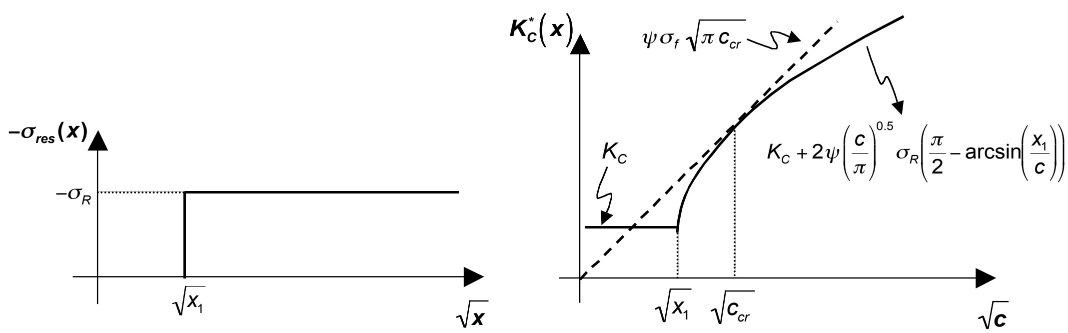

Figure 2: Step residual stress profile and corresponding apparent fracture toughness. The tangent straight dashed line is drawn to calculate the failure stress for crack lengths lower than c_{cr}.

3 EXPERIMENTAL PROCEDURE

In order to reduce the above concepts to practice, ceramic laminates with stress profiles specifically designed to improve the mechanical behaviour were produced and characterized. The thermal expansion coefficient as required for the development of the residual stress profile was tailored by considering composites in the alumina/zirconia (AZ) system for the production of the single laminae. Here the composites are labelled as AZzy where the last two characters correspond to the volume percent content of zirconia. Laminae of different composition were produced by tape casting, staked together and sintered as reported elsewhere (M. Bertoldi et al. [23]). In this work a stress-free temperature equal to 1200°C was assumed according to previous results (M. P. Rao et al. [22]).

Figure 3 shows the residual stress profile calculated by Eq. (4) for the laminate labeled as AZ-1 produced by stacking in the order the following layers: AZ40 ($t = 35$ μm) on the surface, AZ20 ($t = 35$ μm), AZ0 ($t = 90$ μm), AZ20 ($t = 35$ μm) and AZ40 ($t = 522$ μm); the sequence is then repeated in the reverse order to obtain a symmetric multilayer. The composition and thickness of the layers and their stacking order have been selected to produce a ceramic laminate with a "constant" strength of ≈ 700 MPa. The apparent fracture toughness curve and corresponding residual stress profile (Fig. 3) were correspondingly tailored to promote the stable growth of surface defects as deep as ≈ 150 μm.

Bars (6 mm x 48 mm) of the AZ-1 composite were fabricated. Edges were slightly chamfered to remove macroscopic defects and geometrical irregularities. No further polishing and finishing operations were performed on the sample surfaces or edges to avoid any artificial reduction of flaws severity. Sixteen samples were fracture by four-points bending tests. In order to establish the invariance of strength with flaw size, some specimens were also pre-cracked by Vickers indentation using loads ranging from 10 N to 100 N. Three indentations were produced in the centre of the perspective tensile surface. Monolithic samples (AZ0 and AZ40) were also produced and tested in the same conditions for comparison.

4 RESULTS AND DISCUSSION

The average bending strength measured on the AZ-1 samples was equal to 594 ± 59 MPa. The bending strength of the monolithic samples was equal to 418 ± 43 MPa and 741 ± 86 MPa for AZ0 and AZ40 laminates, respectively. One can easily observe that the strength values measured on the AZ-1 laminate are in good agreement with the design value. Conversely, the strength scatter is not substantially reduced. This can be explained by considering the size of the critical flaws as shown in Fig. 3. Most of the defects are shorter than ≈ 15 μm and propagate at stress levels higher than the designed strength, producing a scatter similar to homogeneous laminate. The designed stress, (≈ 500 MPa) corresponds therefore to the minimum value for the strength data, being indeed in a very good agreement with the minimum strength measured by the experiments.

Figure 4 shows that the failure stress of engineered AZ-1 laminates is also independent from the initial flaw size while, as expected, the strength of monolithic laminates is strongly dependent on the size of the indentation cracks.

The presence of stable crack growth phenomena was observed in the engineered AZ-1 laminates. The samples subjected to bending loads exhibited in fact multiple cracking before final failure, i.e. several cracks are formed on the tensile surface of the specimen above a specific threshold stress (≈ 300 MPa). Such cracks are arrested or undergo to stable growth and only one leads to the failure of the body. Such behaviour indicates that the material shows also some warning before final failure, this phenomenon being quite unusual in brittle materials.

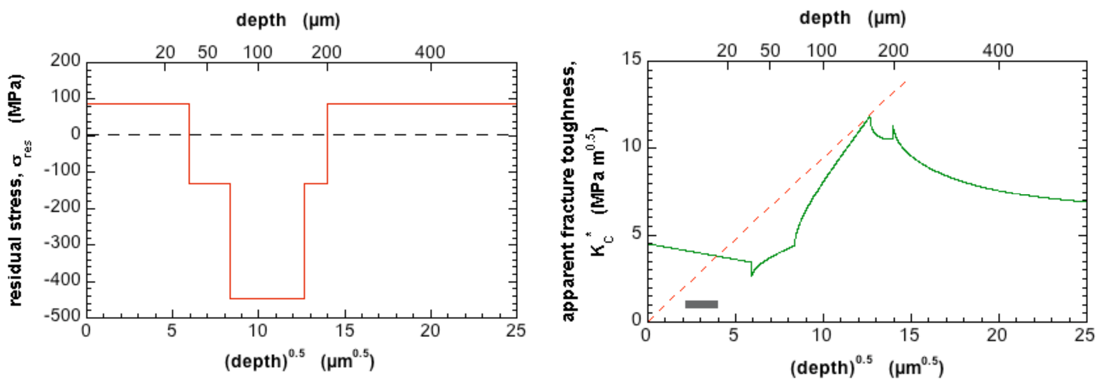

Figure 3: Design residual stress profile and corresponding apparent fracture toughness. The dashed tangent line shows the construction used for the calculation of the failure stress. The grey bar represents the flaw size interval.

4 REFERENCES

[1] B. R. Lawn, Fracture of brittle solids, 2nd Ed., Cambridge University Press: Cambridge, UK (1993).

[2] J. B. Davis et al., Fabrication and crack deflection in ceramic laminates with porous interlayer. J. Am. Ceram. Soc., **83** [10] 2369-74 (2000).

[3] W. J. Clegg et al., A simple way to make tough ceramics. Nature, **347**, 455-57 (1990).

[4] W. M. Kriven et al., High-strength, flaw-tolerant, oxide ceramic composite, U.S. Pat. No. 5,948,516, Sept. 7, 1999.

[5] R. E. Mistler et al., Strengthening alumina substrates by incorporating grain growth inhibitor in surface and promoter in interior. U.S. Pat. No. 3,652,378, March 28, 1972.

[6] M. P. Harmer et al., Unique opportunities for microstructural engineering with duplex and laminar ceramic composites. J. Am. Ceram. Soc.. **75** [7] 1715-28 (1992).

[7] C. J. Russo et al., Design of laminated ceramic composite for improved strength and toughness. J. Am. Ceram. Soc., **75** [12] 3396-400 (1992).

[8] R. Latkshminarayanan et al., Toughening of layered ceramic composites with residual surface compression. J. Am. Ceram. Soc., **79** [1] 79-87 (1996).

[9] M. P. Rao et al., Laminar Ceramics That Exhibit a Threshold Strength. Science, **286**, 102-5 (1999).

[10] I. W. Donald, Methods for improving the mechanical properties of oxide glasses. J. Mater. Sci., **24**, 4177-208 (1989).

[11] R. F. Bartolomew and H. M. Garfinkel, Glass science and technology, Vol. **5**, Ed. by D. R. Uhlmann and N. J. Kreidl, Academic Press, New York (1980).

[12] F. F. Lange, Powder processing science and technology for increased reliability. J. Am. Ceram. Soc., **72** [1] 3-15 (1989).

[13] B. C. Yu and F. F. Lange, Colloidal isopressing: a new shape forming method. Adv. Mater., **13** [4] 276-80 (2001).

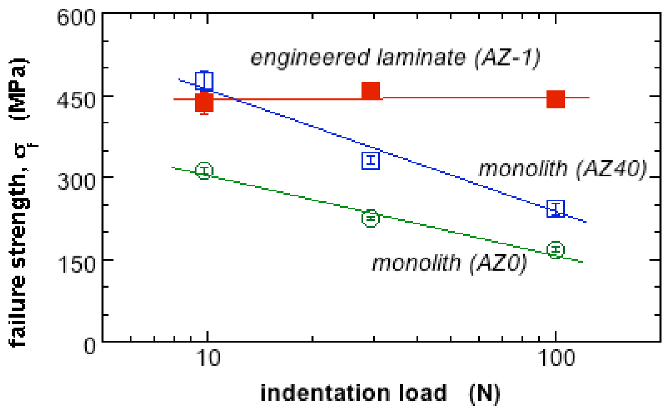

Figure 4: Failure strength data as function of the indentation load for the engineered AMZ laminate and monolithic samples (AZ0 and AZ40).

[14] V. M. Sglavo et al., Flaw-insensitive ion-exchanged glass: I, theoretical aspects, J. Am. Ceram. Soc., **84** [8] 1827-31 (2001).

[15] V. M. Sglavo and D. J. Green, Flaw-insensitive ion-exchanged glass: II, production and mechanical performance. J. Am. Ceram. Soc., **84** [8] 1832-38 (2001).

[16] D. J. Green et al., Crack arrest and multiple cracking in glass using designed residual stress profiles. Science, **283**, 1295-97 (1999).

[17] D. J. Green, An introduction to the mechanical properties of ceramics, Cambridge University Press: Cambridge, UK (1998).

[18] M. Bertoldi, High reliability ceramic laminates by design, PhD Thesis, University of Trento (Italy), 2004.

[19] R. J. Moon et al., R-curve behaviour in alumina/zirconia composites with repeating graded layers. Eng. Fract. Mech., **69**, 1647-65 (2002).

[20] T. J. Chung et al., Fracture toughness and R-curve behaviour of Al_2O_3/Al FGMs. Ceramic Transaction, Vol. **114**, 789-96 (2001).

[21] J. C. Halpin, Primer on composite materials analysis, Second Edition, Technomic Publishing Company, Inc., USA (1992).

[22] M. P. Rao et al., Residual stress induced R-curves in laminar ceramics that exhibit a threshold strength, J. Am. Ceram. Soc., **84** [11] 2722-24 (2001).

[23] M. Bertoldi et al., Production of multilayer ceramic laminates with improved mechanical properties, in Advances in Ceramic Matrix Composites IX, Ceramic Transactions, The American Ceramic Society, USA, Vol. 153, pp. 89-102 (2003).