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Effective fracture toughness in Al_2O_3–Al_2O_3/ZrO_2 laminates

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Abstract

During the processing of laminar ceramics, biaxial residual stresses can arise due to the thermal mismatch between different layers. For ceramic multilayers, the beneficial consequences of compressive stresses at the surface are well known: increase in strength, apparent toughness and reliability. Nevertheless, the resulting tensile stresses may induce a negative influence in the effective fracture toughness if the tensile stresses are high.

The weight function technique is used to assess the stress intensity factor corresponding to the residual stresses field. The influence of geometrical parameters such as thickness, number of layers and tension/compression thickness ratio is analyzed. For different multilayers ($Al_2O_3 - xAl_2O_3/(1-x)ZrO_2$), effective *R*-curves are presented.

The existence of an optimal architecture that maximizes the toughening is exposed as well as two tendencies on the apparent *R*-curve that define different fracture patterns: brittle failure or layer-by-layer fracture.

Keywords: Composites; Toughening; Al_2O_3; ZrO_2; Laminate

1. Introduction

Ceramic composites have a broad range of industrial applications. They have been extensively developed for structural components in order to improve the mechanical, chemical and thermal performance of engineering devices. However, despite a high hardness, an excellent oxidation resistance, and high temperature stability, ceramics are inherently brittle. One of the strategies to decrease brittleness is through the design of ceramic laminates with residual stresses.[1,2]

Laminates can improve mechanical performance since surface compression introduces a closure stress that protects against flaws. Two strategies of laminate design have been previously presented: first, laminates with a weak interface that deflects cracks, thus preventing catastrophic failure[3,4] and second, laminates with strong interfaces. Since strong interfaces will transmit residual stresses during cooling from sintering temperature, one can benefit of a phase transformation[4] or a thermal mismatch[5] to induce compressive stresses at the surface.

This paper examines laminates with strong interfaces, in particular multilayers made of alumina (A) and an alumina–zirconia composite (AZ). Those multilayers with an A-outer layer are shielded due to the minor thermal expansion of A compared to the composite AZ.

Although fracture toughness of a layered composite can be experimentally measured, it is only an apparent or effective value because of the influence of the residual stress. Besides, different shielding effects or intrinsic properties of the structure, such as bridging associated to grain size, rend difficult the interpretation of toughness measurements.

The apparent *R*-curve of a laminate can be calculated considering the equilibrium condition at the crack tip, i.e. crack propagation is possible if the stress intensity at the crack tip, K_{tip}, equals or exceeds the intrinsic material toughness K_0:

$$K_{tip}(a) \geq K_{c,0} \text{ being } K_{tip}(a) = K_{appl}(a) + K_{res}(a), \quad (1)$$

where $K_{appl}(a)$ is the applied stress intensity and $K_{res}(a)$ is the stress intensity contribution from the residual stress. Solving for $K_{appl}(a)$ holds

$$K_{appl}(a) \geq K_{c,0} - K_{res}(a) = K_{R,\text{effective}}, \quad (2)$$

where $K_{appl}(a)$ equals the desired effective *R*-curve. $K_{R,\text{effective}}(a)$.

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In fracture mechanics, both residual and applied stresses are usually included in the crack driving force. However, it is useful to consider the residual stresses as part of the crack resistance. Thus, in laminates with compressive stress at the surface, the higher resistance to failure results from a reduction of the crack driving force rather than from an increase in the intrinsic material resistance to crack extension.

The term $K_{res}(a)$ can – as an approximation – be assessed by means of the weight function approach,[6] that allows us to calculate the stress intensity factor $K(a)$, for an edge crack of length a for an arbitrary stress distribution acting normal to the fracture path. The weight function procedure developed by Bueckner[7] simplifies the determination of $K(a)$ since most of the numerical methods require separate calculations for each given stress distribution and each crack length. This method is of particular interest when the material is submitted to a "complicated" stress profile such as creep,[8] residual stresses in tempered glasses,[9] or residual stresses in multilayers.[2] Applying this concept to our residual stress profile σ_{res} results:

$$K_{res}(a) = \int_0^a h(x, a)\sigma_{res}(x)\,dx, \tag{3}$$

where $h(x, a)$ is the suitable weight function, a the crack length, and x is the distance from the surface.

Previous works by Fett et al.[10,11] validate the applicability of this methodology to inhomogeneous materials. The weight function presented in Eq. (4) was developed using the boundary collocation method.[12] It models materials with an homogeneous Young's modulus. It will be used as a first approximation. For inhomogeneous materials a suitable weight function will depend on $E(x)$. The consequences of using this simplified weight function for a laminate will be discussed later.

The weight function is

$$h(x, a) = \sqrt{\frac{2}{\pi a}}\frac{1}{\sqrt{1-(x/a)}(1-(a/W))^{1.5}} \times \left[\left(1-\frac{a}{W}\right)^{1.5} + \sum A_{\nu\mu}\left(1-\frac{x}{a}\right)^{\nu+1}\left(\frac{a}{W}\right)^{\mu}\right], \tag{4}$$

where the coefficients $A_{\nu\mu}$ can be found in.[8,12] W is the total thickness. It is worth of note that is not dependent on Young's modulus exclusively in the case of a homogeneous material.

In order to calculate the residual stresses in a laminate the following approximation was used.[13] Far away from the free surface,[14] the residual stress, σ_R, in each layer is uniform and biaxial. For the different layers A or AZ:

$$\sigma_{res,A} = -E'_A\frac{\int_{T_0}^{T_{sf}} \Delta\alpha\,dT}{1+((N+1)/(N-1)(e/\lambda))} \quad \text{and}$$

$$\sigma_{res,AZ} = -\sigma_{R,A}\frac{t_A}{t_{AZ}}\frac{(N+1)}{(N-1)}, \tag{5}$$

where $E' = E/(1-\nu)$, ν being the Poisson's ratio, $\Delta\alpha = (\alpha_{AZ} - \alpha_A)$ the difference of the thermal expansion coefficients of AZ and A, respectively, T_{sf} the temperature below which the residual stresses arise, T_0 the room temperature, N the number of layers, $\lambda = t_{AZ}/t_A$ is the layer thickness ratio, and $e = E'_A/E'_{AZ}$. The indexes make reference to the materials. Since the reason of the residual stresses is a deformation constraint (due to the thermal mismatch), the stress profile has to be proportional to the Young's modulus in the corresponding layer. For the assessment of the residual stress profile the different elastic moduli were considered.

Table 1
Properties of the material layers

	E (GPa)	ν	α_{tech} (10^{-6} K^{-1}) (0–1150 °C)	K_0 (MPa m$^{1/2}$)
A	391	0.241	8.64	3.8
AZ (60A40Z)	305	0.257	9.24	4.28

In this paper, symmetrical N-layer laminates with compressive stresses at the surface were studied (N being an odd number to fulfil the condition of symmetry). All the layers made of the same material (A or AZ, respectively) have the same thickness, so the laminate is well defined by the thicknesses t_A and t_{AZ}, or the total thickness W and λ. Through the paper, W will be considered constant and equals to $W = 1.5$ mm, according to a possible design condition. The corresponding effective R-curves are calculated according to the procedure explained above. The influence of the residual stress field, defined by geometrical and material properties, on the apparent R-curve is examined in detail. The results are expressed for the laminated system $Al_2O_3 - xAl_2O_3/(1-x)ZrO_2$, but the conclusions can be extended for any ceramic multilayer system with ideally strong interfaces.

2. Experimental procedure

This study entailed the use of a high-purity (99.7%) alumina powder (Alcoa A16-SG, Alcoa Aluminium Co., New York, NY) with an average particle size of 0.3 μm, and a zirconia powder (TZ3Y-S, Tosoh Corp., Tokyo, Japan) doped with 3 mol% Y_2O_3 with an average particle size of 0.3 μm. On the basis of previous experiments,[15] the different powders were mixed with organic binders, dispersants, plasticizers, and solvents to obtain suitable slips for tape casting. Slurry compositions were the same for both Al_2O_3 and Al_2O_3/ZrO_2 composite powders. Sheets of pure alumina (A) and of the composite containing 60 vol% alumina and 40 vol% zirconia (AZ) were produced.

Table 1 presents the material properties for the different layers, where E and ν were measured by impulse excitation technique, α by means of a dilatometer between 20 and 1200 °C and the intrinsic toughness K_0 following the VAMAS procedure (single-edge-V-notch beam in four point bending test).[16,17]

3. Results and discussion

As shown by previous authors, the apparent R-curve in multilayers presents an oscillating behaviour[3,4] (see Fig. 1). The toughness increases in the layers under compression with increasing crack length and reaches a local maximum at the interface. It decreases in the tensile layers reaching a local minimum

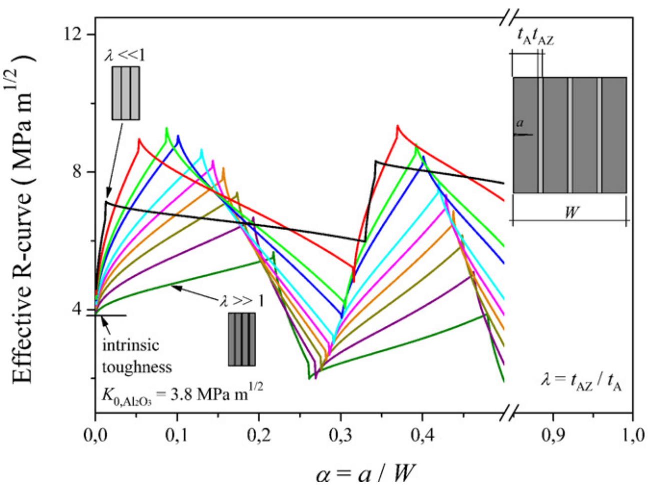

Fig. 1. Influence of the thickness ratio $\lambda = t_{AZ}/t_A$ on the effective *R*-curve. The situation $W = 1.5$ mm and $N = 7$ layers has been chosen to present the results.

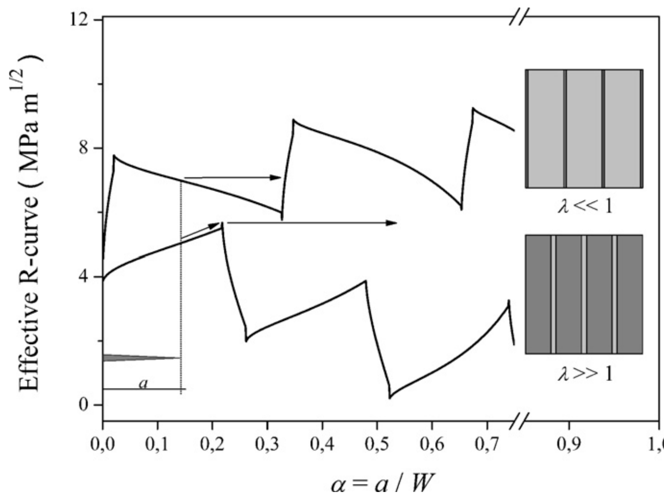

Fig. 2. Two clear tendencies provoking different fracture process.

at the interface. It can be stated that the compressive stresses shield the material against flaws, while the tensile stresses have a detrimental effect on the effective *R*-curve.

As it derives from Eq. (5), the architecture (λ) defines the residual stress field. It was the aim of this investigation to understand how the architecture influences the maximum shielding. In Fig. 1, apparent *R*-curves are presented for different values of λ in the range 0.2–25. Low values of λ corresponds to thin alumina layers t_A in comparison to t_{AZ}, and thus high compressive stresses are present in these layers. That is the reason why the shielding increases so steep in the alumina layers and a high stress intensity factor has to be applied to fail the specimen. For high values of λ, the thickness of alumina layers is much bigger than that of the AZ composite layers and as a result, high tensile stresses arise in the AZ layers, while almost no compressive stress appears. That is the reason that the effective toughness drops in the AZ layers for these laminates. This kind of multilayers, could even present for all the crack lengths a lower apparent toughness, so its mechanical performance is not so interesting as compared to laminates with $\lambda < 1$.

An interesting conclusion drawn from Fig. 1 is the existence of an architecture that maximizes the shielding in the first interface. Opposite to what could be expected, the highest surface compressive stress (the highest λ) does not correspond to the highest shielding in the first layer. Since the maximum shielding in the first layer is obtained at a distance equal to the outer layer thickness, the thickness t_A plays an important role.

This architecture that maximizes the apparent toughness at the first interlayer is especially interesting when short cracks are expected. Otherwise, for long cracks a laminate with $\lambda \ll 1$ could be more adequate due to the overall increase of toughness that is present in this type of multilayer.

We caution the reader about the fact that a weight function that applies to a homogeneous material (*E* constant) has been considered. This approximation results in an error of maximal 10% for the calculated stress intensity factor.[5] The A/AZ laminate contains an AZ core that is less stiff than the A. Compared to a situation with homogeneous stiffness, the A-layers carries more load and the AZ-layers less load, so that the calculated apparent toughness is overestimated in the alumina.

A second conclusion worth of note concerns the fracture process. As shown in Fig. 2, two clearly different behaviours are observed. In both cases, while the crack is propagating through layers under compression the shielding is increasing, reaching a maximum at the interface, but the overall tendencies are different. There are laminates for which the effective toughness presents an overall increase with crack length, while there are laminates that show an overall decrease.

Roughly speaking, those laminates in which the A-compressive stress is higher than the AZ-tensile stress, will present a tendency of toughness increase as long as the crack grows. Those laminates with a higher tensile stress present a tendency of toughness decrease, even reaching fictitious negative values of effective toughness. In the latter type of laminates, the fracture process results in unstable failure after reaching a peak in the *R*-curve. On the other hand in laminates with a tendency of toughness increase, a controlled layer-by-layer fracture pattern is observed. This behaviour has been experimentally observed carrying out 4-point bending tests.[18]

The architecture $\lambda = \lambda_{opt}$ that maximizes the shielding in the first interface, also deserves some attention. In Fig. 3, an envelope is presented covering the maximum shielding for each λ. Obviously all the maxima of these envelopes correspond to λ_{opt}. Fig. 3 also presents the influence of the different architecture parameters (*N* and *W*) on shielding. *N* modifies the residual stress field thus influencing the shielding and *W* normalizes the crack depth in the effective *R*-curve. The envelopes can be obtained by evaluating the effective *R*-curve at the first interface for each architecture. The reader should keep in mind that for this work the stress field considered is given by Eq. (5) that introduces some error in the outer layer since does not consider free surface. FEM calculations demonstrate that the difference is not significant in our case.[18]

As one can appreciate from Fig. 3 the architecture does not influence the position of the maximum. This means that the

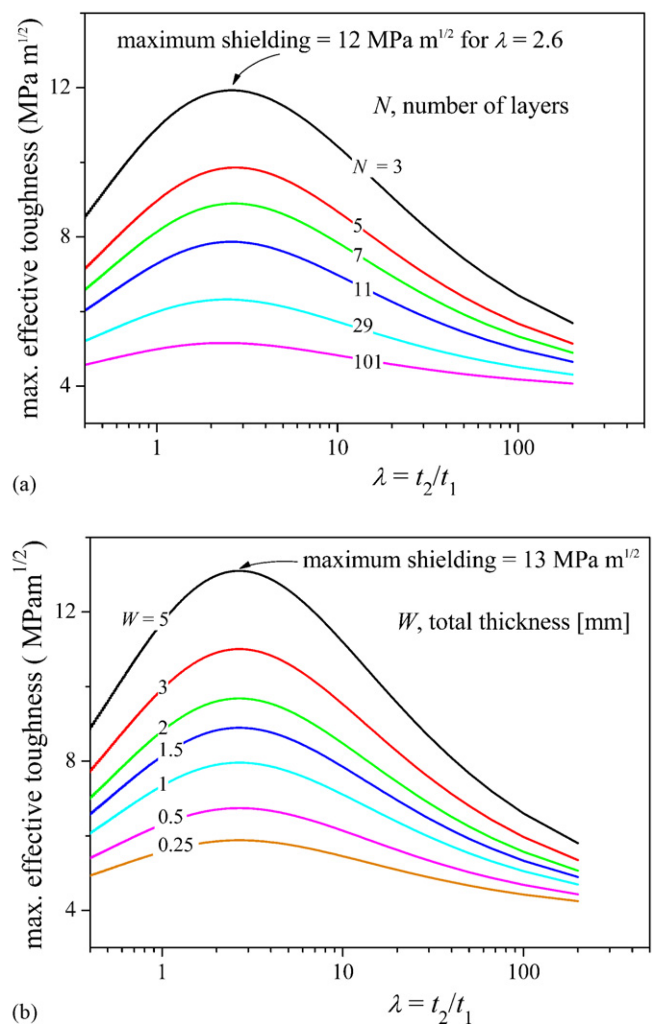

Fig. 3. Influence of architecture on shielding. (a) *N*: number of layers and (b) *W*: total thickness.

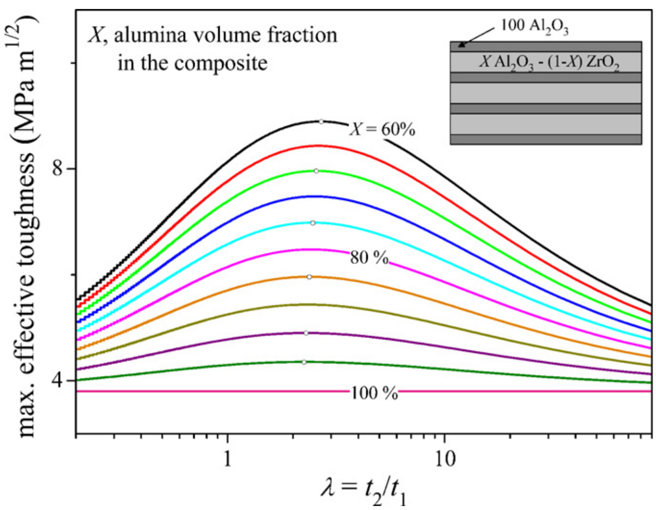

Fig. 4. Influence of the AZ-composite chemistry on the maximum shielding of the outer A-layer.

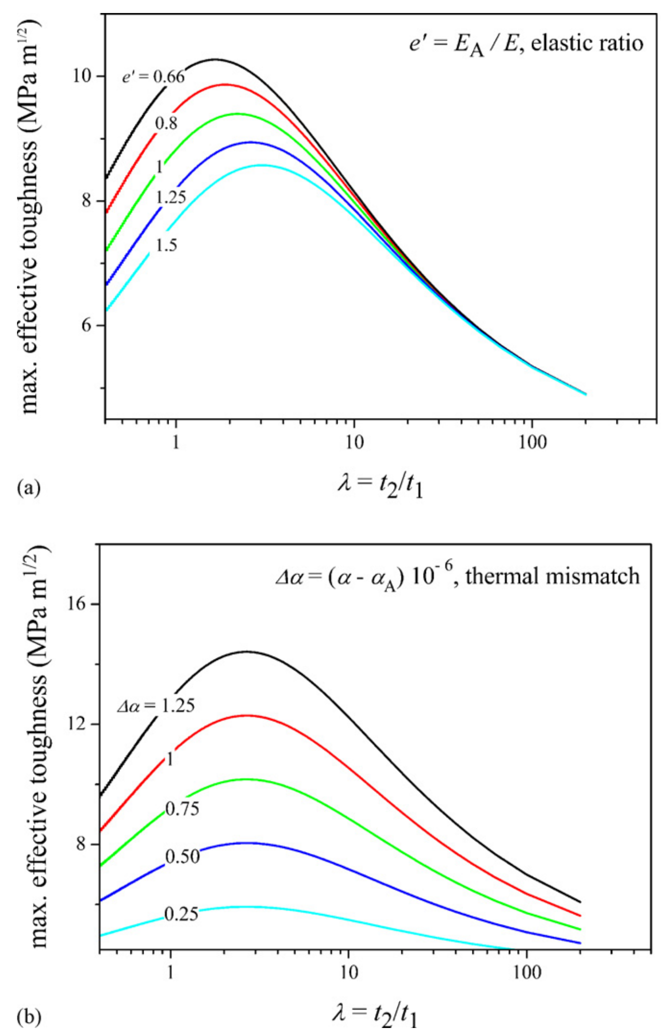

Fig. 5. Influence of the Young's modulus *E* and of the coefficient of thermal expansion α on the maximum shielding.

optimal architecture λ_{opt} is exclusively defined by the elastic constants. It also shows that shielding is more protective with a low number of layers and for thicker specimens. However, for relatively thick layers the authors expect a non-uniform stress field within the layers (Saint Venant principle) and the stress field considered here (Eq. (5)) would not apply.

The influence of the materials properties on the residual stress field is evident by Eq. (5). Fig. 4 presents the maximum shielding in A/AZ laminates for several compositions of the composite AZ. As one can observe the maximum for each composition is obtained for a different λ_{opt}. λ_{opt} varies from 2.25 for 95 vol% alumina to 2.7 for 50 vol% alumina. Properties of the different composites were estimated by applying the rule of mixtures, to the values presented in Table 1.

A more detailed analysis results in that exclusively the Young's modulus ratio influences λ_{opt}, and not the thermal expansion mismatch. Fig. 5a reveals how a stiffer material than alumina in the inner layer will increase the toughness. It results from Fig. 5b, the higher the thermal mismatch is, the higher is the compression in the outer layer and therefore, the higher is the shielding.

4. Summary

Attending to structural considerations an optimal architecture is presented for ceramic multilayers. An overall increasing tendency is observed in laminates with high compressive stresses that can provoke a controlled fracture process. Results are presented for a system alumina/composite alumina–zirconia for which realistic effective toughness up to 13 MPa $m^{1/2}$ can be expected.

Acknowledgments

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