THE ROLE OF TRANSFORMATION TOUGHENING IN PARTICULATE ALUMINA-ZIRCONIA COMPOSITES

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Sommario

La resistenza a frattura dei materiali ceramici è fortemente influenzata dalla attivazione di meccanismi di tenacizzazione. Fra i molti meccanismi riscontrati, in questo lavoro si è esaminato quello associato alla trasformazione di fase con espansione volumetrica caratteristico della zirconia parzialmente stabilizzata. Si è considerato il sistema composito particellato allumina/zirconia e se ne è ottenuta sperimentalmente la tenacità a frattura in funzione dalla percentuale di zirconia. La si è quindi analizzata in base ad un modello meccanico di frattura di una matrice con particelle soggette ad espansione evidenziando che il solo meccanismo di trasformazione di fase non è in grado di spiegare l'evidenza sperimentale.

Abstract

Individual toughening mechanisms affects remarkably the fracture resistance of ceramics. The role of stress-induced martensitic transformation of zirconia particles in the alumina/zirconia material system has been investigated experimentally. The fracture response of the several particulate alumina/zirconia composites was determined according to two techniques. A FE-based model of a propagating fracture in a model microstructure of transforming particles is presented. As the predicted influence of zirconia volume fraction is compared with the experimental evidence, it appears that zirconia transformation mechanism alone cannot explain the measured fracture toughness.

1. Introduction

The resistance of ceramics to crack propagation can be strongly influenced by microstructure and by the use of various reinforcements, [1]. Crack propagation in the microstructure activates different mechanisms, which may oppose or hinder further growth. This so-called toughening effect can act immediately when the crack starts to propagate or it may develop with crack extension. Therefore, toughening mechanisms may be classified into two categories: i) frontal shielding mechanisms acting ahead of the crack tip and ii) wake shielding mechanisms. Examples of the former category are crack front bowing, deflection and twisting, micro-cracking etc; examples of the latter category include stress-induced phase transformation, fibre and grain bridging etc. [1]. In spite of this categorisation, it is widely recognised that often more than one mechanism occurs simultaneously with a combined, enhanced effect.

The aim of this paper is the identification of the role of one such mechanism, associated to the stress-induced phase transformation of metastable zirconia particles, on the fracture toughness behaviour of the alumina/zirconia system. Zirconia-toughened aluminas are ceramic particle composites expected to be harder and more wear resistant than pure zirconia and tougher than pure alumina. In the experimental part of the work, composites with different percentage of reinforcing phase were fabricated and their fracture toughness experimentally determined according to two techniques, [2]. This experimental evidence is interpreted in the light of a model quantifying the role transforming zirconia particles on fracture response, [3].

2. Fracture toughening of zirconia/alumina ceramics

Zirconia has been extensively studied since the discovery of its tetragonal-to-monoclinic (martensitic) phase transformation, which is characterised by a large volume change (3-5%) and shear deformation (1-7%). When local stresses precipitate the phase change, transforming (i.e. expanding) particles have to be accommodated in the stable matrix with the stress distribution due to the matrix/particle compatibility superposed to those due to the external load, thus altering the crack tip stress intensity, [4]. The effectiveness of the mechanism is interpreted adopting the concept that the transformation stresses shield the crack tip from the nominal loading conditions. This martensitic transformation can potentially improve also the fracture toughness of a zirconia-containing ceramic composite. For example, the alumina-zirconia material system would combine the remarkable physical-mechanical properties of alumina, i.e. high wear resistance, and a superior fracture toughness compared to the monolithic material.

In parallel to material development, mechanics-based models of material behaviour should be put forward, [1]. Continuum fracture models have been formulated for partially stabilised zirconia, PSZ, and tetragonal zirconia polycrystals, TZP, materials as the transformation-affected zone surrounding the crack is expected to be significantly larger than the size of the individual metastable particle, [5-7]. In this way, the presence of a rising R-curve, was predicted with a toughening effect which increased from 25% to 35% the intrinsic toughness of the zirconia.

In the case of zirconia-reinforced alumina matrix, the particles embedded in the brittle matrix are relatively large when compared to the extent of the highly stressed region at the crack tip. Therefore, a finite-element-based model of the transient behaviour of a crack advancing in a matrix containing a regular distribution of transforming particles was developed in [3] to analyse the associated toughening effect. The influence of several material parameters, such as the volume percentage of transforming phase, particle arrangement and size on the crack growth response, was analytically quantified and it will briefly summarised in a subsequent section of the paper.

3. Experimental evidence

Materials and methods

The material tested were sintered single oxides: alumina, A, and 3% mol. yttria-stabilized zirconia, Z, and mixed oxides, homogeneous mixtures of 3% mol. yttria-stabilized zirconia and different percentages of alumina, denominated TZ3Y20A, TZ3Y40A, TZ3Y60A, TZ3Y80A, respectively with 20%, 40% 60% and 80 wt % of alumina, [8]. The average size of alumina and zirconia grains in the tested materials are reported in Table I.

The fracture response of the alumina/zirconia system was initially investigated with the indentation fracture toughness technique, [2]. The method is widely used to estimate the fracture toughness of ceramics from the length of cracks developing at the corners of a pyramidal-shaped impression left on the material surface by a Vickers indenter. Although many fracture toughness formulas (for the different crack systems, indenter geometry and material) are available in the literature, previous work, [9], verified the accuracy of the Anstis equation [10], used here for indentation toughness calculation.

Table I - Average size of alumina and zirconia grains in the tested materials.

Grain size, μm	TZ3Y80A	TZ3Y60A	TZ3Y40A	TZ3Y20A	Z
Al_2O_3	0.60	0.37	0.29	0.56	--
ZrO_2	0.25	0.25	0.36	0.41	0.57

One complicating factor of the indentation fracture method is the short crack lengths (< 500μm) involved. Therefore, relatively long-crack fracture tests were performed on prismatic bars (3 x 4 x 50 mm^3) of selected alumina/zirconia composites (i.e. TZ3Y20A and TZ3Y). The fracture testing method used consisted of a two-step procedure, [9]: i) a natural through-thickness pre-crack is introduced in the prismatic bar according to the bridge indentation (SEPB) technique, and ii) the fracture toughness test is then performed on the pre-cracked bar under four-point bending loading.

Fracture toughness data

A summary of the indentation fracture experiments on the alumina/zirconia materials system is given in Fig. 1. As previous investigations of the fracture behaviour of pure alumina, [11], showed the relevant influence of grain size on fracture toughness, a reference fracture toughness of 3 MPa√m was assumed for a pure alumina of micron sized grains. Fig. 1 shows that the response of the indentation fracture toughness as a function of zirconia content, which does not depend significantly on the indentation load. The limited long crack fracture data are also inserted in Fig. 1 (identified by 4PBS) and found to agree with indentation fracture tests. Interestingly, no significant increase in toughness is found up to a content of 50 % zirconia. Higher toughness is instead found at higher zirconia percentage, still compatible with the pure zirconia value of approx 4 MPa√m.

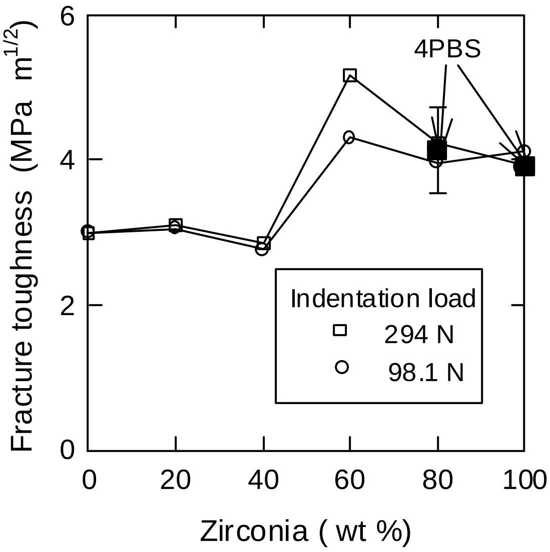


Fig. 1 - Fracture toughness of alumina/zirconia composites as a function of zirconia weight fraction.

4. A model of the transformation toughening mechanism

The fracture model of the transformation toughening mechanism, [3], is briefly reviewed as it will be used in the next section to discuss the experimental evidence. It is based on a two stress-intensity-factor approach, which assumes that local at the crack tip a small transformation-affected region exists. Outside this crack tip zone, the stress field is given by the linear elastic solution $\sigma_{ij} = K_0\ \Sigma_{ij}(r,\theta)$ where K_0 is the stress intensity factor determined by the applied load and geometry and $\Sigma_{ij}(r,\theta)$ are known functions from LEFM theory. Experimentally, the measured fracture toughness, K_c , is the critical value of K_0. Inside the toughening-mechanism-affected zone, the stress field is characterised by a local stress intensity factor, K_{tip}. Fracture propagation occurs when $K_{tip} = K_e$, where K_e is considered the intrinsic fracture resistance of the material. The increment in (measured) fracture toughness over the intrinsic toughness is termed toughening effect: it may be affected by any local mechanism and is characterised by the stress intensity variation $\Delta K = K_0 - K_{tip}$. When $K_{tip} < K_0$, the active mechanism (i.e. martensitic transformation in this case) shields the tip from the applied loads and the measured fracture toughness $K_c = K_e + \Delta K$. The stress intensity variation ΔK can act immediately when the crack starts to propagate or it may develop with crack advance. Typically, the crack resistance curve shows a gradual increase to a plateau: this steady state response gives the effective increment in fracture toughness.

Crack tip parameter calculation

In the modelling work of [3], a superposition approach was used to compute the relevant stress intensity factors.

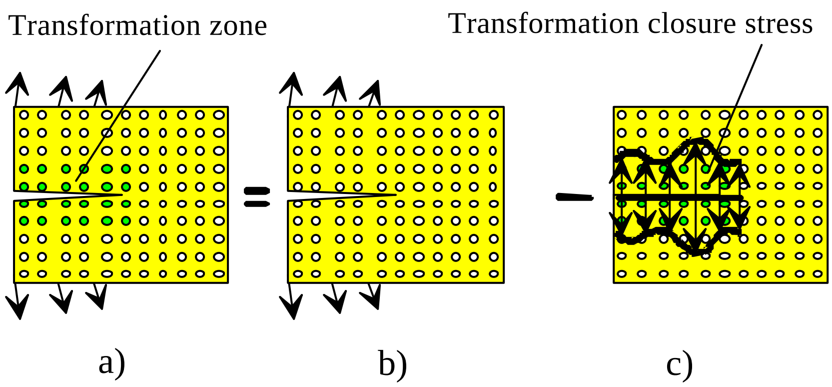


Fig. 2 - Particle transformation model: superposition approach for K_{tip} determination.

The scheme of Fig. 2 helps explain the approach: the transformation-affected K_{tip}, Fig. 2a, is obtained by superposition of the far field K_0 (no transformation), Fig. 2b, and the separate effect of transforming particles, Fig. 2c. Particle transformation is however constrained by the surrounding material and a contact stress distribution develops on the crack faces. As crack propagation must occur in the presence of an open crack tip, the closure stress distribution of Fig. 2c has to be removed. In [3], the closure stresses were obtained with a finite element approach for different particle fractions, size and distribution. The fundamental stress-intensity solution for a point force acting on the crack surface of an infinite sheet was used to evaluate normalised ΔK curves.

Model verification

Several models from the literature developed for 100% zirconia ceramics were used to assess the present modelling approach before its application to the alumina/zirconia system. The normalised toughening curves predicted by the analytical and numerical models of [5-7] are presented in Fig. 3 along with the present FE-based response. $\Delta a/L$ is the normalised crack increment. A reasonable correlation is found and the predicted saturation toughening is within the 25%-35% range. The present model shows a slight oscillation possibly of numerical origin.

The alumina/zirconia system

The main result of the application of the present model to the alumina/zirconia system is given in Fig. 4 in the form of (normalised ΔK vs. transforming zirconia content) plot. It shows that steady state toughening increases non-linearly with zirconia volume fraction. Lower and upper bound curves are shown as the oscillatory characteristic of the computed R-curve response is more significant in the particle composites than in Fig. 3.

In this case, it is affected by the presence of discrete transforming particles rather than a homogenised continuum. The trend presented above confirm previous studies, [1], and is justified by the competitive effects of the transforming zirconia particles as their percentage increases.

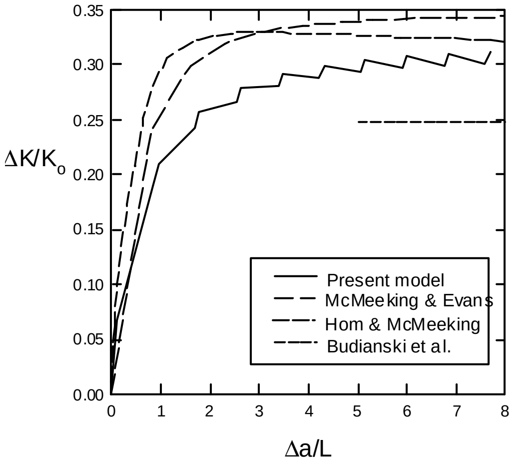


Fig. 3 - Predicted toughening response for a transforming zirconia.

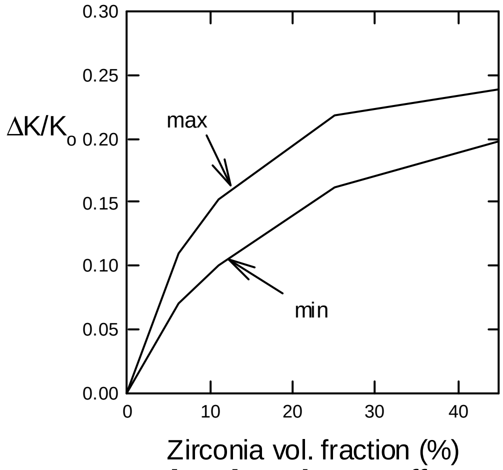


Fig. 4 - Predicted toughening effect as a function of zirconia volume fraction.

5. Discussion

In this section the previous analytical and experimental results are correlated and discussed to quantify the role of transformation toughening in the alumina/zirconia materials system with the help of Fig. 5. Inspection of Fig. 5 reveals that the toughening model overestimates the material response up to 50% of zirconia content in the alumina/zirconia composite. On the other hand, it underestimates the material response at very high zirconia content.

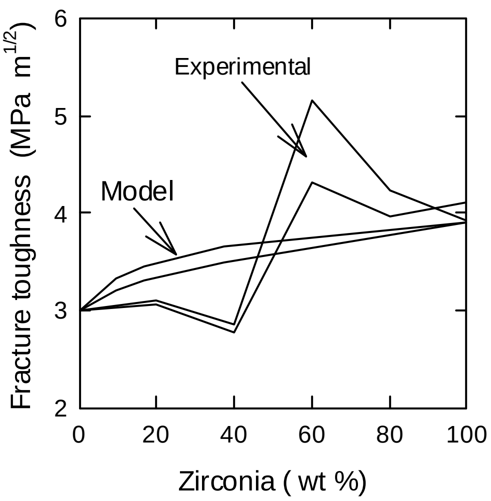
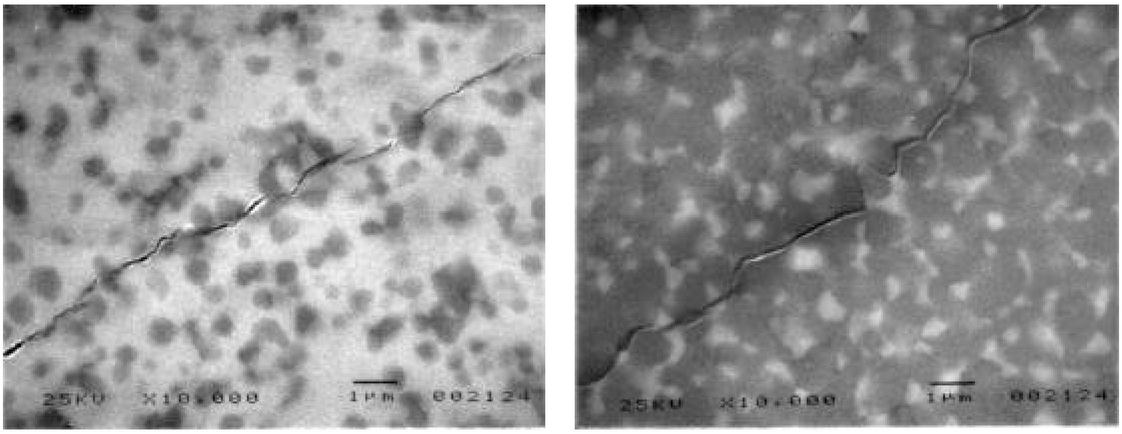


Fig. 5 - Fracture toughness of alumina/zirconia composites as a function of zirconia content: particle transformation model vs. experimental.

It has to be reminded that transformation toughening is but one of the different mechanisms identified to discuss fracture of ceramic materials and, in most instances, more than one occurs simultaneously. Toughening mechanisms such as crack front bowing, deflection and twisting and grains bridging are often found in monolithic ceramics. Secondary mechanisms such as residual stresses due to mismatch between the coefficients of thermal expansion are expected in particulate composites. Therefore, to obtain further insight in material performance an investigation in the scanning electron microscope of the microstructure-crack interaction was carried out, [12]. Examples of cracks in high and low zirconia composites are reported in Fig. 6a and b. The following is found:

- no grain bridging and crack deflection and a limited transformation effect are observed in pure zirconia materials;
- no significant crack deflection and higher transgranular fracture of zirconia grains is present in the 80% and 60% zirconia composites;
- in the low zirconia composite, no grain bridging is observed while crack deflection is more significant than in the previous case. Fracture occurs along alumina grain boundaries in according to its larger sizes and thermal expansion coefficient.



a) 20% alumina/80% zirconia. *b) 80% alumina/20% zirconia.*

Fig. 6 - Fracture path in selected alumina/zirconia composites.

These results can be explained considering that for this kind of zirconia, TZP, due to the very small size of the zirconia grains, a limited phase transformation capability is recognised, [13]. This effect, enhanced in the tested low zirconia composites, is able to justify their lower toughness values. For the high zirconia content composites, additional mechanisms to the martensitic transformation are activated. In particular, the presence of alumina causes a strengthening of the zirconia grains boundaries [14] and and activation of crack deflection, thus increasing the overall toughness of the composite with respect to pure zirconia .

6. Conclusions

The resistance of ceramics to crack propagation can be strongly influenced by microstructure design. In this work, experimental evidence obtained in the zirconia/alumina system has been discussed in the light of a mechanics model of the toughening associated to the stress-induced phase transformation of zirconia. Although the model correctly predicts a rising R-curve behaviour and a toughening response as function of zirconia content, the divergence with experimental results shows that microstructural differences, due to characteristics of the raw materials and sintering cycles, are able to strongly influence the martensitic transformation and activate different toughening mechanisms.

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