EFFECT OF THE BRIDGING MECHANISM
ON
CERAMICS TOUGHNESS

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Abstract: Grain localised bridging is one of the toughening mechanisms responsible for flaw tolerance in non-transforming ceramics. Bridging grains are wedged in the microstructure by internal compressive forces, which lead to an increase in fracture toughness as the crack grows (R-curve behaviour). The form of the R-curve depends upon factors such as the type, shape and size of bridging grains, and the residual stress field surrounding the bridging sites. Furthermore, when two different phases are present in the microstructure, wedging stresses are augmented by thermal expansion mismatch, which conducts to toughness improvement and to the possibility of exhibiting R-curve behaviour. In this article the main factors contributing to grain bridging enhancement are analysed, with special emphasis given to the effect of grain size in duplex (alumina-zirconia) ceramics.

Resumo: A existência de ligações locais entre os grãos é um dos mecanismos responsáveis pela tolerância às fissuras em cerâmicos não transformáveis. Os grãos ligados estão inseridos na microestrutura por forças internas de compressão, o que leva a um aumento da tenacidade à medida que a fissura se propaga (curva em R). A forma da curva em R depende de factores tais como o tipo, forma e tamanho dos grãos ligados, e o campo de tensões residuais que rodeiam os locais de ligação. Quando duas fases diferentes estão presentes na microestrutura, as tensões locais aumentam devido à diferença dos coeficientes de expansão térmica das duas fases, o que leva a um melhoramento da tenacidade e a uma maior probabilidade de exibir curva em R. Nesta comunicação são analisados os principais factores que contribuem para um acentuamento do mecanismo de ligação, dando-se especial ênfase ao efeito do tamanho de grão em cerâmicos duplex (alumina-zircónia).

1. INTRODUCTION

The brittle nature of ceramics, due to independent slip systems, ionic or covalent bonds and exhibition of long-range order, is continuously demanding an active research to increase their fracture toughness/resistance.

In the past, researchers tried to improve the fracture strength of ceramic materials by producing powders that were highly sinterable and ceramics with fewer flaws (conventional "flaw elimination" approach). Later, it was recognised that the resistance to damage in service was a further issue and that toughening these materials ("flaw tolerance" approach) could enhance their damage resistance [1,2].

Significant toughening of non-transforming ceramics can be achieved, at the microstructure level, via interaction of a propagating crack with the microstructure units (crack tip shielding mechanisms). In this case, the overall toughness (T) of the ceramic is [3,4]:

$$T = T_0 + T_\mu \qquad (1a)$$

or the crack resistance (R) is:

$$R = R_0 + R_\mu \qquad (1b)$$

where,

T_0 - intrinsic toughness

T_μ - extrinsic toughness mechanism (crack tip shielding)

R_0 - fracture resistance energy

R_μ - crack resistance energy contribution.

The critical condition for crack extension is then given by:

$$K_c = K_a = T_0 + T_\mu = T \qquad (2a)$$

or

$$G_c = G_a = R_0 + R_\mu = R \qquad (2b)$$

where,

K_c, K_a - critical and applied stress intensity factor

G_c, G_a - critical and applied mechanical strain energy release rate.

Due to the cumulative and irreversible nature of the crack tip shielding events, it is logical that the toughness of the ceramic increases with the crack extension (T- or R-curve-behaviour), which implies that the toughening term T_μ (or R_μ) is a function of the crack length, c. Therefore, eqs. (2a) and (2b) take the form:

$$T(c) = T_0 + T_\mu(c) \qquad (3a)$$

or

$$R(c) = R_0 + R_\mu(c) \qquad (3b)$$

Figure 1 presents a comparison of fracture strength (σ_f) and toughness versus the crack size (c) for materials exhibiting non-R-curve and R-curve behaviour.

The processes responsible for the crack tip shielding occur away from the crack tip, either in the region ahead of the crack tip (referred to as the "frontal zone") or behind the crack tip ("wake zone")- figure 2 [3].
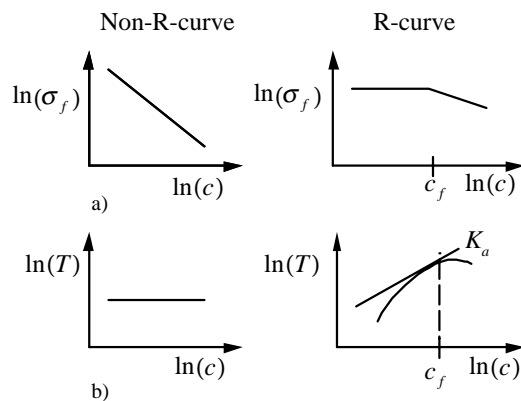

Figure 1 Comparison of materials exhibiting non-R-curve and R-curve behaviour; (a) for non-R-curve materials, the fracture strength (σ_f) decreases with increasing flaw size. R-curve materials, however, exhibit a range of crack size over which the fracture strength is invariant, i.e.,

they are "flaw tolerant"; (b) for non-R-curve materials, the toughness (T) is a constant, independent of crack size. For R-curve materials, the toughness increases with crack size. c_f denotes the crack size below which the fracture stress is constant (redrawn after Harmer et al. [1]).
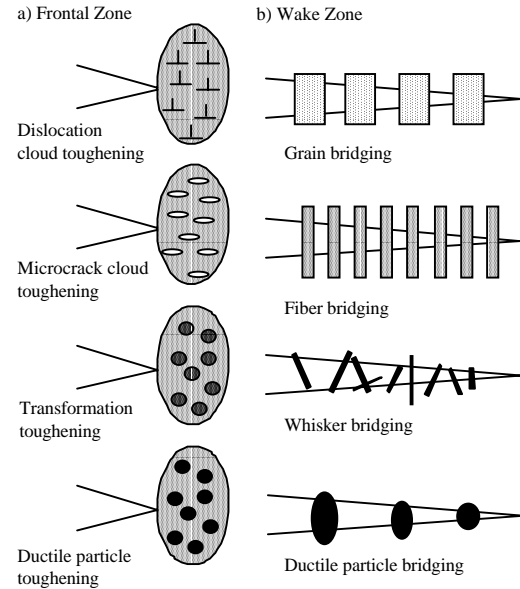

Figure 2 Schematic illustrations of crack tip shielding processes in ceramics (redrawn after Lawn [3]).

Although different mechanisms can be effective in toughening ceramics materials, in this article only the details of the grain localised crack bridging mechanism will be presented (for more information about the bridging processes in ceramics, see reference [2]).

2. GRAIN LOCALISED CRACK BRIDGING

Cracks in many polycrystalline ceramics follow the grain boundaries (intergranular fracture), thus interacting with the microstructure. Earlier, it was believed that such crack-microstructure deflections were responsible for toughness improvement.

In 1982, a research group led by Steinbrech [5] carried out a "re-notching" experiment on alumina (figure 3).

In this study, the crack resistance was measured during the stable extension of a crack in a notched bend specimen, made of a coarse-grain alumina, and it was demonstrated the importance of the

shielding events occurring in the wake of the crack tip. Essentially, they found that the crack resistance increases as the crack grows (a-b, fig.3(a)). Subsequently, they unloaded the specimen and carefully sawed out the wake of the extended crack, thereby advancing the notch close to the crack tip (position c)-the crack resistance was reduced to the original value at the bottom of the R-curve, instead of rising monolitically (b-d'). With further extension of the crack and development of a new wake (position d) the crack resistance was found to increase again [5].
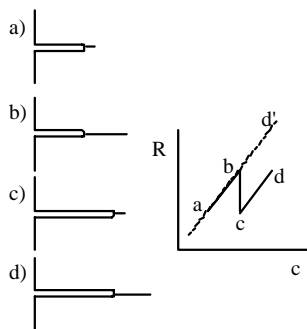

Figure 3 Knehans-Steinbrech experiment; (a) notched specimen; (b) propagation from the notch; (c) saw cut, to remove wake zone from extended crack; (d) re-propagation. Saw cut removes shielding tractions, as indicated by decrement b—c in R-curve (redrawn after Knehans and Steinbrech [5]).

Some systematic experiments on different grain size aluminas, have shown that the toughening observed is due to the shielding occurring with the increasing of grain size, thereby confirming a strong dependence of R-curve on the microstructure [1,3,6].

It was later realised that intact grain ligaments (grain bridges) in the wake of the crack tip exert frictional closure forces on the crack walls as the crack extends. Internal stresses are believed to control the bridge-restraining stress, which in turn increases with internal stress level. This fact dictates the amount of frictional energy dissipated during bridge pull-out and therefore the toughening contribution [7,8].

Optical microscopic observations carried out using both short-crack (indentation) and long-crack (double cantilever beam) test geometries in an alumina with pronounced T-curve behaviour have shown an interesting aspect: repeated "popping-in" of the crack very few grain dimensions by arrest, under an increasing load. Subsequent examination

of the crack wake during such loading revealed large-scale pullout of individual bridging grains from their respective sockets. A closer look at the bridging sites revealed frictional and geometrical interlocking between the bridging grains and the matrix grains. An examination of the entire crack showed evidence of intact bridging grains and the matrix grains. The density of bridging sites was found to be quite high, on an average two to three grain dimensions apart [6]. Figure 4 is a schematic illustration of the bridge formation and evolution.
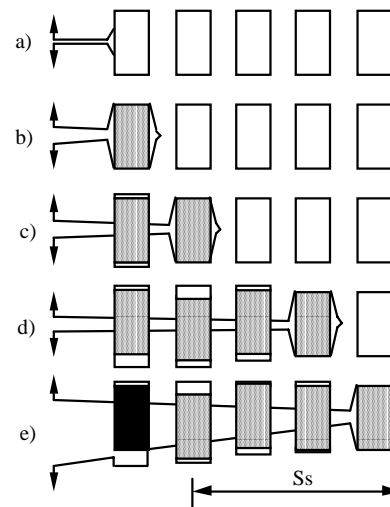

Figure 4 Schematic illustration of the bridge evolution and the successive formation of the bridging zone. Open grains denote potential bridges; shaded grains represent active bridges and closed grains disengaged bridges. (a) crack deflection, (b) debonding, (c) grain pullout, (d) expansion of the bridging zone, and (e) formation of the steady-state (Ss) bridging zone (redrawn after Padture [4]).

It was later shown that crack bridging is even more widespread in various other non-cubic ceramics [8].

The coarsening of alumina microstructures has been shown to lead to an improved long-crack toughness [4,7]. However, it has been observed, that when the grain size exceeds a certain critical value, non-cubic ceramics undergo spontaneous micro cracking (additional dissipation of energy takes place). This has been attributed to the presence of internal residual stresses associated with the thermal expansion mismatch/anisotropy in these microstructures [8,9]. The tensile component of these stresses acts to "open" sub-facet flaws. The size of such sub-facet flaws scales with the microstructure (grain size). Therefore, at some critical grain size (and sub-facet flaw size) the

stress intensity due to the internal residual stresses exceeds the intrinsic grain boundary toughness, and spontaneous micro fracture takes place.

From the exposed, it can be concluded that the grain localised bridging mechanism, in the ideal case, produces a ceramic where the fracture strength is independent of the flaw size (see figure 1). This means that the strength of the material is less sensitive to processing defects and accidents. Although this is a very important mechanism, it should be kept in mind that multiple toughening mechanisms are likely to operate simultaneously (for instance, bridging, micro cracking and transformation toughening can be present at the same time), and consequently the ceramic microstructures should be properly tailored [1-4].

3. SINGLE-PHASE/TWO-PHASE CERAMICS

The form of the R-curve depend upon such factors as:

1 – Size and shape of the bridging grains,
2 – Spacing between the bridges,
3 – Residual stress field surrounding the bridging sites.

It is well established that the R-curve behaviour of alumina is due to the formation of frictional tractions (grain bridges) between opposing crack faces in the crack wake [6]. One of the ways to enhance microstructural bridging in alumina is to increase the grain size. While this is effective in increasing the grain pull-out distance and hence promoting flaw tolerance, it does so at the expense of strength at small flaw sizes [10]. A more effective alternative is the introduction of a high number density of isolated elongated grains (effective bridge sites) into the microstructure [11]. However, single-phase ceramics are limited by the degree of anisotropy (which determines the maximum possible mismatch in thermal expansion coefficient between neighbouring grains). On the other hand, by judiciously incorporating the grain growth inhibiting second phase particles within the microstructure one can control the grain size and grain size distribution, and the degree of thermal mismatch [1,9,11]. For example, alumina-silicon nitride ceramics with strong R-curve behaviour, have a microstructure, which contains some large β-Si_3N_4 grains, presenting high peak fracture toughness 8-12 $MPA.m^{1/2}$ and high strength-800 MPa [12]. In two-phase ceramics, like Al_2O_3-c-ZrO_2 [13], Al_2O_3-Al_2TiO_5 [4], Al_2O_3-$3Al_2O_3.2SiO_2$ [14] and others, the residual stresses may be enhanced by the addition of the second-phase, whose thermal expansion coefficient shows the

desired degree of mismatch with that of the matrix. The Al_2O_3-Al_2TiO_5 system shows the greatest degree of flaw tolerance as a result of the highest degree of thermal mismatch [2]. It is important to note that if the microstructure scaled up beyond a certain limit the high thermal mismatch can give rise to micro-cracking [9,14]. In order to tailor a specific type of microstructure and enhance the fracture toughness in two-phase ceramics, different approaches have been used. For instance, using of a liquid phase at the sintering temperature, which upon cooling is retained (or not- transient liquid phase sintering) as a glassy or crystalline (after an heat treatment) phase, larger grains can be more easily obtained [1,13].

Figure 5 shows in situ SEM studies of crack propagation in an Al_2O_3-Al_2TiO_5 composite. The advancing crack tip was seen to be attracted to the alumina/aluminium titanate (A-AT) interphase interfaces. This observation implies that the high residual stress (and possibly the elastic modulus mismatch) due to the presence of aluminium titanate is instrumental in the formation of bridging elements in the A-AT composites.
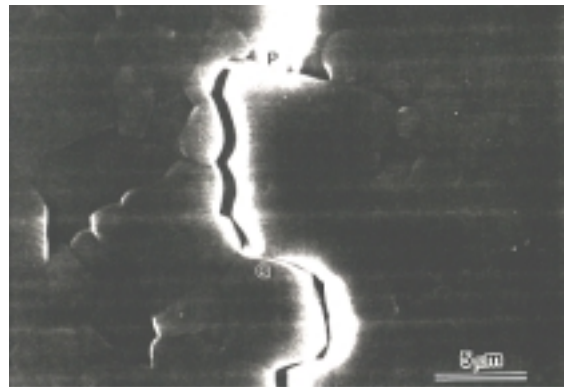

Figure 5 Micrograph of a bridging site taken in situ in the SEM (secondary electrons) during crack propagation in an alumina/aluminium titanate. P and Q denote frictions points during grain pull-out (courtesy of Nitin Padture).

In addition to increasing the level of residual stresses (by the addition of second phases), further enhancement in R-curve behaviour can be achieved by deliberate introduction of microstructural inhomogeneities (example: producing a "duplex-bimodal" structure by the addition of spray-dried agglomerates) [3,14]. Values of 8MPA.$m^{1/2}$ have been obtained in Al_2O_3-Al_2TiO_5 with bimodal structure [4].

In order to check for evidence of R-curve behaviour in a duplex (Al_2O_3-c-ZrO_2) ceramic, hereafter designed by AZ50 (50 % vol. of each phase); samples with different grain sizes were made.

Introducing indentation cracks and then checking for evidence of crack-wake bridging will evaluate the potential for the coarsest-grained duplex microstructures to exhibit R-curve.

In order to generate samples with very large grains (ideally around 10µm) [1,7], different amounts (0-10 % vol.) of anorthite glass ($CaO.Al_2O_3.2SiO_2$) were added to AZ50.

4. MATERIALS AND METHODS

Ultra high purity (99.995%) alpha alumina powder (AKP-HP, Sumitomo Chemical Company, Japan) with a mean particle size of 0.45 µm, cubic zirconia (8 mol % Y_2O_3, Tosoh Corporation, Japan) with a mean particle size of 0.59 µm, and anorthite (Alcoa Industrial Chemicals, USA) glass were used as starting powders. Glass particles smaller than 1 µm, were obtained by sedimentation under gravity. Powders were mixed in the correct proportions in 200-proof ethanol and ball milled (YTZ balls, Tosoh Corporation, USA) for 24 hours to produce AZ50 and AZ50 samples with 0.5, 1, 5 and 10 % vol. of anorthite. After milling, the slurry was magnetically stirred during drying and the dried cake was then crushed in a polyethylene bag with a teflon rolling pin.

Samples were uniaxially pressed in a cylindrical steel die, at 35 MPa, and isostatically pressed at 350 MPa. Pellets were cut in quarters, calcined in air at 950° C for 16 hours, then sintered in air at 1650° C for 1/2 hour. Densities were measured using the Archimedes method. Densities higher than 96% of the theoretical density were obtained for all samples. Pellets were then annealed in air at 1650° C for times ranging from 0 to 54 hours. All sintering and annealing treatments were conducted inside alumina crucibles. Samples were surrounded with calcined powder of the same composition, in order to minimise contamination and prevent volatilisation.

Pellets were polished [15], using standard materialographic techniques, and thermally etched at 1350° C or 1500° C for times ranging from 0.5 to 3 hours. Scanning electron microscopy (SEM-JEOL 6300F, JEOL, Japan), operated at 5 KeV, was used to characterise the microstructures. Grain sizes were determined from SEM micrographs using the linear intercept method (assuming a grain size of 1.56 times the mean intercept) with at least 400 grains/sample counted. The volume fraction of

each phase was evaluated with a transparent sheet containing a grid with 221 points, which was placed over each micrograph. The effective test line length was obtained by subtracting the line length intersected by porosity and/or liquid phase.

5. RESULTS

The goal of this research was the generation of duplex microstructures with large grain sizes for both phases (ideally around 10 µm), and evaluation of their potential to exhibit R-curve behaviour. Grain growth annealings (see figure 6) have shown that even after 54 hours, the maximum grain sizes obtained were only around 7 µm. If the annealing time were extended even further, 10 µm grains could be obtained, however this would not have any practical interest.
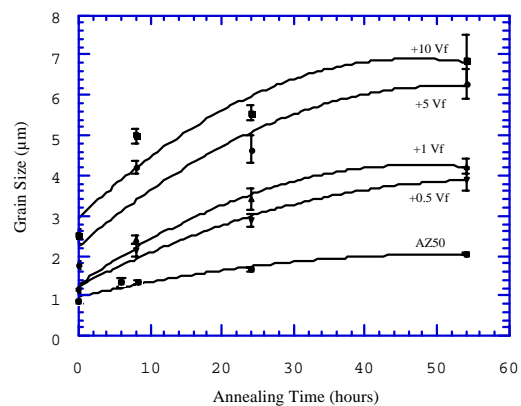
Fig. 6 Effect of volume fraction of anorthite glass (Vf) and annealing time, at 1650° C, on grain size of the AZ50 composite. Both phases have shown similar growth rates; therefore the grain size given represents the average for all grains.

Coarse-grained samples are not easily obtained, since it is well known [1,13] that in this system the limited grain growth rate is attributed to the limited mutual solid solubility, the increased diffusion path length relative to the single-phase materials, and the physical constraint provided by the two phases. Previous results obtained by the author [13] have shown that in this system grain growth is interface controlled, which means that a future approach to generate larger grains should involve an enhancement of the atoms transfer rate to the liquid phase.

In order to determine the toughness values, the indentation technique with direct crack

measurements, described by Antis et al. [16], was employed. Some cracks, made with a Vickers indenter (5 Kg load), were obtained in samples with different grain sizes, and the resulting microstructural features analysed in a SEM. Figure 7 shows how these cracks interact with the different phases in two samples with different grain sizes, 2 and 7 µm, respectively. All the samples observed exhibit a mixture of inter/transgranular crack propagation (similarly to what happens in samples with small grains) and also some bridges. The fracture toughness K_c measured from the indentation test is 3.1 MPa.m$^{1/2}$ for AZ50 (2 µm grain size) and 3.5 MPa.m$^{1/2}$ for AZ50 + 10 % vol. glass anorthite (7 µm grain size). This means that even in the samples with the largest grains, a significant enhancement of the crack-wake bridging mechanism, and consequently any significantly improvement in the fracture toughness, should not be expected.
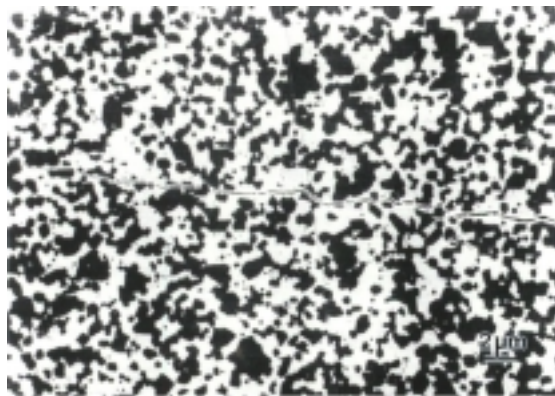
a)

b)

Fig. 7 Crack propagation in AZ50 samples with different amounts of glass and different grain sizes: a) AZ50 + 1 % vol. glass annealed for 0 h; b) AZ50 + 10 % vol. glass annealed for 54 hours.

These results are not a surprise, considering that the largest grains obtained are still small and that the thermal expansion difference between the two phases (α_{Al}=9x10^{-6}C^{-1}, α_{c-ZrO_2}=10x10^{-6}C^{-1}) [13] is very small to produce large residual stresses surrounding the bridging sites. In this aspect alumina-aluminium titanate seems to be a more promising material.

6. CONCLUSIONS

Grain localised crack bridging is one of the shielding mechanisms responsible for toughness improvement in non-transforming ceramics. In order to enhance the toughness is necessary to control some microstructural variables, namely, internal residual stresses and microstructural coarseness.

Increasing the grain size, increases the distance that the crack must propagate before it encounters a bridge, which conducts to an increase in toughness. However, coarsening of the microstructure by uniform grain growth (scaling) resulted in a relatively weak material and severe microcracking is susceptible to appear. Coarsening of duplex microstructures is very difficult, due to the limited large range interdiffusion, the physical constraint provided by the two phases and the limited mutual solubility of the two phases. Liquid phase sintering promotes faster grain growth rates and microstructures with larger grains, however the resistance can be decreased (although this can be minimised with the crystallisation of the glass). Liquid phase sintering (and transient liquid phase sintering) are promising opportunities to develop ceramics with tailored microstructures and high toughness (susceptible of exhibiting R-curve behaviour).

Duplex ceramics seem to have an improvement in flaw tolerance and R-curve properties over single-phase ceramics. An increase in residual stresses (change in the interfacial characteristics), derived from different thermal expansion coefficients of the two phases, appears to be the responsible for this observation. In AZ50 system, R-curve behaviour is not observed due to the difficulty in achieving large grains and the similar thermal expansion coefficients of the two phases.

Duplex bimodal composites, such as alumina-aluminium titanate, could exhibit even more flaw tolerance than duplex ceramics.

ACKNOWLEDGEMENTS

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