R-CURVE BEHAVIOR OF OXIDE CERAMICS

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Abstract. Brittle ceramics react in a more tolerant way to the presence of defects if toughness rises with crack extension. Macroscopic and micro-mechanical aspects of this R-curve behavior are presented for two oxide ceramics (Al_2O_3, MgO partially stabilized ZrO_2) and are discussed in terms of crack tip shielding mechanisms.

1. INTRODUCTION

In the last twenty years, ceramic materials have won increasing importance and significance for structural applications despite their inherent brittleness [1, 2]. Progress has been prompted both by the range of applications suitable for ceramics and by the development of improved materials [3-5]. In respect to materials improvements, great efforts have been and continue to be made to enhance mechanical properties through detailed microstructural design. Two alternative concepts for microstructural design which relate directly to the unstable behavior of ceramic materials containing flaws, and hence determine strength and toughness, dominate the developments.

Strength improvements can in the sense of the Griffith instability criterion, be achieved through a reduction in microstructural scale, provided that the critical flaw size responsible for failure, correlates with the grain size. Current efforts to develop high strength fine grain ceramics provide an impressive confirmation of this fracture mechanics concept [6].

However, the strength improvements that are won through attention to microstructural refinement are in themselves unable to reduce the extraordinary flaw sensitivity and the associated susceptibility to brittle fracture which is shown by ceramic materials. Local stress concentrations can again be responsible for catastrophic failure of the material.

An alternative concept for ceramics development has involved the search for materials which do not fail directly as a consequence of instabilities caused by existing defects but which rather show a relatively stable growth of initial flaws on loading. In contrast to the materials which have reduced flaw sizes but which nonetheless remain flaw sensitive, ceramics are now sought which can react in a tolerant way to the presence of defects. Crack tolerant behavior becomes possible if the toughness is increased with increasing crack length (R-curve behavior). Such an effect requires microstructural toughening mechanisms which involve increasing degrees of energy dissipation as a crack grows.

The present paper addresses toughening aspects of two oxide ceramics. The R-curve behavior of Al_2O_3 and MgO partially stabilized ZrO_2, both used for manyfold structural applications, are reviewed. Results of controlled, stable crack propagation studies are presented for long cracks and surface flaws, which elucidate the macroscopic R-curve behavior and the underlying microscopic mechanisms.

2. TESTING METHODOLOGY

Ceramic materials often fail in a catastrophic manner after a range of elastic behavior so that description of this brittle behavior can be treated with the help of linear elastic fracture mechanics. The crack instability criterion as proposed by Griffith

$$\sigma_B = \frac{1}{Y} \frac{K_{IC}}{\sqrt{a_c}} \qquad (1)$$

combines strength, σ_B, and toughness, K_{IC}, with the critical crack length, a_c. Here Y is a parameter which depends on the crack and specimen geometries.
In case of R-curve behavior stable crack growth occurs prior to instability [7] and equation (1) thus becomes

$$\sigma_B = \frac{1}{Y} \frac{K_R(a)}{\sqrt{a_o + \Delta a}} \qquad (2)$$

with $a = a_o + \Delta a$.

To measure the rising toughness, K_R (a), controlled, slow crack propagation studies rather than fast fracture experiments are required.
Experimentally such controlled crack growth is achieved if the change in crack driving stress intensity with incremental crack growth is smaller than the respective increase in toughness [7].

Specimen geometries typically used in fracture mechanics tests (SENB, DCB, CT (Fig. 1)) satisfy this condition if they contain deep precracks or starter notches. Stable growth over several milimeter of crack extension is possible and thus such long crack measurements provide information about the long range toughening potential of a ceramic. On the other hand, short crack measurements (Fig. 1), covering the growth of "natural" surfaces cracks, reflect more realistic the impact of the R-curve effect on component failure.

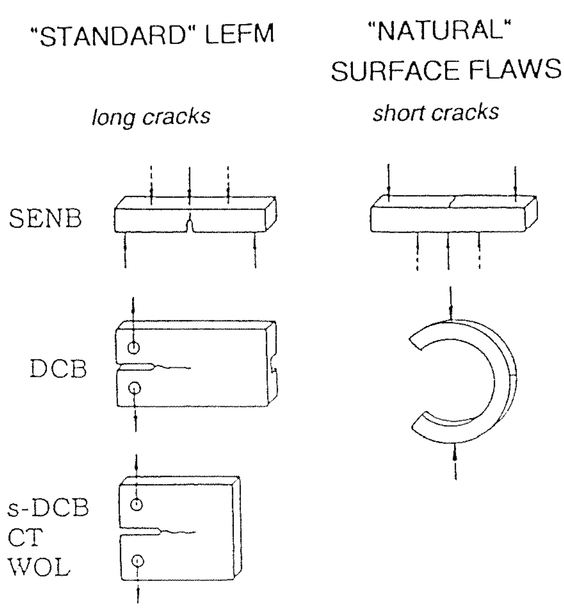

Fig. 1: Testing geometries for R-curve determination of ceramics

In the following sections R-curve results for long and short cracks of various Al_2O_3 ceramics and Mg-PSZ are presented and discussed.

3. "LONG CRACK" R-CURVE BEHAVIOR

3.1 Experimental R-curves

Crack resistance curves determined from stable crack propagation experiments with short double cantilever beam (s-DCB) geometry are shown in Fig. 2 for Al_2O_3 and Mg-PSZ [5]. In both cases material variants, optimized with respect to toughening, were used, i.e. coarse grained Al_2O_3 with intergranular fracture [8] and heat treated Mg-PSZ with high thermal shock resistance [9]. The transformation toughened Mg-PSZ exhibits a higher toughness level and also R-curve behavior is more pronounced. The plateau toughness value in the order of 15 MPa $\sqrt{m}$ is among the highest determined to date for monolithic ceramics [3, 5]. But also an impressive increase in toughness from 2 MPa $\sqrt{m}$ up to about 6 MPa $\sqrt{m}$ is observed in the Al_2O_3. In both ceramics extended crack growth occurs ($\Delta a \geq 1$ mm) before the high plateau toughness is finally reached.

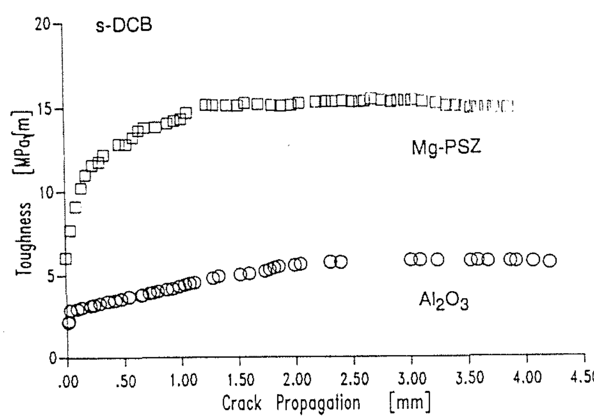

Fig. 2: "Long crack" R-curves of coarse grained Al_2O_3 ($D \approx 20$ µm) and MgO partially stabilized ZrO_2

However, it has to be exphasized that the rise in the R-curves in Fig. 2 and the plateau values are not unique properties for the given ceramics. R-curve behavior also depends on the microstructure and on the chemical composition of a ceramic and furthermore the crack history prior to the crack extension experiment is of importance [8].
To illustrate some of the intrinsic and extrinsic parameters which influence R-curve behavior, load-displacement curves are compiled in Fig. 3 from crack propagation studies in Al_2O_3. The curves, which reflect the amount of energy dissipated during crack extension, show the influence of mode of microstructure, grain size, loading rate, environment and temperature, respectively.

From the area under the load-displacement curves, which is assumed to be proportional to toughness, it can be deduced:

- intergranular fracture dissipates more energy than transgranular
- coarse grained material requires higher work of fracture than fine grained
- fast loading rates require more energy
- work of fracture decreases in the sequence oil, air, water
- work of fracture decreases with temperature, but increases in case of Al_2O_3 with glassy grain boundary phase.

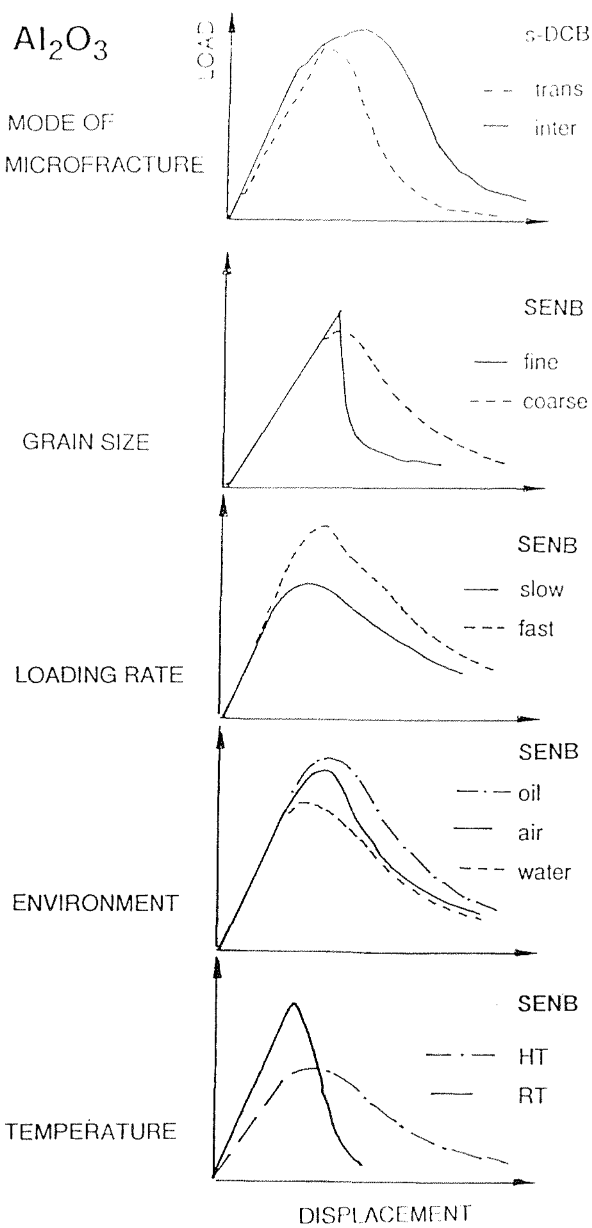

Fig. 3: Parameters which influence the toughening effect in Al_2O_3

3.2 Toughening mechanisms

The microstructural mechanisms which cause the macroscopic R-curve behavior of Al_2O_3 and Mg-PSZ have crack tip shielding in common [3, 5]. In both materials an increasing screening of the crack tip from the applied stress occurs as the crack grows. However, in Al_2O_3 the effect originates from contact bridging of the crack surfaces whereas Mg-PSZ residual stresses in a transformation zone are effective.

The micro-mechanics modelling of screening effects in the context of the macroscopic toughness is generally developed on the basis of energy concepts [3]. An energy term which arises from the screening effect is then added to the local energy requirement for the occurrence of fracture at the crack tip (G_0).

$$K_R = [E(G_0 + \Delta G_c)]^{1/2} \qquad (3)$$

The toughness and energy requirement have been linked in eqn (3) in the usual manner by way of the modulus of elasticity (E).

3.2.1 Contact shielding in Al_2O_3

Al_2O_3 is the ceramic material which has been most studied in connection with R-curve behavior arising from contact screening [8, 10-16]. Using renotch experiments (removal of the crack faces following a given crack growth) attention has been given to the toughening mechanisms which arise behind the crack tip (wake-effects) [10]. Such crack face interactions are recognized for instance from the change in crack path development which occurs in large grained alumina ceramics at increased crack opening (Fig. 4). The freeing of local keying points by the forming of side cracks can be seen in the two light micrographs which show the same specimen-location after two stages of crack growth. The crack face contacts exerting closing forces are not limited to the sample surface but occur also in the bulk of the material. Fig. 5 indicates qualitatively this effect in a two-step crack extension experiment with large-grained alumina (D = 20 μm). In crack extension experiments using s-DCB specimens, stable cracks of a given length can be enlarged (Fig. 5 (a)). From the load displacement behavior - with the characteristic departure from the linear elastic rise before reaching the maximum and with the following gradual reduction in required load, the alumina ceramic demonstrates R-curve behavior [7]. In the second part of the experiment, a tensile load is applied to the sample after a counter-notch has been made up to the crack tip (Fig. 5 (b)). It is then clear that the material which has already been broken can only

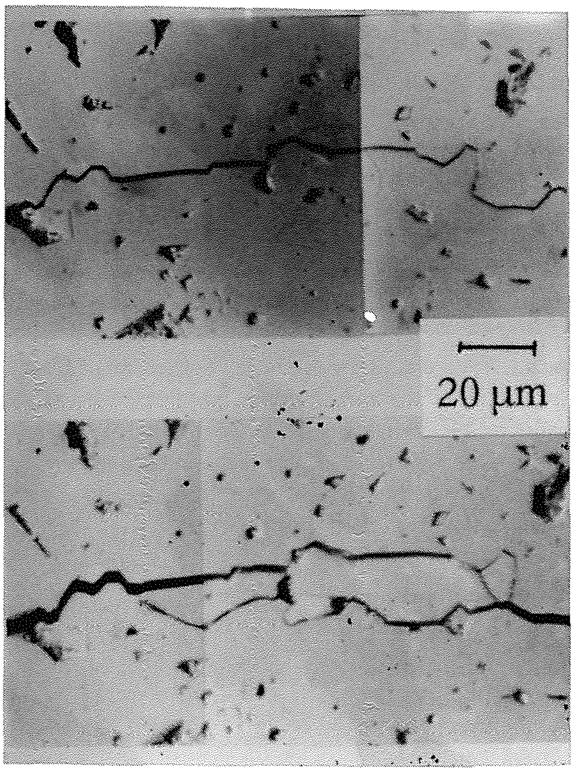

Fig. 4: Side-cracking in the wake regime of Al_2O_3 due to crack bridging forces

be fully separated with further energy dissipation owing the interlocking material at the crack faces. Systematic measurements with a similar method made by White et al. confirm the existence of this toughening mechanism for a variety of oxide materials [17].

Micromechanic modelling of the interlocking effect for the friction controlled separation of the bridging elements (Fig. 6) leads to [18]

$$\sigma(u) = \sigma_m \left(1 - \frac{u}{u_{max}}\right)^n \qquad (4)$$

where the stress falls from a maximum value $\sigma = \sigma_m$ directy behind the crack tip (crack opening $u = 0$) to the value $\sigma = 0$ at the end of the crack face interaction zone (where $u = u_{max}$) at a rate dependent upon the parameter n.

The additional toughening effect then is given by

$$\Delta G_c \approx 2\, f_B \int_0^{u_{max}} \sigma(u)\, du \qquad (5)$$

where the maximum separation distance, u_{max}, is determined by the point at which the crack closing force goes to zero in the stress versus crack opening distance behavior. The density of active crack bridges is taken care of in the parameter f_B.

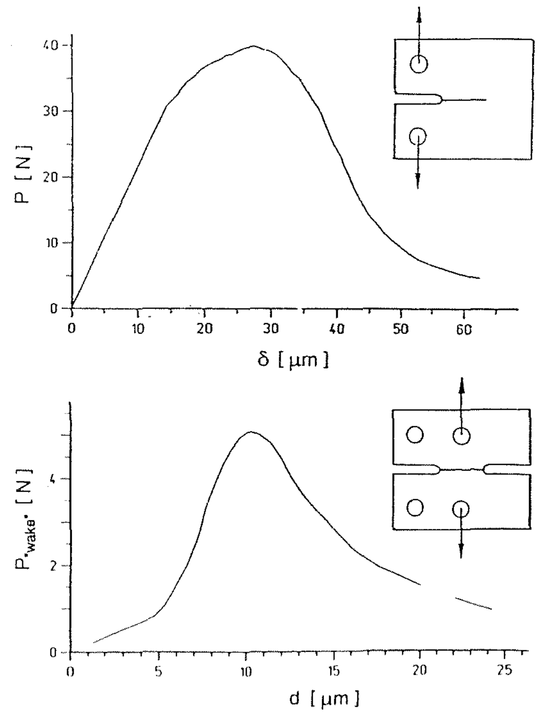

Fig. 5: Post-s-DCB tensile fracture test to demonstrate bridging forces between crack surfaces

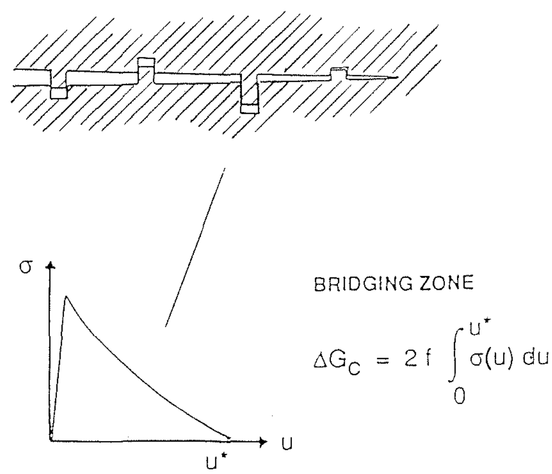

Fig. 6: Interlocking effect of crack surfaces with friction controlled separation of bridging elements

For the modelling of toughness from long cracks in test geometries of limited specimen size, eqn (5) changes to [8]

$$\Delta G_c = 2 \int_0^{z_{max}} \sigma u(z) \frac{du}{da}\, dz \qquad (6)$$

where z is taken parallel to the crack.

Equation (6) contains a geometry dependence of the toughening effect which can be derived on the basis of the methods of weight functions [19] and which has also been found experimentally from measurements with different crack and test geometries [8]. The grain size dependence of the R-curve behavior of Al_2O_3 is likewise explained by the toughening model of crack face bridges when reference is made to the respective maximum separation length $u_{max} \approx ¼ D$ [8].

Following the above micromechanical approach it seems resonable to consider the stress-separation function $\sigma(u)$ rather than the macroscopic R-curve as the important material property.

3.2.2 Transformation zone shielding in Mg-PSZ

The effectiveness of transformation toughening in zirconia-containing ceramics is shown clearly by the example of Mg-PSZ in Fig. 3. Even more striking R-curve behavior with plateau toughnesses $K_R \geq 18$ MPa $\sqrt{m}$ can be achieved by further optimization of the stress-induced microstructural transformations that are found in certain materials groups [20, 21].

In Mg-PSZ, metastable tetragonal (t) precipitates form the strengthening elements in a crack-surrounding process zone. In the stress field of the crack, there occurs a volume dilatation and a shearing as the elements transform to the equilibrium monoclinic (m) symmetry. The size of the transformation zone is dependent on the transformation propensity of the t-precipitates. Furthermore the toughening effect is determined by the number density of the transforming precipitates. According to Evans [3] the contribution of zone shielding to the required fracture energy is given by

$$\Delta G_c \approx 2 f_z \ \sigma_o \cdot \varepsilon_o \cdot h \qquad (7)$$

where f_z is the density of transformable t-ZrO_2 particles, h is the size of the zone, and the product $\sigma_o \cdot \varepsilon_c$ defines the mechanical energy dissipated in the non-linear $t \rightarrow m$ transformation of a precipitate (Fig. 7).

In high toughness Mg-PSZ, transformation zones of up to $h \approx 1$ mm can be formed [22]. As a consequence of the volume dilatation during the tetragonal to monoclinic transformation ($\Delta V = 4.7$ % in the absence of constraints [23]), the zone can be optically observed in the form of surface uplifting [21]. Although there is currently no exact correlation between microstructure, zone size and R-curve behavior [3], it has long been recognized that selected heat treatments can be used to influence the transformation propensity of the tetragonal precipitates and hence the transformation toughening of the material [9].

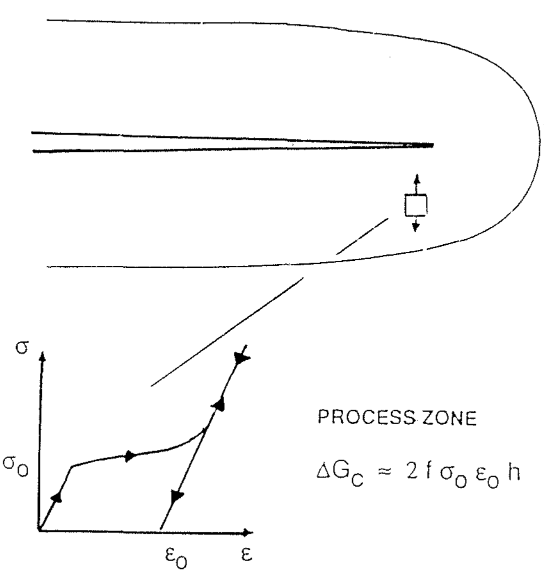

Fig. 7: Process zone shielding through non-linear stress induced transformation

However, if the optimization efforts are stretched too far, i.e. the tetragonal precipitates are tailored too metastable, aging can occur even at room temperature [21]. Fig. 8 shows R-curves of a Mg-PSZ material which in the "as received" condition only exhibits a "moderate" plateau toughness ($K_R \approx 9$ MPa $\sqrt{m}$). After short annealing at 1000°C for 20 min, which causes back-transformation of the m-precipitates into t-symmetry, the "reconditioned" material shows a maximum toughness of $K_R \approx 18$ MPa $\sqrt{m}$, i.e. an increase by 100 %.

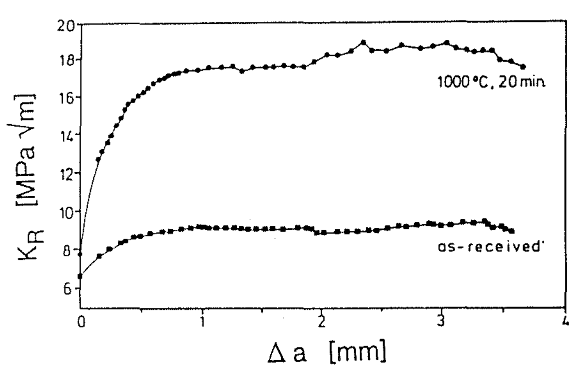

Fig. 8: Annealing of Mg-PSZ which aged at room temperature. Monoclinic precipitates are back-transformed to tetragonal symmetry to increase the density of transformation toughening elements.

4. "SHORT CRACK" R-CURVE BEHAVIOR

The long-crack R-curves described in the previous sections are useful to gain insight into the toughening behavior of alumina and zirconia. Furthermore the ultimate toughening potential after extended crack growth is recognized. The high toughness values cited for modern structural ceramics often correspond to the plateau values of long-crack R-curves.

However, such toughnesses are not really relevant for most structural applications, because experience tells that the fracture of ceramic components is not determined by cracks of some millimeters in length. Typically crack instability already occurs at much shorter crack sizes. Thus failure-relevant crack growth studies specifically must focus on the propagation and toughness behavior of short cracks and "natural" flaws.

The R-curve behavior of short cracks, frequently introduced by indentation techniques, has been studied in the case of Al_2O_3 intensively by Lawn and coworkers, e.g. [14]. The results are in qualitative agreement with the long crack observations, i.e. the grain size dependence and the crack bridging effects also hold for short cracks.
To further characterize and analyze R-curve behavior beyond artificially introduced cracks, controlled crack propagation experiments with "natural" flaws are necessary (Fig. 1). Although R-curve behavior automatically satisfies the condition of limited stable crack growth [7] experimental problems emanate if the effect is not big enough to be recognized in a mechanical test. Only few of such measurements have been reported to date [24-27], some of them also utilizing microscopic in situ observation of crack growth [24, 26]. Note that "natural" crack was put in quotation marks throughout this paper because microscopic crack observation requires well polished specimen surfaces.

Again coarse grained Al_2O_3 and high toughness Mg-PSZ had been the materials of choice. Fig. 9 shows in both materials such "natural" surface cracks which were generated on the tensile surface of bend bars by simple load increase. Interestingly the mechanical response of Al_2O_3 and Mg-PSZ upon loading significantly differs. Al_2O_3 behaves linear elastic in the bending test, whereas relaxation effects occur prior to and simultaneously with crack growth in Mg-PSZ due to far-field transformation of the highly metastable tetragonal precipitates [26]. Only after equilibrium between applied load and crack length is obtained the toughness can be determined using eqn. (2).

The resulting "natural" crack R-curves of Al_2O_3 are plotted in Fig. 10 together with the respective curves for indentation crack growth and long crack propagation in SENB geometry. The short crack R-curve starts at a considerably lower stress intensity level and crack instability occurs before the plateau toughness of the long cracks is reached. The low crack initiation stress intensity of the "natural" flaws (≈ 1 MPa $\sqrt{m}$) is presumably related to localized microstructural heterogeneities. They either may weaken the material or induce residual stress fields [28]. Often more than one crack forms. They can all grow but the one with the shallowest R-curve becomes unstable first.

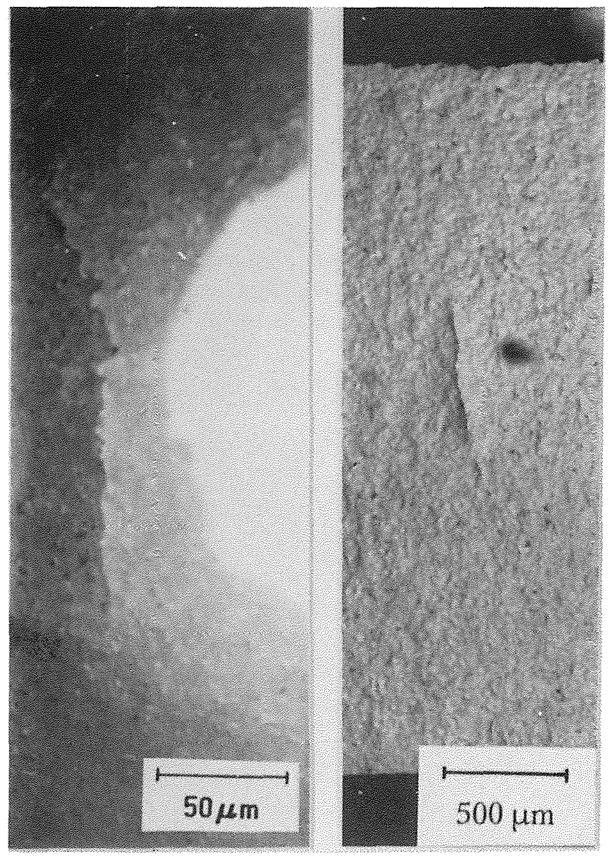

Fig. 9: "Natural" surface cracks in Al_2O_3 (left) and Mg-PSZ (right)

Similar R-curve results are obtained with short cracks in Mg-PSZ (Fig. 11). Again crack initiation is observed at much lower stress intensities compared to those where long cracks start to grow, and again the high plateau value, e.g. $K_R = 15$ MPa $\sqrt{m}$, is not reached with "natural" surface cracks.

The differences between "natural" and long cracks are important from the application point of view. Under technically relevant loading conditions specimens or components obviously cannot utilize the full potential of the R-curve behavior of ceramics. Clearly toughness or resistance data referring to the plateau value from long crack measurements overestimate the values pertinent for service requirements.

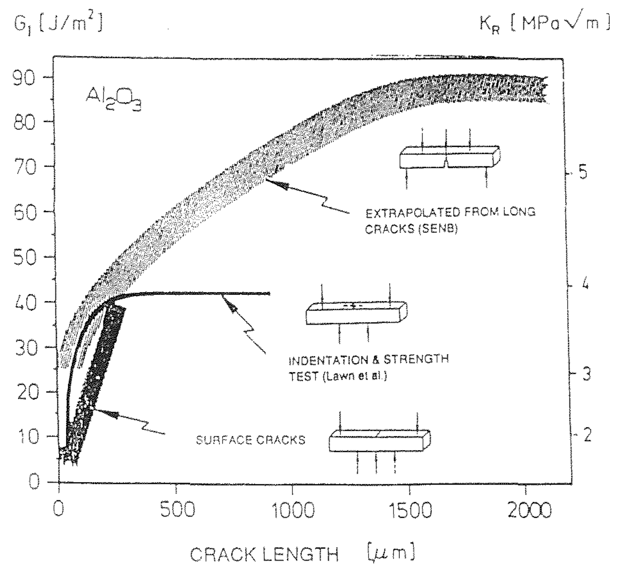

Fig. 10: R-curve behavior of Al_2O_3. Comparison of short and long crack experiments. Also indentation strength in bending results derived from [14] are included.

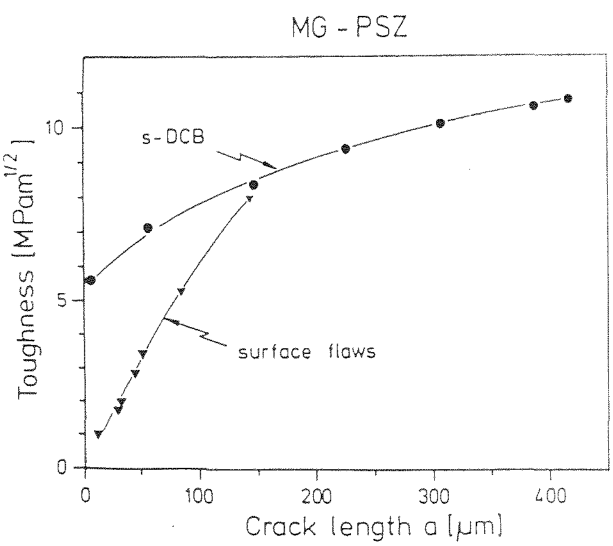

Fig. 11: Comparison of R-curve behavior of Mg-PSZ. Long crack (s-DCB) and surface crack (bending) experiments

5. CONCLUSIONS

The R-curve behavior of two oxide ceramics was described which originates from an increasing crack tip shielding during crack extension. Besides material properties, crack and testing geometry influence the shape of the R-curve.

In the case of Al_2O_3, which may also serve as a model material for non-transforming monolithic ceramics, the R-curve behavior is wake-controlled, i.e. interlocking grains form bridging elements which exhibit sliding friction during separation. The stress-separation function rather than the macroscopic R-curve is seen as the important material property.

The R-curve behavior of Mg-PSZ is governed by process zone shielding, i.e. the transformation zone around the crack generates residual stresses which oppose the applied stress intensity.

Although long crack studies are necessary to establish the toughening potential of a given ceramic material and to deduce the microstructural mechanisms, information about the failure relevant toughness properties is only obtained by "natural" flaw experiments.

Long crack R-curve data overestimate for both oxide ceramics, Al_2O_3 and Mg-PSZ, the toughness increase actualy available before component failure.

6. ACKNOWLEDGEMENTS

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