MECHANISMS OF TOUGHENING IN CERAMICS

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INTRODUCTION

Considerable effort has been expended to produce structural ceramics with enhanced toughness with some success, although toughening mechanisms which remain effective at high working temperatures remain elusive. Parallel efforts are underway for concrete and polymeric materials. A micro-mechanics of toughening of brittle solids is beginning to emerge. For ceramics, the mechanisms include transformation toughening, toughening by a metallic particulate phase, and fiber reinforcement. Brittle polycrystalline or multi-phase solids also have "inherent" toughening mechanisms connected with their heterogeneity which are not well understood. An overview of some of these mechanisms is given in this talk with the aim of highlighting the approaches and issues from a mechanics perspective. Transformation toughening and toughening by a metallic phase will be discussed briefly first. The bulk of the talk is concerned with the status of inherent toughening mechanisms in single phase polycrystalline ceramics.

2. TRANSFORMATION TOUGHENING

A theory of transformation toughening began to emerge just over five years ago after it had been discovered experimentally that a second phase constituent which undergoes a stress-induced martensitic phase transformation could be used to toughen structural ceramics. In the intervening years theory and experiment have reinforced one another, and there now exists a reasonable understanding of many aspects of the phenomenon [1]. Materials have been produced recently with toughnesses higher than ever before recorded for ceramics. One of the first clear successes of the mechanics modeling was the prediction of crack growth resistance for transformation toughened materials (i.e. R-curve behavior in fracture terminology). Stable crack growth was then observed and within the last few years R-curves have been measured for a number of zirconia toughened materials.

The mechanics approach involves the formulation of a phenomenological continuum model of the stress-strain behavior of the two-phase material and then the use of this constitutive model to determine the effect of transformation on behavior at a macroscopic crack tip. The approach parallels that taken to understand the role of plasticity in the fracture of metals. The two most important ingredients to the constitutive theory are the multi-axial stress condition for nucleation of transformation and the characterization of the transformation strain at the continuum level. For zirconia particles transforming from the tetragonal to the monoclinic phase, a 4% volume expansion occurs. A shear transformation accompanies the dilatation but the amount which occurs at the continuum level is uncertain because a particle usually

transforms into multiple layers with alternating shearing. The simplest constitutive models are based on a critical mean stress condition for transformation and a purely dilatational transformation strain, although these are undoubtedly too simple for a fully quantitative theory.

The role of transformation in toughening is examined within the context of the continuum theory by the extent to which the intensity of the near-tip stress field is altered, or shielded, by the zone of transformation induced at the tip. According to calculations based on the simplest constitutive model, the transformation zone has no net effect on the near-tip intensity for a stationary crack. Only when the crack advances leaving behind a wake of transformed material does significant crack tip shielding take place. Toughening is inherently tied to stable crack growth resistance.

3. CERMETS ~ TOUGHENING BY METAL PARTICLES

Tungsten carbide/cobalt used for some time as a material for cutting tools exploits its metal phase to toughen an otherwise brittle matrix material. Any macroscopic crack which occurs in the material leaves behind metal particles, or ligaments, which bridge the crack surface just behind the crack tip restraining its opening. For this mechanism to be effective it is essential that the particles be well-bonded to the matrix and that the advancing crack tip be drawn to particles so that bridges are left behind. Basic mechanics models [2] relate the near-tip stress intensity to the remote, or applied stress intensity, as altered by the bridging particles. Imposition of the condition that the near-tip stress intensity be at the level needed to crack the matrix (times a factor proportional to the area fraction of the matrix) leads to the toughening enhancement due to bridging. This enhancement depends on the ratio of the work of fracture of the particles to the work of fracture of the unreinforced matrix. The modeling of the toughening enhancement stands at a somewhat perplexing state. To bring the models in line with experimental data for several systems (Al_2O_3/Al and WC/Co) requires that the bridging stress developed in the particles be more than twenty times their uniaxial flow stress. Constraint by the surrounding matrix does raise the bridging stress to levels as large as 6 or 7 times the uniaxial flow stress, as model experiments have revealed [3]. But a level of twenty times the flow stress seems unrealistically large.

A tentative modification of the bridging model which might bring it into line with experiments involves the idea that the stress intensity at the tip may have to be well above the level needed just to crack the matrix if the crack front is trapped by the particles. The trapping of a bowed dislocation line by arrays of obstacles is an analogous phenomenon. Preliminary calculations [4] suggest that stress intensity levels between two and three times the critical matrix toughness level may be required to advance the crack front through a line of particles with an area fraction around 20%. Thus the critical crack-tip toughness for a bridged crack should (perhaps) be the critical intensity required to advance the trapped crack front rather than that of the pure matrix. Clearly, there are important aspects of the phenomenon that remain obscure.

4. TOUGHNESS OF SINGLE PHASE POLYCRYSTALLINE CERAMICS

It can fairly be said that the mechanics of the toughness of single phase polycrystalline ceramics is not even qualitatively understood. Consider alumina, Al_2O_3 , at room temperature as an example. The toughness for intrinsic grain boundary cracking (measured in terms of a critical energy release rate) is generally agreed to be about $2Jm^{-2}$; the toughness associated with the most cleavable planes of a single crystal is roughly $8Jm^{-2}$; and toughness measured in the

macroscopic cracking of high quality polycrystalline alumina usually lies between 20 and 50 Jm⁻², depending on the grain size. The mystery is why the polycrystalline toughness is so high given the observational fact that the macroscopic crack does advance by predominantly grain boundary cracking. Obviously, the notion of a "path of least resistance" is not at play here. Various inherent mechanisms of toughening have been suggested to explain the source of the polycrystalline toughness [5] and these will be reviewed here from the mechanics perspective. It is our opinion that the mechanism (or mechanisms) has not been identified with any convincing certainty, and this is borne out by the divergence of views of workers in the field. A quantitative understanding of the inherent mechanisms of polycrystalline toughness of brittle solids is likely to impact our understanding of toughening mechanisms in general.

Central to each of the mechanisms which have been considered is the heterogeneity at the micro-scale of the polycrystal. Grains and grain boundaries have random orientation. Most single crystals are elastically anisotropic resulting in stress concentrations (singularities) at grain boundary junctions and vertices. Non-cubic crystals have a certain degree of anisotropy in thermal expansion properties; and, because polycrystalline materials are usually formed at sintering temperatures, at room or operating temperatures there are usually substantial residual stresses varying from crystal to crystal. These residual stresses increase with increasing grain size, and above a critical grain size most non-cubic polycrystalline ceramics undergo spontaneous micro-cracking along grain boundaries upon cooling from the fabrication temperatures to room temperature. For Al_2O_3 , this critical grain size is on the order of 200 to 400 μ m. Below the critical grain size grain boundary cracks are stable until they finally coalesce with the macro-crack. The micro-cracks are thought to be nucleated at grain boundary junctions or vertices where local stress concentrations occur [6]. They typically run over one grain boundary facet until they arrest at another grain boundary junction [7].

Thus, a macroscopic crack making its way through a brittle polycrystalline ceramic must contend with having to deflect to accommodate the varying orientations of the grain boundaries, with residual stresses varying from grain to grain, with the possibility of a zone of stress induced micro-cracks at its tip, and with any uncracked ligaments left behind the tip bridging its opening.

4.1. Crack Deflection Toughening

Macroscopic cracks in many polycrystalline ceramics do advance along grain boundaries since the grain boundary toughness is substantially less than the lowest cleavage toughness of the crystals. The variation in the orientation of the grain boundary facets and in the residual stress force the crack to advance in a tortuous manner which is not readily analyzed. The simplest approach of adding the additional surface energy associated with the extra area of grain boundary surface generated by the tortuosity is not valid because part of the energy supplied to create the crack surface comes from the residual stress. Most workers in the field seem to accept the outcome of the highly approximate analysis given in [8] where it is concluded that deflection effects in materials with more or less equiaxial grains may account for about a factor of 2 increase in toughness (measured in energy units) above the grain boundary value. The modeling does not specifically account for residual stress effects, and it is conceivable that there is a contribution to toughening from crack front trapping by residual stress.

4.2. Micro-crack Toughening

The idea behind the mechanism of micro-crack toughening is that stable grain boundary micro-cracks are nucleated by the high stresses in the vicinity of the macroscopic crack tip. These micro-cracks then lower the stress experienced by the tip. This shielding effect has been

studied from two vantage points: one involving a smearing out of the effects of the micro-cracks appropriate to a zone of profuse micro-cracking and the other treating the interaction of the macro tip with discrete micro-cracks. No clear-cut conclusion from these theoretical studies has yet emerged, but an attempt will be made below to summarize the current status.

Direct experimental evidence of profuse micro-cracking surrounding a macro-crack is limited, partly because of the difficulty of making such observations. Detailed documentation of a micro-crack zone in a two phase alumina/zirconia material has recently been published [7], including information on the spatial distribution of the micro-cracks and on their orientation. In this particular material there appeared to be no preferred orientation of the micro-cracks relative to the stresses at the macro-crack tip, as might be expected given the large and essentially random variation of the residual stresses in the grains. A similar study of a relatively large grain, single phase alumina [9] revealed no micro-crack zone but did provide evidence of substantial crack bridging by uncracked grains, as will be discussed further in the next subsection. In any case, direct experimental evidence for the micro-crack toughening mechanism in single phase polycrystalline ceramics does not yet exist.

The theoretical approach to analyzing the shielding effect of a zone of profuse micro-cracks surrounding the macro-crack tip has close parallels to the approach taken to transformation toughening [10-12]. There are two separate contributions to shielding -- one arising from the reduced effective moduli in the zone and the other due to the partial release of residual stress which is formally equivalent to a transformation strain. When the micro-cracks are randomly orientated the reduced moduli remain isotropic and the equivalent transformation strain is a pure dilatation. Just as in the case of transformation toughening, the effect of the release of residual stress on shielding is proportional to the square root of the zone size and the shielding increases as the crack advances leaving micro-cracks in its wake. By contrast, the shielding contribution due to the reduced moduli is independent of the zone size, is relatively insensitive to the shape of the zone, and increases, but only slightly, with crack advance. Thus, any R-curve effect is mainly due to the release of residual stress. An attempt was made in [7] to make a detailed comparison between the theoretical micro-cracking models and experimental data for the alumina/zirconia material using observed distributions of micro-cracks and measured information on the release of residual stress. In this particular case, the two contributions to the toughness were roughly equal resulting in a total toughness increase of about a factor of four (measured in energy units). This level of increase appears to be representative; that is, significantly higher toughening should probably not be expected from profuse micro-cracking.

The second theoretical approach to micro-crack toughening employs two-dimensional, plane strain solutions for a macro-crack interacting with discrete micro-cracks in the vicinity of its tip, as represented by the studies in [13-17]. These investigations focus on how the stress intensity at the macro-crack tip is altered -- lowered or elevated -- by the configuration of micro-cracks. They have exclusively considered the weakening of the solid and have not included the contribution from the release of residual stress. A recent unpublished study in [17] gives the maximum mutual shielding possible in the interaction between a semi-infinite macro-crack loaded in mode I and a single micro-crack. That is, the configuration of the micro-crack is varied to minimize the energy release rate of the macro-crack subject to the constraint that the energy release rate at either tip of the micro-crack itself not exceed that of the macro-crack. The resulting minimum energy release rate of the macro-crack is reduced to 46% of the "applied" level and one of the tips of the micro-crack attains this same level. In other words, the maximum toughening which can be achieved by a single micro-crack is roughly a factor of 2 in energy terms, assuming no contribution from the release of residual stress is taken into account. For reasons now discussed, we believe this is likely to be representative of the maximum toughening increment expected from micro-crack shielding.

As already mentioned, shielding is independent of the size of the micro-crack zone. Moreover, calculations in [10] reveal that an annular zone of randomly orientated profusely distributed micro-cracks with a central core of uncracked material centered at the macro-crack tip results in very little change in the stress intensity at the macro-crack. Thus, one concludes that it is mainly the micro-cracks nearest to the macro-crack tip which determine the net shielding, or anti-shielding, of the macro-tip. This conclusion appears to be borne out by recent calculations for large numbers of discrete micro-cracks. Since the maximum mutual shielding from a single micro-crack is about .5, we expect this is more or less representative for any array. The mechanics of what determines a typical array has thus far not been addressed.

4.3. Macro-crack Bridging

Several mechanisms involving bridging of a macro-crack tip have been identified as playing an important role in toughening: metallic particles in brittle matrices discussed in Section 3 and ceramic fibers in ceramic matrices. Clear evidence of bridging by uncracked grains has been observed in large grain, single phase alumina [9]. This material also displayed significant R-curve behavior with the crack resistance still increasing after several millimeters of crack advance. The bridging grains were observed to survive as far back as one hundred grain diameters behind the tip.

The role the bridges play in reducing the stress intensity at the crack front (i.e. shielding the lead crack tip) is readily understood and analyzed [18]. The puzzling aspect of the phenomenon is how the bridging grains are able to survive far back from the lead tip since the stress intensity levels where the crack surface impinges on these grains are exceptionally high according to any relatively straightforward estimate. One plausible suggestion [19] invokes local residual stresses at the bridging grains for their "protection". Suppose, for example, that prior to any cracking a grain destined to be a bridge supports a residual tensile stress normal to the potential crack plane. Surrounding this grain on the same plane will be a compressive stress which is largest at the equator of the grain and falls off away from it. This compressive region will tend to protect the grain from any crack engulfing it on the plane in question. Statistically, only a relatively small fraction of the grains will be favorably orientated and positioned to be so protected, and thus the area fraction of surviving bridges is not expected to be large. A first quantitative model of the protective role of residual stress [17] shows some promise in rationalizing the survivability of the bridges but not for bridges as far back as those observed in [9]. At this writing, the issue remains a puzzle.

4.4. Concluding Remarks on Toughening Mechanisms in Polycrystalline Ceramics

We have tried to make the case that a micro-mechanics model of the inherent toughening of single phase polycrystalline ceramics does not yet appear to exist. Specifically, with alumina as an example, an accepted explanation does not exist for how the toughness of the polycrystalline material (in energy units) can be on the order of 10 to 20 times the intrinsic grain boundary toughness, even though the cracking process is largely grain boundary cracking. It may be that the several mechanisms discussed above act in concert adding up to give the inherent toughness of the polycrystalline material. It seems equally likely that there may be other mechanisms or other features to the mechanisms mentioned above which remain to be uncovered. Further background, insight, and access to the literature can be found in the recent review articles [1, 5, 20].

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REFERENCES

- [1] Evans, A. G. and Cannon, R. M., Acta metall. 34 (1986) 761.
- [2] Budiansky, B., Amazigo, J. C. and Evans, A. G., J. Mech. Phys. Solids, 36 (1988) 167.
- [3] Eberhardt, J. and Ashby, M. F., "Flow Characteristics of Highly Constrained Metal Wires", Report, Engineering Department, Cambridge University, Cambridge, England (December 1987).
- [4] Fares, N. and Rice, J. R., work in progress, Division of Applied Sciences, Harvard University.
- [5] Freiman, S. W., Ceramic Bulletin, **67** (1988) 392.
- [6] Tvergaard, V. and Hutchinson, J. W., J. Am. Ceram. Soc., 71 (1988) 157.
- [7] Rühle, M., Evans, A. G., McMeeking, R. M., Charalambides, P. G., and Hutchinson, J. W., Acta metall. 35 (1987) 2701.
- [8] Faber, K. T. and Evans, A. G., Acta metall. 31 (1983) 565.
- [9] Swanson, P. L., Fairbanks, C. J., Lawn, B. R., Mai, Y-W., and Hockey, B. J., J. Am. Ceram. Soc. 70 (1987) 279.
- [10] Hutchinson, J. W., Acta metall. 35 (1987) 1605.
- [11] Ortiz, M., J. Appl. Mech. 54 (1987) 54.
- [12] Charalambides, P. G. and McMeeking, R. M., Mechanics of Materials 6 (1987) 71.
- [13] Kachanov, M., Int. J. Solids Structures 23 (1987) 23.
- [14] Rubinstein, A. A., Int. J. Fracture 27 (1985) 113.
- [15] Horii, M. and Nemat-Nasser, S., J. Mech. Phys. Solids 35 (1987) 601.
- [16] Rose, F., Int. J. Fracture 31 (1986) 233.
- [17] Shum, D. and Hutchinson, J. W., work in progress, Division of Applied Sciences, Harvard University.
- [18] Mai, Y.-W. and Lawn, B. R., J. Am. Ceram. Soc. 70 (1987) 289.
- [19] Swain, M. V., J. Mater. Sci. Lett. 5 (1986) 1313.
- [20] Clarke, D. R. and Faber, K. T., J. Phys. Chem. Solids 48 (1987) 1115.