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A qualitative model for the development of tough ceramics

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A qualitative working model is presented in this paper for the development of tough ceramics. The model shows that toughness can be imparted only to dispersed multiphase ceramics in preference to single phase ceramics. A simultaneous improvement in strength can be achieved by securing a strong bond between the dispersed phase and the matrix. Eutectic composites of mixed oxides merit serious consideration as potential tough ceramics.

1. Introduction

Ceramics have been traditionally referred to as a class of materials which fail in a brittle manner when subjected to a mechanical or thermal stress. In practice, it means that once a crack is initiated in ceramics, there is no resistance to propagation and consequently, the ceramics fail catastrophically. The potential for such catastrophic failure has limited the use of ceramics in many applications. At the same time, there is a growing need to use ceramics in designs where increasingly demanding mechanical and thermal stresses combined with high temperature corrosion, severely restrict the use of metals. In the metal vapour arc lamp, in MHD power conversion, in high temperature turbine blades, for example, certain structural parts are subjected to such a combined load of mechanical and thermal stresses that ceramics merit serious consideration as structural components. In order to meet this growing demand, the objective to date has been to make stronger and stronger ceramics, so that a higher fracture-initiation stress exists at failure. But here again, once the fracture is initiated, the higher kinetic energy from stronger ceramics produces a fracture which is, nevertheless, catastrophic and even more spectacular. Failure occurs without a warning.

On the contrary, if the propagation of a crack, once past the stage of initiation, can be truly arrested, structurally sound components can be fabricated with ceramics. This is the key to producing tough ceramic. Indeed, there has been mounting evidences [1-4] in recent years that not all ceramics necessarily fail in a brittle manner when subjected to a load and do exhibit some degree of toughness. This paper is an attempt to understand the toughness in ceramics in terms of crack propagation and to delineate a general approach of fabricating the same. Details are expected to vary with a particular system.

It is desirable to stress at this point that there is no universally agreed upon definition of toughness in ceramics. It must, however, represent some property of the material which will give a measure of resistance to crack propagation when subjected to a mechanical or thermal stress. Various toughness parameters in the literature [5] relate the energy required to fracture a specimen (W) to the area of the material fractured (A), so that the toughness parameter is proportional to W/A. Since considerable confusion exists [6] regarding the definition and measurement of W, the toughness parameter is expressed in the following in terms of critical strain energy release rate (or critical energy dissipation rate). This parameter, designated as G_c, is shown to be the truly fundamental toughness characteristic of the ceramics.

2. Crack propagation: single versus multiphase ceramics

The working model for a tough ceramic as presented in this paper is based on the concept of stable crack propagation as developed by Glucklich and co-workers for cement composites [3, 7, 8]. The essence of this concept is that when crack arresters are present in a brittle matrix, the energy demand curve for crack propagation is nonlinear in contrast to a linear demand curve for the Griffith material. However, before we go into the details, it is desirable to establish some fundamental characteristics of crack propagation in terms of brittle fracture.

According to Griffith, the energy balance for fracture in an ideal homogeneous brittle material can be represented by the curves shown in Fig. 1. This is an example of an elliptical flaw of the semi-major axis C, perpendicular to a uniform tension field σ. The energy demand curve is a straight line represented by the equation: $W_d = 4C\gamma$, where γ is the surface energy. The energy release curve is a second power parabola: $W_r = \pi C^2\sigma^2/E$, where E is the Young's modulus. Crack instability occurs when the slopes of the two curves are equal:

$$\frac{\partial W_r}{\partial C} = \frac{\partial W_d}{\partial C}\,. \tag{1}$$

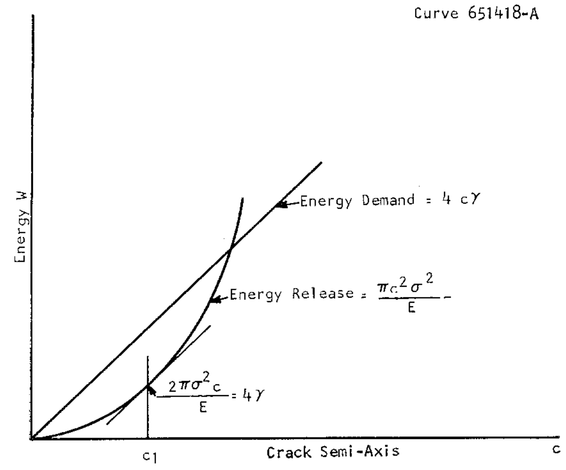

Figure 1 Crack propagation in ideal Griffith material.

Implicit in the result of Equation 1 is the fact that the instability (propagation) is assured in advance, once the crack is initiated. The instability is thus "built in" in Griffith's criteria of brittle fracture. In single phase homogeneous brittle material, since the energy demand is the same everywhere, the initiation and the propagation of a crack therefore coincide, and the fracture is instantaneous as in glass. The development of tough ceramic, where the objective is to promote a stage of stable crack propagation, is thus denied by single phase homogeneous brittle materials, irrespective of strength. The situation is, however, different with multiphase materials.

In multiphase ceramics, there is a distinct division in energy demand between various phases, so that the stages of initiation and propagation are separate. A crack, for example, may develop in a weaker matrix, and while propagating, may encounter a zone of tougher second phase. To clear the obstacle, the crack may cut through or detour around this zone; in either way, the energy demand will suddenly increase, and the propagation will be halted unless there is an increment in operating stress. Thus, the presence of selective inhomogeneities in a ceramic material will prevent the premature growth of an unstable crack.

Following the above argument, Glucklich [7, 8] has proposed that the energy demand curve for a heterogeneous material is nonlinear in contrast to a linear demand curve for the homogeneous material. The nonlinearity arises from the ability of the secondary phase to stabilize a growing crack, by requiring more energy to clear the obstacle. The demand curve is, therefore, concave upward [7]; the energy demand per unit crack length is thus an increasing fuction of crack length C:

$$\frac{\partial^2 W_d}{\partial C^2} > 0\,. \tag{2}$$

The condition as predicted by Equation 2 demands that $\partial W_d/\partial C$ must constantly be increased as the crack propagates. This requires that a similar increase be made in $\partial W_r/\partial C$ by raising the applied stress σ. The condition for stable crack propagation:

$$\frac{\partial W_r}{\partial C} \leqslant \frac{\partial W_d}{\partial C} \tag{3}$$

is thus always satisfied when the demand curve is nonlinear.

Following Glucklich, the energy balance for crack propagation in a multiphase ceramic is represented by the curves shown in Fig. 2. For simplicity, the demand curve, which is concave upward, is illustrated as a smooth curve. An initial crack of length C_0, will begin to grow under a stress σ_0, but it will be soon stopped at C_1 because of the increase in energy demand at this point. For the crack to continue, the stress has to be raised to σ_1. With this new operating stress level, the crack will grow to C_2 until a further increase in stress level is permitted. This phenomena will continue to a crack length of C_5 with a corresponding increase in stress of σ_5. The crack will thus propagate in steps, alternately from instability to stability, and the final strength will exceed that of crack initiation. At point A, the slopes of the energy demand and the energy release curves are equal and the crack will now grow spontaneously. The above, of course, is a highly idealized case of a tailored ceramic where the crack, as it grows, encounters an increasingly tougher medium. The crack propagation in a real multiphase ceramic may differ considerably in detail, but the overall growth mechanism will be similar to the mechanism proposed above.

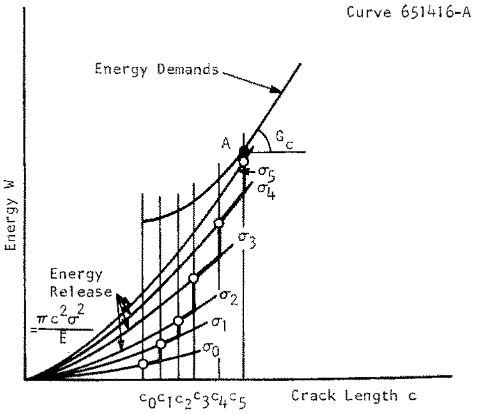

Figure 2 Forced crack propagation in a multiphase material.

3. Toughness versus strength in ceramics

In contrast to a homogeneous material (ideal Griffith case), the strength-determining property of multiphase ceramics is thus seen to be somewhat complex. In an ideal material, the surface energy, γ, alone determines the resistance to crack propagation or toughness of the material. In a real material such as a ceramic, the equivalent to γ would be the rate of energy absorption (or dissipation). This value is significant only at the onset of instability (point A in Fig. 2) and is then designated as G_c, the critical strain energy release rate (or the critical energy dissipation rate). G_c is, therefore, the maximum slope attained by the energy demand curve. In a real material, it can be regarded as the sum of all individual slopes caused by various crack arresting factors. With metals, G_c is a characteristic constant of the material. With ceramics, to a first approximation, G_c may also be regarded as a material constant, and its magnitude may also be affected by factors such as kind and size of the various phases, their volumes, density, and microstructure, etc., as in metals. For example, when the dispersed phase is very small in size, the crack may not be aware of its presence and there will be no improvement in G_c. Increasing the size of the dispersed phase will make the crack depend on the obstacle present in the matrix; during propagation, the crack must then either penetrate or detour around the second phase and consequently, there will be an increment in G_c.

Increasing the volume of the second phase will similarly affect the value of G_c. However, when both the volume and the size are significantly increased, the second phase particles become too close to each other, macroscopic defects (e.g. void) generally appear in the composite and there is a lowering of G_c (there is no energy demand for propagation of a crack through a void). In essence, G_c will go through a maximum with increase in size and volume of the dispersed phase. The literature data of fracture energy (which is proportional to G_c) of alumina-glass [9], hydrated alumina-epoxy [10], and glass-epoxy [13] composites are in agreement with the above observation.

It is thus apparent from the discussion that the addition of a second phase to the matrix influences the resistance to crack propagation or toughness (G_c) of the ceramic. It is also clear from the discussion that when the secondary phase has a G_c value higher than that of the matrix, toughness is considerably increased. By the same token, the toughness is decreased when the G_c value of the secondary phase is lower than that of the matrix. The example of the former is the alumina dispersed glass composite [9] as mentioned above, and that of the latter is the silicon carbide-dispersed-silicon nitride composite [11]. It is known that silicon carbide [12] has a fracture energy typically lower than that of silicon nitride [11]. The result is a reduction in composite fracture energy as demonstrated by the experiment. Only when the particle size of silicon carbide is considerably larger than the matrix grain size, does the composite show an increase in fracture energy. The hypothesis advanced for this latter behaviour was that the crack front could only interact with particles that were larger than the grain size of the matrix [11]. On the contrary, it is likely that the increase in fracture energy is caused by poor bond strength between the dispersed phase and the matrix as discussed below.

It is shown in the foregoing that when a dispersed phase is carefully chosen, the energy demand for crack propagation is increased and a toughness is imparted to ceramics. Furthermore, since the fracture occurs at a stress higher than the fracture-initiation stress, it follows that there will be an increment in final strength according to the model presented above. Barring few instances, the experimental data are in disagreement with this prediction. High values of fracture energies have been generally accompanied with low values of strength. Analysis of the literature data indicates that this discrepancy results from the existence of an imperfect bonding between the dispersed phase and the matrix. If the area adjacent to the dispersed phase is weak due to poor bonding, the crack will preferentially grow in this region because the energy demand is least in the weak interface. The fracture energy will increase but the strength will suffer. Experimental support for this observation can be found in works concerning the epoxy-glass system [13]. Here, the degree of interfacial bonding was controlled by pretreating the glass spheres, prior to fabrication, with organic agents which imparted different degrees of interfacial bonding. The highest fracture energy was obtained with the weakest interfacial bonding. At the same time, there was a substantial decrease in strength relative to that attained in composites made from untreated spheres. The same argument can be applied to the SiC–Si_3N_4 system [11], where an increase in fracture energy was observed when the SiC particle size (32 μm) was much larger than the Si_3N_4 matrix grain size (5 μm). It is obvious that the interfacial area is larger in this case than when the size of the dispersed phase was small and is weaker than the matrix phase due to differential thermal contraction of each phase ($\alpha_{Si_3N_4} = 3.6 \times 10^{-6}$ °C^{-1} and $\alpha_{SiC} = 5 \times 10^{-6}$ °C^{-1}) and high fabrication temperature (1750° C). The area adjacent to the dispersed phase will, therefore, be the most preferred path of crack growth. Once this situation exists, fracture energy will increase but strength will decrease, as found in the experiment. If, however, the interfacial bonding is strong, the crack will deviate from the area adjacent to the dispersed phase. As a result, both the strength and fracture energy will increase. In order to improve the strength of the composite, the bonding strength between the dispersed phase and matrix should be strong, preferably, as good as that of the matrix. Imperfect bonding results from a number of factors such as the size and volume of the dispersed phase, elastic moduli of the phases, residual and localized stresses and so on. These factors either independently or in combination produce flaws at the interface which then act as precursors to Griffith failure. The high fracture energy but low strength of alumina-glass composite clearly results from an imperfect bonding between the glass and the alumina. This explanation is true for many other composites where two or more components are incorporated by standard ceramic techniques such as hot pressing.

Finally, it is instructive to evaluate the present status of engineering ceramics with regard to desired level of toughness. Recently, Davidge [14] has addressed this question by comparing the measured values of fracture energies with those calculated from elastic energy consideration at the level of fracture stress of the ceramics [6]. The latter values represent the works of fracture necessary to prevent the complete failure. It is shown from the calculation that the fracture energies of the existing engineering ceramics such as the oxides of aluminium and magnesium with a typical value of ~ 20 to 50 J m^{-2}, are more than an order of magnitude lower than those required to prevent catastrophic failure. On the contrary, composites with fracture energy values greater than 1000 J m^{-2} merit serious consideration as tough ceramics. These conclusions are in agreement with the predictions made in this paper. It is thus apparent that unless means are developed to improve the fracture energies of single phase ceramics, multiphase composites offer the best hope of obtaining ceramic materials that are both strong and tough. The validity of this approach has been recently demonstrated by Davidge [14] in certain ceramic composites containing carbon fibre with a matrix of glass or cement. He showed that there was not only a several order of magnitude increase in fracture energies, but also a marked improvement in strength. This suggests that a strong interfacial bonding was achieved between the fibre and the matrix during hot pressing. However, the implication that a strong bonding was due to interfacial chemical reaction between the fibre and the matrix appears detrimental to the long term stability of the fibre at elevated temperature. For less demanding thermal environment, ceramic composites such as those reported by Davidge seem satisfactory. The potential for an ideal combination of a high strength and high fracture energy is believed to exist in "self-generated" composites. They are derived from eutectic melts which solidify to yield a peculiar type of microstructure. Often they consist of rods or plates of one phase in a matrix of another. It is possible that the orientation of these phases can be preferentially controlled to achieve the perfect bonding with the matrix. It will require a major technological advance to fabricate oxide eutectics in sufficiently large dimension to be of use as structural components, but since single crystal sapphire of large dimensions has been fabricated from the melt, it is not considered impractical.

4. Summary

It is shown that the toughness in ceramics can be best described in terms of critical strain energy release rate, G_c, (or critical energy dissipation rate). It is further shown that the addition of a second phase dispersed in a ceramic matrix influences the resistance to crack propagation or toughness of the ceramic. The toughness increases when the second phase has a higher G_c value than that of the matrix and decreases when the second phase has a lower G_c than that of the matrix. From the model presented in this paper, it is clear that tough ceramic cannot be made from single phase homogeneous material; the route to tough ceramic exists through the development of multiphase composite. However, to increase the strength as well as toughness, the bonding between the dispersed phases and the matrix should be at least as strong as that of the matrix itself. From this standpoint, the "self-generated" composites derived from the eutectics of mixed oxides merit serious consideration as potential tough ceramics.

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