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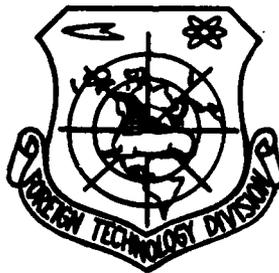
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BRITTLENESS AND TOUGHENING OF CERAMICS

by

Guo Jingkun



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BRITTLENESS AND TOUGHENING OF CERAMICS

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Abstract

Improving the brittleness of ceramic materials has long been a problem of concern to ceramists. Fiber-reinforcing and zirconia phase transformation toughening have been proven to be effective approaches in practice for improving the brittleness as well as the strength of ceramics. This paper will briefly discuss some problems in the study of the above two approaches. The importance of chemical compatibility between fiber and matrix is pointed out in the study of fiber-reinforced ceramic composite materials; the matches of the two in thermal expansion and elastic modulus can not be neglected either, and the moderate bonding of the two boundary surfaces is considered appropriate. In the study of zirconia phase transformation toughening, problems such as the retention of tetragonal zirconia ($t\text{-ZrO}_2$), the transformable $t\text{-ZrO}_2$ induced by stress and the transformable $t\text{-ZrO}_2$ which contributes to the toughening actions, etc. are discussed. The related properties of Y-TZP and ZTA are also listed in this paper.

Chinese Journal of Ceramics, 1987, 13(1)

I. Preface

When comparing ceramic material with metallic material, many inherent properties of the former, such as high temperature dynamic property, chemical corrosion resistance property, electric insulation, hardness, wear resistance, etc. are better than those of the latter, but the nature of atom arrangement in the molecular structure has determined that ceramic materials lack the kind of elastic transformation ability like that of metallic materials. Therefore, other than increasing the surface energy of the newly fractured surface there are almost no other functions that can absorb energy during the fracturing process. This result has determined the brittle nature of ceramic materials. The brittle nature of ceramic materials is difficult to change, but it can be improved using various approaches. This is a problem we have always been concerned ^{about} and studied about.

After going through an extended period of complications, people have found two more effective approaches by which the brittleness of ceramic materials can be improved, i.e. fiber-reinforcing and using zirconia phase transformation toughening. The results of our studies on the above two areas are briefly introduced below.

II. Fiber-Reinforcing Ceramic Composite Materials

1. Compatibility between fiber and matrix

In order to make good composite materials, the chemical and physical compatibility between fiber and matrix must be considered first. Chemical compatibility mainly means that fiber and matrix must not react chemically under the required temperature, and of course this also includes that the fiber itself does not result in any property degeneration under the said temperature. Physical compatibility mainly means that the two thermal expansion properties can match and that they should make the matrix portion (ceramics or glass) of composite material produced capable of sustaining under the state of compressive stress, thus making it possible for the composite material to possess strengthening and toughening properties. Secondly, it is the matches of the two in elastic modulus. Only when fiber possesses higher elastic modulus can it be possible for the fiber to share more load.

2. Several promising composite material systems

1) C fiber/Quartz glass composite material^[1]

C fiber and quartz glass are not likely to react chemically under proper production temperature, and the axial thermal expansion coefficient of C fiber matches the thermal expansion coefficient of quartz glass which facilitates the formation of composite material. Table 1 is the related properties of the said composite material.

It can be seen from Table 1 that both strength and toughness of C fiber/quartz composite material have been greatly increased compared to those of quartz glass itself. Bending strength increases 11 times and the work of fracture increases by two degrees of magnitude. It also has extremely good thermal shock resistance property and is a promising heat resistance material.

Table 1 Properties of C fiber/Quartz glass composite

Material	C fiber/Quartz glass composite	Quartz glass
Bulk density(g/cm ³)	2.0	2.16
Fiber content(vol%)	30	
Bending strength(MPa)	600	51.5
Work of fracture(J/m ²)	7.9 × 10 ⁴	5.9—11.3
Thermal diffusion coefficient (cm ² /s)	0.0077(900°C) ^a	

• Fiber cross arrangement.

2) C fiber/Si₃N₄ composite material^[2]

C fiber will react with Si₃N₄ at temperature above 1600°C (Figs. 1 and 2), and meanwhile since the mismatch of the two thermal expansion coefficients often results in cracks in the matrix of composite material, ways must be found to reduce the production temperature of composite materials of this system and to adjust the incompatibility in their thermal expansion. Table 2 shows the properties of C fiber/Si₃N₄ composite material. It is observed that, as a reinforcing agent for Si₃N₄, C fiber can make its toughness increase greatly while being unable to also make its strength increase. This is due to the reason that the elastic modulus of matrix is greater than the elastic modulus of fiber.

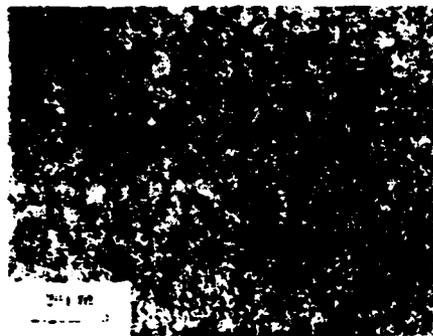


Fig. 1 No indication of occurrence of chemical reactions between C fiber and Si_3N_4 at 1600°C.

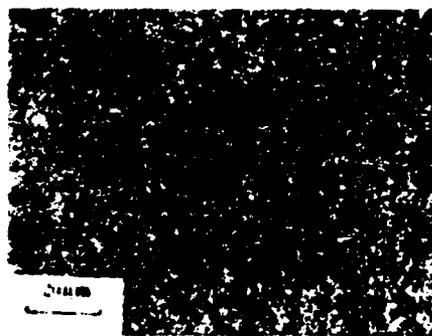


Fig. 2 Obvious reaction has occurred between C fiber and Si_3N_4 at 1650°C.

Table 2 Properties of C fiber/ Si_3N_4 composite

	C fiber/SMZ- Si_3N_4	SMZ- Si_3N_4
Bulk density(g/cm ³)	2.7	3.44
Fiber content(vol%)	30	
Bending strength(MPa)	454 ± 42	473 ± 30
Work of fracture(J/m ²)	4770 ± 770	19.3 ± 0.2
Fracture toughness, K_{Ic} (MPa·m ^{1/2})	15.6 ± 1.2	3.7 ± 0.7
Thermal expansion coefficient, $\times 10^{-6}$ (R.T.-1000°C)	2.51	4.62

• SMZ consists of Li_2O , MgO , SiO_2 , and ZrO_2 .

3) Dielectric composite material

As a dielectric material, it is also required that it possesses better toughness thereby setting a certain limit on the selection of fiber and matrix materials. BN fiber and quartz glass are the reinforcing agents currently available for selection.

BN fiber/ Si_3N_4 is a composite material system available for selection. In order to better preserve BN fiber technologically, the reactive burned Si_3N_4 (RBSN) may be used as the matrix, BN fiber/quartz glass is also a better matching system under proper technological production temperature. Quartz glass fiber/Aluminum phosphate is a composite material system with characteristics of being technologically simple and convenient to process and being easy to be made into large pieces of products.

These three composite materials possess better dielectric properties. Table 3 lists their related properties. Fig. 3 is the SEM photograph of the cross-section of BN fiber/RBSN.

Table 3 Properties of dielectric composite

Material	BN fiber/ RBSN	Reaction- bonded Si ₃ N ₄ (RBSN)	BN fiber ^(*) / Quartz glass	Quartz glass	Quartz glass fiber/ Aluminum phosphate
Dielectric constant	3.96	8.3	3.79	3.8	2.5-3.5
Dielectric loss (tgδ)	6.7 × 10 ⁻³	9 × 10 ⁻³	<10 ⁻³	<10 ⁻³	1-5 × 10 ⁻³
Bulk density(g/cm ³)	2.16	2.40	2.05	2.16	1.75
Fiber content (wt%)	10		15		20-50
Bending strength (MPa)	57	170	87.1	51.5	40-60
Work of fracture (J/m ²)	34.5	12.7	10.1	5.9-11.3	150-300



Fig. 3 SEM photograph of the cross section of BN-fiber/reaction bonded Si₃N₄ composite



Fig. 4 Fracture of SiC fiber due to fiber mismatch of SiC fiber/Si₃N₄ matrix in thermal expansion coefficient

4) SiC fiber (crystal tassel) reinforcing Si₃N₄ composite material

Excessive disparity in the difference between thermal expansion coefficients of fiber and matrix ($\alpha_{SiC} > \alpha_{Si_3N_4}$, close to one fold) will cause SiC fibers in the composite material produced to fracture. Therefore, it is necessary to adjust the thermal expansion coefficient of matrix. The main technological issue for using crystal tassel as reinforcing agent is uniform dispersion. Fig. 4 shows cracks of fiber occurred due to mismatch of SiC fiber and Si₃N₄ matrix in thermal expansion coefficient.

3. Toughening effects of fiber-reinforced ceramic composite materials

When fiber and matrix are selected properly, the composite material formed will benefit greatly in strength and toughness, such as C fiber/quartz glass composite ma-

terial. The majority of systems, however, can all obtain a higher toughening effect. Several mechanisms which can consume external energy are described in Fig. 5 among which pulling fiber out of matrix requires the most energy. Figs. 6 and 7 show the SEM photographs of the fracture of C fiber/quartz glass composite material and cross-section of C fiber/Si₃N₄ composite material respectively. It can be observed in the photographs that the pulling out of C fibers are obvious, and as a result their toughness is also high.

Therefore, it is best that the following requirements are satisfied in order to produce high strength, high toughness composite materials:

(1) Fiber or crystal tassel with high strength, high modulus (both should be greater than those of the matrix).

(2) Under composite material production conditions (e.g. temperature and atmosphere), properties of fiber or crystal tassel must not degenerate.

(3) Fiber or crystal tassel must not react chemically with the matrix under the conditions listed in item No. (2).

(4) Thermal expansion coefficients match, and it is best if α_{fiber} is appropriately larger than α_{matrix} .

(5) In composite material, it is better that the bonding strength between fiber and matrix reaches this extent; i.e. not only effective transfer of matrix stress onto fiber is guaranteed, but also sufficient length of fiber is pulled out from the matrix.

III. Zirconia Phase Transformation Toughening

Utilizing the volume effect caused during the phase transformation of zirconia in order to reach the goal of toughening the ceramics is one of the breakthroughs in the study of ceramics in the recent 10 years.

Lately we used the general thermal compression burning technology to make the zirconia toughened ceramics, and test results of strength of 1500MPa and fracture toughness of 15.6MPa·m^{1/2} can be reached.

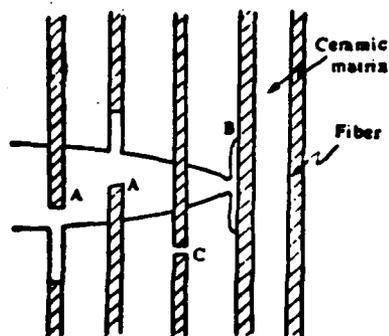


Fig. 5 Several possible mechanisms for energy absorption in fiber-reinforced ceramic composite

Key: A---Fiber pull out; B---Interface debound; C---Fiber fracture.

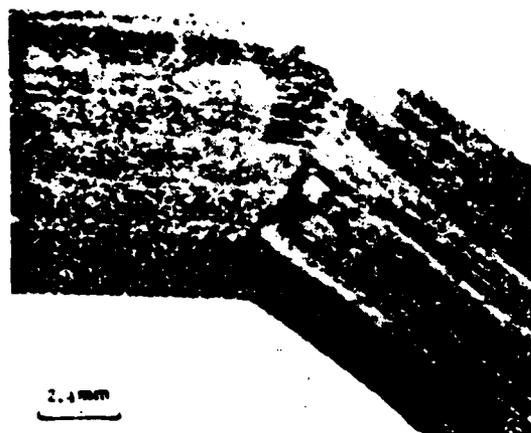


Fig. 6 Fracture of C fiber/SiO₂ composite



Fig. 7 SEM photograph of the cross-section of C fiber/Si₃N₄ composite

In addition to applying technology of Y₂O₃-containing tetragonal ZrO₂ multi-grain (Y-TZP) ceramics and ZrO₂-toughened aluminum oxide (ZTA) ceramics, we have also studied a few basic questions on the phase transformation toughening of ZrO₂^[4-6]. First of all, the strengthening and toughening of ceramics is realized through the process of transforming tetragonal phase ZrO₂ (t-ZrO₂) into single slanting phase of ZrO₂ (m-ZrO₂). Therefore, sufficient t-ZrO₂ must be retained in the ceramics. Secondly, are all t-ZrO₂ capable of being transformed into m-ZrO₂ and what are the transformation conditions? Thirdly, are all phase transformation processes contributive to strengthening and toughening of ceramics? And the last one is how to make the contributive (or ^{also} called effective) phase transformation processes reach their maximum and

be technologically feasible?

1. Retention of tetragonal ZrO_2

First of all, for Y-TZP ceramics it is to retain the maximum limit of t- ZrO_2 in the raw material. Fig. 8 shows the relation between particle size (D_p) of $Y_2O_3-ZrO_2$ powder and fraction ($\chi_m\%$) of m- ZrO_2 . It can be observed that without adding the stabilizing agent Y_2O_3 the phase composition of ZrO_2 powder is still 100% single slanting phase even if its particle size is as small as $0.025\mu m$. In theory, it is estimated that the critical size for retaining tetragonal phase in the ZrO_2 powder is 105\AA , and this is consistent with the aforementioned results. In the meantime, the larger the mean particle size is, the higher its fraction of m- ZrO_2 is; the higher the fraction of Y_2O_3 is, the less the fraction of single slanting ZrO_2 is.

After the Y-TZP ceramics is produced, the influence of ZrO_2 grain size (D) on the fraction of t- ZrO_2 ($\chi_t\%$) is also very important (Fig. 9), and the highest t- ZrO_2 fraction only exists within a very narrow range of grain size.

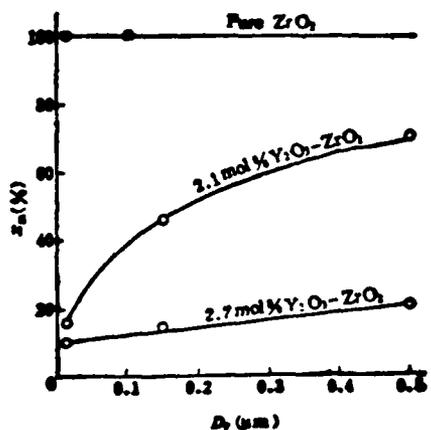


Fig. 8 Particle size of $Y_2O_3-ZrO_2$ (D_p) vs fraction of m- ZrO_2 ($\chi_m\%$)

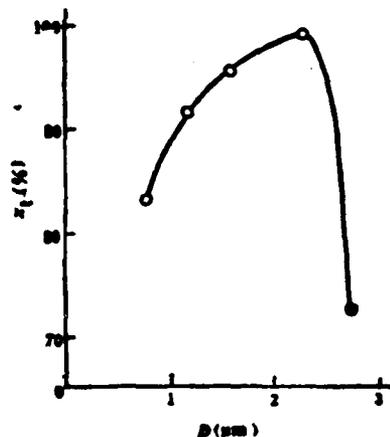


Fig. 9 Mean grain size of ZrO_2 grains in 2.1 mol% $Y_2O_3-ZrO_2$ sample (D) vs fraction of t- ZrO_2 ($\chi_t\%$)

2. Transformable tetragonal ZrO_2

Crushing Y-TZP ceramics thoroughly and grinding it to release the suppressive effect by neighboring crystals should make all transformable t- ZrO_2 transform into m- ZrO_2 . Therefore, the difference in fraction of m- ZrO_2 before and after crushing and

grinding could be interpreted as the volume fraction (ΔV_t) of transformable t-ZrO₂ induced by stress. Fig. 10 shows the relation between Y-TZP grain size (D) and ΔV_t . Fig. 9 corresponds perfectly with Fig. 10 in that the greater the fraction of t-ZrO₂ (χ_t) is, the higher the volume fraction (ΔV_t) is. As it should be, the corresponding fracture toughness is also at its maximum (Fig. 11).

In Y-TZP ceramics, not all t-ZrO₂ can be transformed into m-ZrO₂ as induced by stress. Fig. 12 shows the phase transformation situation on both sides of the crack in the Y-TZP material. On the upper left-hand side of the crack, all tetragonal ZrO₂ grains have been transformed into twin-crystal m-ZrO₂, whereas those on the lower right-hand side of the crack remain as t-ZrO₂. This indicates that not all t-ZrO₂ grains can be transformed.

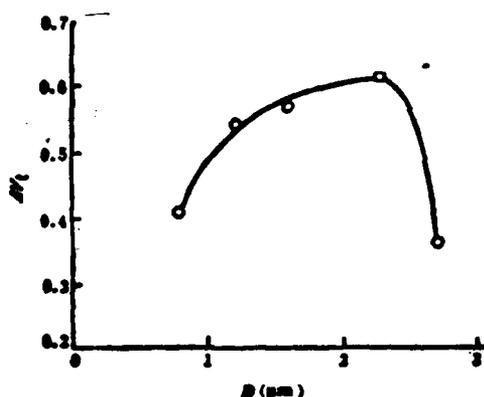


Fig. 10 Mean grain size of ZrO₂ grains in 2.1 mol% Y₂O₃-ZrO₂ sample (D) vs volume fraction of t-ZrO₂ transformable as induced by stress (ΔV_t).

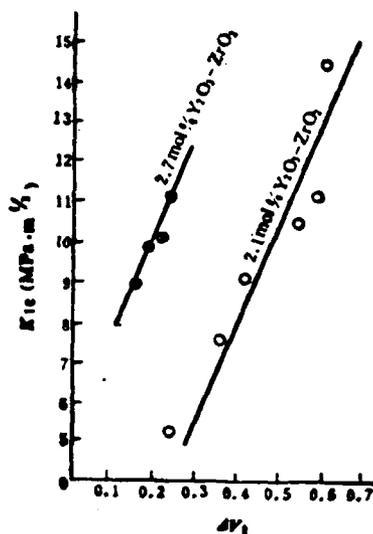


Fig. 11 Volume fraction of t-ZrO₂ transformable as induced by stress (ΔV_t) vs fracture toughness (K_{1c})

3. ZrO₂ grain phase transformation and relations between their crystal orientation and structure

Direct observation and study of phase transformation of ZrO₂ grains using the transmittance electronic microscope indicate that; phase transformation often begins at the crystal boundary; it is observed from the studies of electronic diffraction diagrams of different orientations that when t-ZrO₂ → m-ZrO₂, its crystal structure

changes into $(100)_c // (100)_m$ and that the said plane is the inertia state plane of Ma's [*] body phase transformation whereas its crystal axial orientation relation is $[001]_c // [001]_m$ and $[010]_c // [010]_m$.



Fig. 12 TEM photograph of different t-ZrO₂ phase transformation on both sides of a crack in a Y-TZP ceramic sample

In addition to the above phase transformation and crystal plane orientation relations, it is also related to the extent of structural tightness between adjacent crystal grains. Fig. 13 shows 5 t-ZrO₂ grains connected together and the three grains A, B and C have the same crystal axial orientation $[011]$ after being measured by electronic diffraction. Phase transformation occurs after being irradiated by strong electronic beam. Phase transformation begins at the crystal boundary between crystal grain A and crystal grain D, then it expands to crystal grain A and further to the crystal boundary of crystal grain B causing crystal grain B to transform, but not crystal grain C (see Fig. 14). It is observed from the studies of their electronic diffraction diagrams that the angle between crystal grain A and crystal grain B (200) is 53°, whereas the angle between crystal grain A and crystal grain C (200) is 64°. Moreover, looking from the stand point of adjacent structural arrangement, crystal grain B and crystal grain A are even more tightly packed.

It is concluded from the microscope studies of t-ZrO₂ in ZrO₂-toughened Al₂O₃ ceramics (ZTA) that: phase transformation of t-ZrO₂ is related to its location in the Al₂O₃ matrix. Those t-ZrO₂ grains wrapped inside Al₂O₃ crystal grains are the most

[*] as transliterated



Fig. 13 Y-TZP



Fig. 14 Phase transformation of t-ZrO₂ grains in Fig. 13 occurred after being irradiated by electronic beam

difficult to undergo phase transformation; those located between Al₂O₃ crystal grain boundaries or the intersection of crystal boundaries come in second, and those with other t-ZrO₂ grains adjacently connected come in a further second; those with multi-grain of t-ZrO₂ grouped together are the easiest to undergo phase transformation. This condition may be related to the extent of suppression around t-ZrO₂. Since the t-ZrO₂ inside Al₂O₃ grains is wrapped by them, its state of being suppressed is very difficult to be relieved; whereas the concentrated body t-ZrO₂ is located in the weakest suppression state and therefore is the easiest one to undergo phase transformation.

4. Technological considerations

It is concluded from the above discussions that retaining the maximum limit of t-ZrO₂ fraction in ceramics is the condition which must be achieved in technological consideration. Increasing transformable t-ZrO₂ fraction is technologically related to particle size and grain size, and it is also related to the Y₂O₃ fraction. As far as practical toughening effects are concerned, there is also only a portion in the transformable t-ZrO₂ fraction which exerts practical effects on toughening and it is called the contributive transformable t-ZrO₂. The higher the fraction of contributive t-ZrO₂ is, the greater the value of fracture toughness (K_{1c}), and so is strength. Considering from the stand point of technology, there seems no other more direct production monitoring factors as to how to increase the fraction of contributive t-ZrO₂ other than strictly controlling the technological parameters. This requires more thorough probes. Table 4 lists related properties of yttrium-containing tetragonal ZrO₂ multi-grain and ZrO₂-toughened Al₂O₃ (ZTA).

Table 4 Properties of Y-TZP and ZTA Ceramics

Material	Y-TZP		ZTA			
	Hot pressed	Sintered	ZrO ₂ (vol%)			
			15	25	50	75
Bending strength(MPa)	1530	1020	950	960	1000	1590
Fracture toughness(MPa·m ^{1/2})	15.6	10.3	8.8	6.6	11.1	14.4

IV. Conclusions

The two approaches of fiber-reinforcing and zirconia phase transformation toughening have both proven to be effective in practice for improving the brittleness of ceramics and toughening ceramics. Even though they have each found some practical applications, perfection is still required in order to reach the best technological level; besides, both still have numerous basic problems that await further and more thorough studies to provide theoretical basis for perfection technologically.

Nonetheless, the progress in the studies of both fiber-reinforcing and zirconia phase transformation toughening as well as the results obtained are encouraging.

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