Journal of the European Ceramic Society **19** (1999) 1903–1909

PII: S0955-2219(98)00289-1
0955-2219/99/$ - see front matter

The Effect of Whisker Orientation in SiC Whisker-reinforced Si_3N_4 Ceramic Matrix Composites

Wang Chang-an,[a]* Huang Yong[a] and Zhai Hongxiang[b]

[a]The State Key Lab of New Ceramics and Fine Processing, Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, People's Republic of China
[b]Department of Mechanical Engineering, Northern Jiaotong University, Beijing 100044, People's Republic of China

(Received 13 August 1998; accepted 28 November 1998)

Abstract

Si_3N_4 ceramic matrix composites reinforced by nearly unidirectionally aligned SiC whiskers have been prepared by extrusion and hot pressing. Unlike the case in traditional Si_3N_4 ceramic matrix composites reinforced by random SiC whiskers, the mechanical properties of the composites exhibit a significant dependence on whisker orientation. In the direction of whisker alignment for SiC(w)/Si_3N_4 composites, increments in bending strength and fracture toughness of 200 MPa and 3 MPa·$m^{1/2}$ are obtained respectively, compared to the values in the direction perpendicular to whisker alignment. Based on microscopic fractographic observation and micromechanics analyses, the effects of whisker orientation on toughening mechanisms are discussed. The results indicate that the whisker orientation, θ, is a decisive factor for the essential toughening mechanisms of whiskers. Only in the case of small θ and weak interface can whisker pullout occur, and whisker has maximum toughening effect. The results show that effects of whisker strengthening and toughening can be improved simultaneously through whisker oriented alignment. © 1999 Elsevier Science Limited. All rights reserved

Keywords: composites, whisker, mechanical properties, Si_3N_4, SiC.

1 Introduction

It is well known that whiskers are effective reinforcements for ceramics, and considerable development of whisker-reinforced ceramic matrix composites (CMCs) has been made in recent decades.[1–4] It is believed that by incorporating whiskers into a ceramic matrix with careful process control, not only fracture toughness but also strength can be significantly increased, and the scatter in strength can be reduced.[5,6] Another advantage of the whisker reinforced CMCs system is its superior oxidation resistance at high temperature compared with most continuous fiber or metal dispersion reinforced CMCs.[7]

A major purpose of the toughening lies in improving the strength reliability and the flaw tolerance of the brittle ceramics.[7] This issue is closely related with the fracture resistance character of the toughened material. The enhanced fracture toughness of CMCs with whiskers is believed to originate mainly from the additional energy dissipation through interaction between whiskers and matrix in the crack bridging zone behind the crack tip.[2,7–9]

Previous work has suggested that the essential toughening mechanisms of whisker reinforced CMCs include whisker bridging, frictional pullout, and crack deflection.[2,8–10] However, these experiments did not take into account the effect of whisker orientation, which is considered as one of the sensitive parameters affecting toughening process. For continuous fiber reinforced composites, fibers have one or two preferred orientations, and the reinforcing mechanics has been extensively studied.[11] However, for whisker reinforced composites, whiskers are usually randomly oriented, consequently the detailed toughening process appears to be more complicated, because in this case the toughening process would be expected to be sensitive to the whisker orientation.[12–14] In most long grain or whisker reinforced CMCs, the

*To whom correspondence should be addressed.

reinforcements are randomly oriented within surrounding matrix, while if the processing methods involving extrusion,[15] tape-casting[16] are taken, certain preferred orientation of whiskers may occur.

The present work seeks to investigate the relation between toughening action and orientation of whiskers in whisker-reinforced ceramic composites. For this purpose, SiC(w)/Si_3N_4 composites with nearly unidirectionally aligned SiC whiskers were prepared by extrusion and hot-pressing, and the relationship between whisker orientation and mechanical properties of SiC(w)/Si_3N_4 composites were examined. Based on an analysis of the microstructure, three essential toughening mechanisms are presented. Furthermore, micromechanics analysis are presented which examine the effects of whisker orientation on toughening mechanisms.

2 Experimental Procedure

The starting powder was prepared by ball milling of a commercial-grade Si_3N_4 powder (E-10, Ube Industries, Ltd., Tokyo, Japan) with sintering additives (8 wt% Y_2O_3, 1·5 wt% MgO and 2·5 wt% Al_2O_3) for 24 h in ethanol. A commercial-grade SiC whiskers (TWS-400, Tokai Carbon Co., Ltd., Kanagawa, Japan) were mixed with the starting powder by ball milling in ethanol for 24 h. The whisker content was 20 wt%. After drying, the blended powder was mixed with 10 wt% polyethylene ethanol (PVA) and 3 wt% glycerine and 2 wt% liquid paraffin and kneaded by roll mixer, and extruded through a 30 mm diameter tubing to form green wires of 1 mm diameter. The wires were placed in graphite dies parallel to the axial direction of wires. After burning-out the organic binders, the compacts were hot-pressed at 1820°C for 90 min in 0·1 MPa N_2 atmosphere under 22 MPa pressure.

The hot-pressed plates were cut into bars for testing mechanical properties along different direction at an angle of θ with respect to the direction of whisker oriented alignment, namely defined as whisker orientation angle as shown in Fig. 1. Bending strength was determined by three point bend testing (test bars $4\times3\times36\,mm^3$). The tensile surface of the samples was polished with diamond paste down to 1 μm and the corners were rounded with a 15 mm diamond grinding wheel. Fracture toughness was measured by SENB method (test bars $4\times6\times30\,mm^3$), and the width of the notch is less than 0·25 mm.

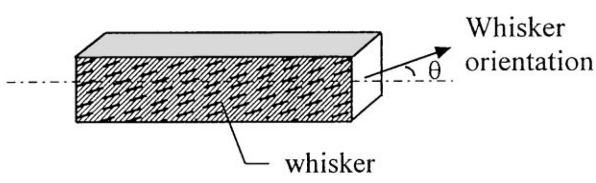

Fig. 1. The schematic diagram of the definition of whisker orientation angle θ.

3 Experimental Results

3.1 Mechanical properties of SiC(w)/Si_3N_4 composites in the different whisker orientation

Figure 2 shows a SEM image which indicates the degree of alignment of the SiC(w)/Si_3N_4 composites. It can be seen that whiskers in the SiC(w)/Si_3N_4 composites are nearly unidirectionally aligned.

Figures 3 and 4 show the bending strength and fracture toughness of the SiC(w)/Si_3N_4 composites at the different whisker orientation angles, respectively. It can be seen from Fig. 3 that the bending strength of the composites is relatively insensitive to whisker orientation angle when $\theta \leq 60°$, while there is a significant drop at larger whisker orientation

Fig. 2. SEM micrograph of oriented SiC(w)/Si_3N_4 composites.

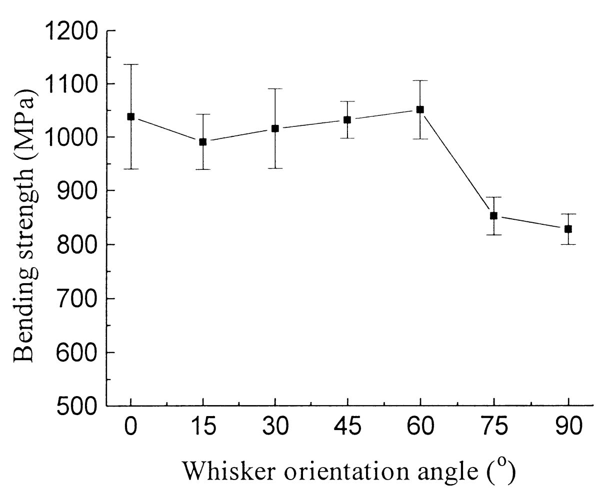

Fig. 3. The bending strength of oriented SiC(w)/Si_3N_4 composites versus whisker orientation angle.

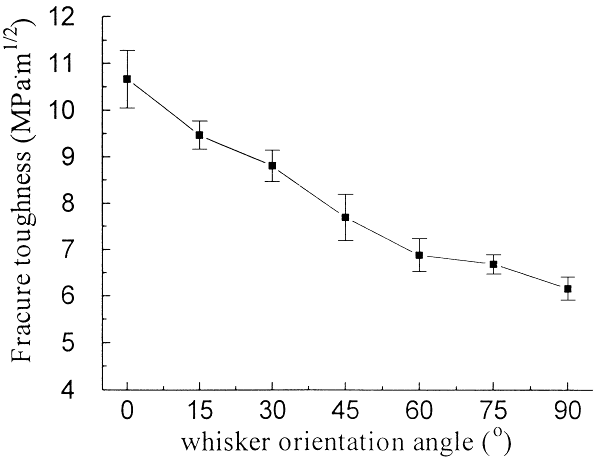

Fig. 4. The fracture toughness of oriented $SiC(w)/Si_3N_4$ composites versus whisker orientation angle.

angle as 75° and 90°. On the other hand, the fracture toughness of the $SiC(w)/Si_3N_4$ composites exhibits an approximately linear decrease with increasing whisker orientation angle, as shown in Fig. 4. From both Figs 3 and 4, it can be found that mechanical properties of oriented $SiC(w)/Si_3N_4$ composites exhibit obvious anisotropy, and reach maximum values, 1038 MPa for bending strength and 10·7 $MPa{\cdot}m^{1/2}$ for fracture toughness, at the whisker orientation angle $\theta = 0°$, i.e. in the direction of whisker oriented alignment. The results suggest that effects of whisker strengthening and toughening can be improved simultaneously through whisker oriented alignment.

3.2 Fracture surface observations and three main modes of whisker toughening mechanisms

In order to understand the relationship between whisker reinforcing mechanisms and whisker orientation, fracture surfaces of the specimens were observed with SEM. Figure 5 shows a set of typical micrographs of the specimen with different whisker orientation angle. The common images observed are the exposed whiskers out of the cracked plane and the whisker marks due to whisker separated from the matrix. It can be seen that either the length of the exposed whiskers or the shape of the whisker marks changes with the whisker orientation angle. For a small orientation angle, the length of the exposed whiskers is small and the whisker marks are approximately circular in shape. With the angle increasing, the exposed whiskers lengthen and the whisker marks change to oblique half-cylindrical shape.

It can be seen from Fig. 5 that whisker orientation strongly influences the fracture process of the $SiC(w)/Si_3N_4$ composites. For a small orientation angle, whisker pull-out appears to be the dominant mechanism contributing to reinforcing effect, while for a large orientation angle, interfaces between whisker and matrix debond and whiskers peel off from the interfaces. Obviously, the latter process will consume less energy than the former, correspondingly, the reinforcing effect for the small orientation angle will be larger than that for the large orientation angle.

At a larger orientation angle, the debonded part of whisker bears not only a tensile force but also an additional bending moment at the same time. Consequently, the whisker is broken usually at its root near the crack plane. This is easy to observe from the fractured surfaces. For this reason, whisker pullout becomes difficult even with a weak whisker-matrix interface, unless the orientation angle is small enough. It can be seen from Fig. 5 that the amount of the pullout-hole-like marks reduces with increasing orientation angle. Due to a certain orientation angle, the debonded whisker leads to mechanical damage to the matrix near the root of the whisker just likes a crowbar upon the matrix. The oblique half-cylindrical marks are clearly the result of matrix damage.

Figure 6 shows three representative crack features for whisker-reinforced ceramic matrix composites: (a) a broken whisker ejected from the cracked plane; (b) a whisker being pulled out of the matrix; (c) the local matrix damage around the root of whisker. According to the observation, we consider three essential modes, whisker bridging, whisker pull out, and matrix damage, as the essential toughening mechanisms for whiskers. Other toughening mechanisms for whiskers, such as crack deflection, can be ascribed to these essential modes. The three essential modes are illustrated in Fig. 7 for further analyzing and discussing the dependence of the toughening mechanism on reinforcement orientation.

4 Analysis and Discussion

Figure 7 illustrates the mechanical condition of a single whisker having partially debonded with the matrix in the crack-bridging zone behind the crack tip. S is traction force from the normal direction of crack plane, u is the crack opening corresponding to S, and θ is the orientation angle of whisker with respects to the direction of S, the same as the meaning above-mentioned. During crack separating, the whisker bears a tensile force and a bending moment at the same time, separately from the parallel component and the vertical component of S, and passes the forces to the interface and the bonding matrix. As the maximum tensile stress is concentrated on the surface of the root of whisker, the effect of the shear stress between whisker and

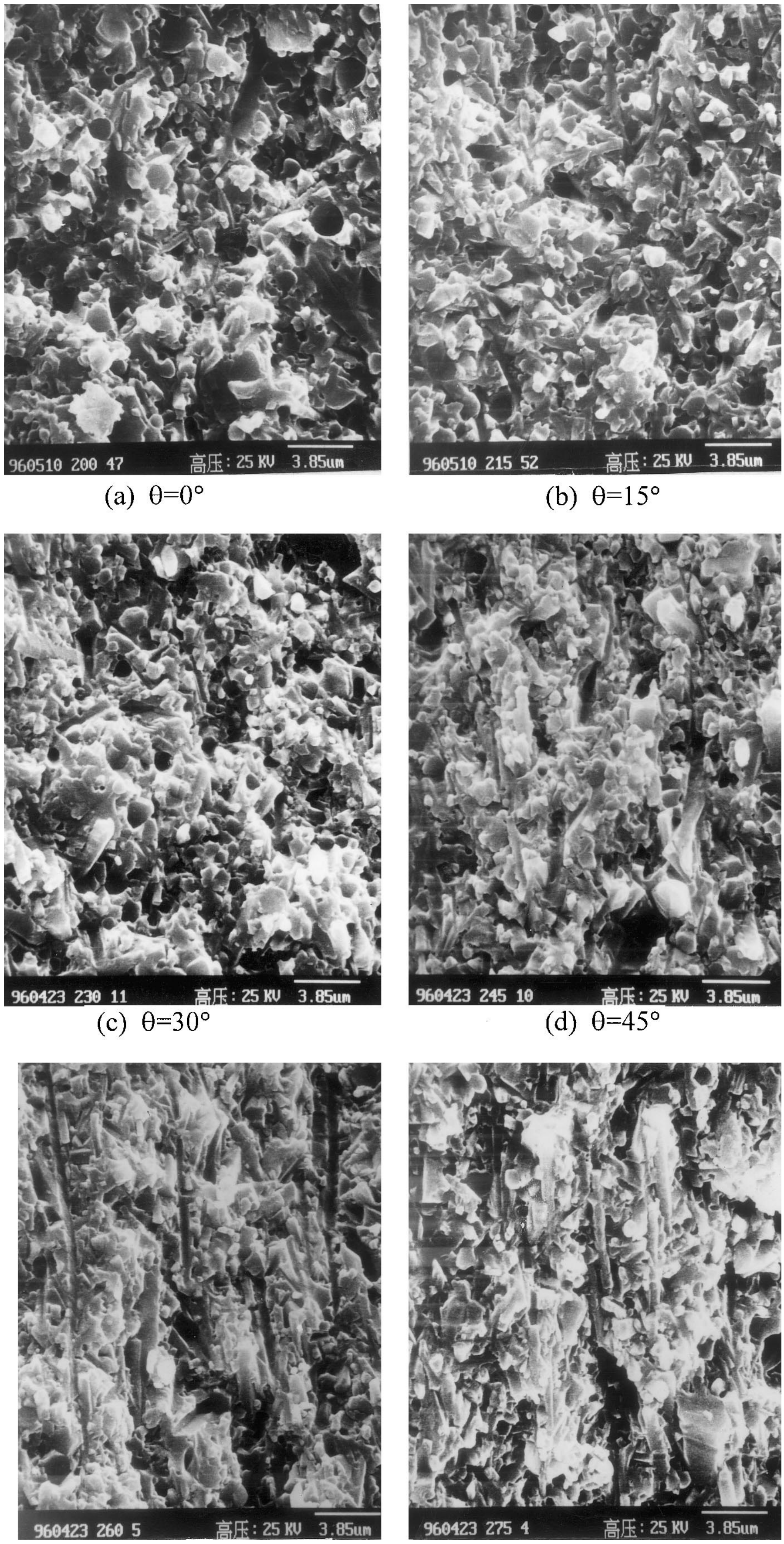

Fig. 5. Fracture surfaces of the oriented SiC(w)/Si_3N_4 composites with different whisker orientation angle: (a)–(f) respectively correspond to $\theta = 0-75°$.

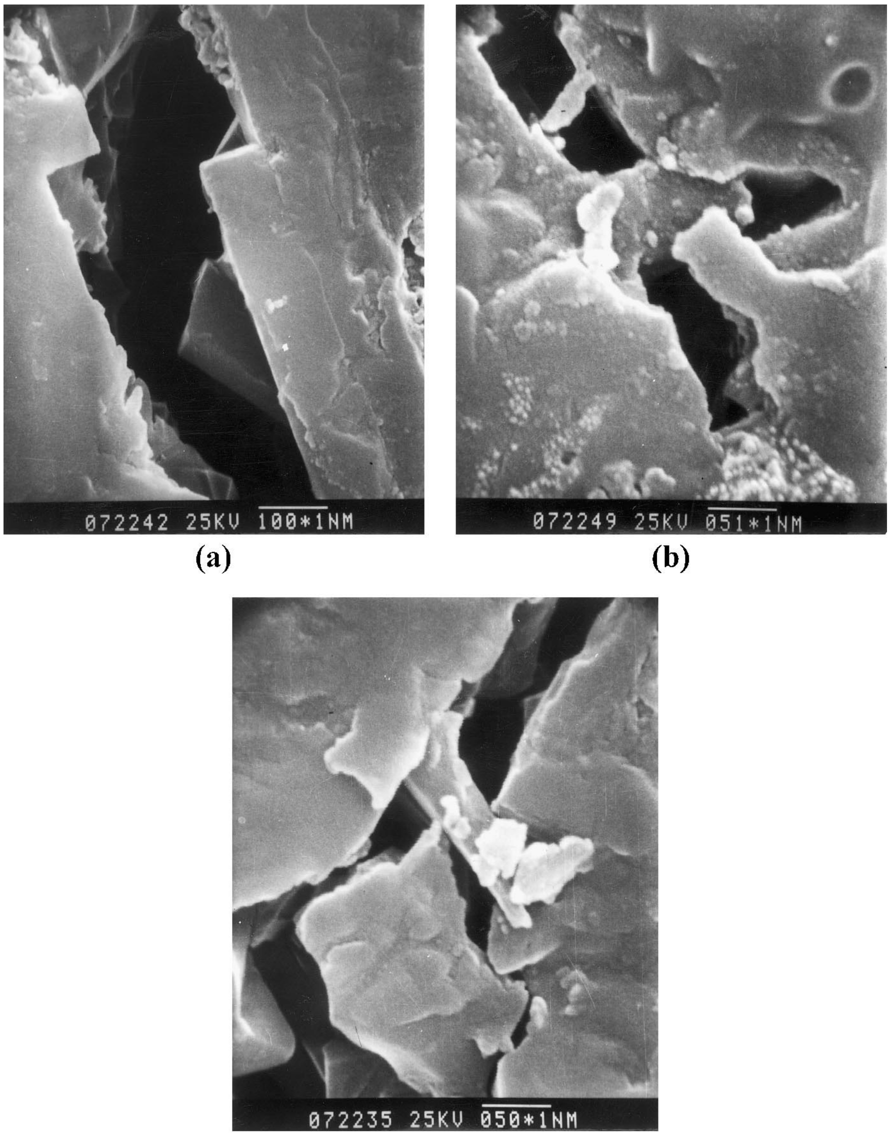

Fig. 6. Results caused by the whisker bridging, the whisker pullout, and the local matrix damage: (a) broken whisker ejected from the cracked plane; (b) whisker being pulled out of the matrix; (c) local matrix damage around the root of whisker.

matrix should be also considered for determining the maximum main stress, σ_w, on the whisker. For the interface between whisker and matrix, it can be generally considered that there is an interfacial layer with a certain thickness. It bears a shear force from the parallel component of S and a compressive force from the vertical component of S as well as a residual stress σ_r, and leads to a main stress, σ_i. As to the local matrix near the root of the whisker, the real stress condition may be quite complex due to the complexity of the shape and the load in there. As a first approximation, it can be simplified as a wedge that is infinitely great in thickness and bears a uniform compressive force generating from the vertical component of S on one side of it. This leads to a maximum tensile stress, σ_m, on the surface of the wedge, on the side of the compressive part, in terms of the classic elastic–mechanics solution.[17]

In terms of such analyses, the critical conditions for the whisker breaking, pullout, and the local matrix damage can be obtained as:[14]

$$U_c = \frac{2(L_{db}/R)\varphi_i(\theta)\cos\theta}{\varphi_2(\theta)\left(1+\sqrt{1+(L_d/R)^{-2}[\varphi_2(\theta)]^{-2}}\right)}, \quad (1)$$

for whisker breaking

$$U_c = \frac{2(\sigma_{ic}/\sigma_{wc})(L_d/R)(L_{db}/R)\varphi_1(\theta)\cos\theta}{\left[(1-\sigma_r\sigma_i)\tan\theta+\sqrt{\tan^2\theta+(1-\sigma_r/\sigma_i)}\right]^{-1}},$$

for whisker pullout

(2)

$$U_c = \frac{(\sigma_{mc}/\sigma_{wc})(L_d/R)(L_{db}/R)(\tan\alpha - \alpha)\varphi_1(\theta)\cos\theta}{\alpha\tan\theta},$$
for local matrix damage
(3)

where $U_c = E_w u_{cr}/R\sigma_{wc}$, $\varphi_1(\theta) = 1 + 4(L_{db}/\mathrm{R})^2 \tan^2\theta$, $\varphi_2(\theta) = 1 + 8(L_{db}/R)\tan\theta$, $\alpha = \pi/2 - \theta$. E_w, R, L_{db} and L_d are microstructural parameters denoting separately Young's modulus, radius, debonding length and bonding length of the whisker. σ_{wc}, σ_{ic} and σ_{mc} are microstructural parameters denoting the critical value of σ_w, σ_i and σ_m, respectively. u_{cr} is the critical crack opening corresponding to that any one of σ_{wc}, σ_{ic} and σ_{mc} is reached.

It is known, from eqns (1)–(3), that for given microstructural parameters the U_c is different depending on θ. This then leads to a dependence of the toughening mechanisms on orientation angle. Figure 8 shows a typical U_c–θ relationship determined by eqns (1)–(3). The signs, (1), (2) and (3), near the curves denote the equation number; on the corresponding curve σ_w, σ_i and σ_m reach their critical value. The shadow area surrounded by the curves stands for the whisker bridging. In this stage of crack separation the whisker bears the traction force, and absorbs energy through elastic deformation. Once u reaches any one of the curves ($u = u_{cr}$) the bridging action ends, and then the whisker is broken or pulled out or the local matrix is damaged depending on θ. In general, only in the case of small θ and weak interface can the pullout mechanism occur. For intermediate θ, the bridging action can be continued until the whisker breaking.

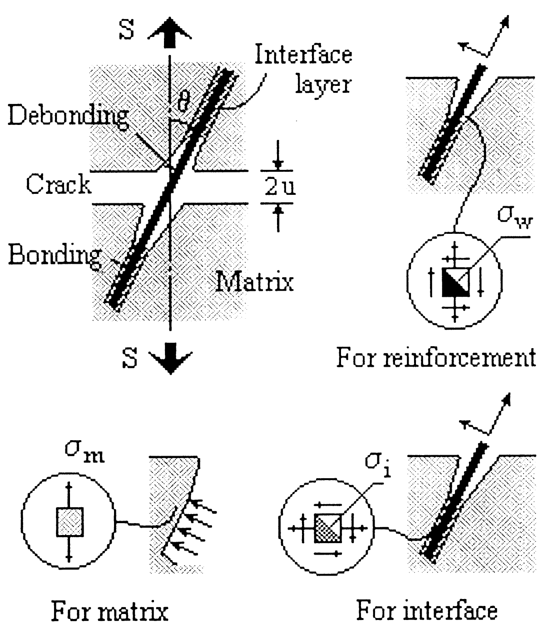

Fig. 7. Schematic diagram indicating mechanical condition of a single reinforcement having partially debonded with the matrix in the crack bridging zone behind the crack tip.

When θ is large, the local matrix will be damaged prior to the whisker breaking or pullout. These analyses are essentially in agreement with the above experimental results.

In the case of whisker oriented alignment, at a small orientation angle θ of whisker, most of whiskers are perpendicular to the crack plane, i.e. parallel to the direction of stress, so that whiskers can effectively transfer stress and a zone of whisker bridging at crack tip can form and develop well. Consequently, debonding and whisker pulling-out in this case will consume much more stored elastic strain energy and fracture toughness of the composites will be increased obviously. With the increasing of whisker orientation angle θ, the number of whiskers tilted to the normal of crack plane increases. In this case, it becomes more and more difficult for bridging whiskers to pull-out so that the contribution to toughness will decrease. That is why in the above experiments the fracture toughness of the composites gradually decreases with increasing whisker orientation angle. The bending strength of the composites versus whisker orientation angle θ exhibits the same tendency as that of the fracture toughness, only the tendency of drop is inapparent in the range of θ from 0° to 60°. As we know, $SiC(w)/Si_3N_4$ composites usually display intergranular fracture, therefore the bending strength of the composites is influenced by grain boundary phase or interfacial phase as well as whisker orientation. In the range of small and intermediate θ, the bonding strength of the grain boundary or interfacial phases may be main factor influencing the strength of the composites. However in the range of large θ, most of whiskers are nearly parallel to the crack plane so that they cannot transfer much stress from matrix, consequently the strength of the composites drops sensitively.

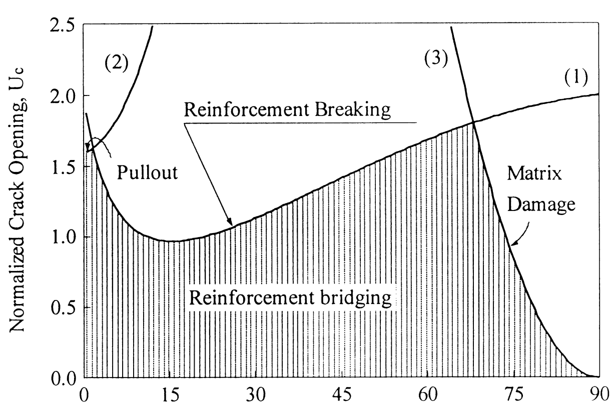

Fig. 8. Typical relationship between the toughening mechanisms and reinforcement orientation, for a single reinforcement having partially debonded with the matrix in the crack-bridging zone behind the crack tip.

5 Conclusions

1. The mechanical properties of the oriented $SiC(w)/Si_3N_4$ composites exhibit a significant dependence on whisker orientation. Along the direction of whisker oriented alignment, mechanical properties of the composites reach maximum values, 1038 MPa for bending strength and 10·7 MPa·$m^{1/2}$ for fracture toughness, respectively. With the increase of whisker orientation angle, both of strength and toughness of the composites decrease.
2. Based on observing and analyzing microstructure, three essential toughening mechanisms for whiskers in $SiC(w)/Si_3N_4$ composites were presented: whisker bridging, whisker pull out and matrix damage.
3. Micromechanics analysis indicates that the whisker orientation, θ, is a decisive factor for the essential toughening mechanisms of whiskers. Only in the case of small θ and weak interface can the pullout mechanism occur. For intermediate θ, the bridging action can be continued until the whisker breaks. When θ is large the local matrix will be damaged prior to the whisker breaking or pullout. These analysis agrees with the experimental results well.

Acknowledgements

This research work was supported by 'The High Technology Research and Development Program' ('863 Program') of China and the National Science Foundation of China (grant no. 59632090).

References

1. Wei, G. C. and Becher, P. F., Development of SiC-whisker-reinforced ceramics. *Am. Ceram. Soc. Bull.*, 1985, **64**, 298–304.
2. Becher, P. F., Hsuen, C. H., Angelini, P. and Tiegs, T. N., Toughening behavior in whisker-reinforced ceramic matrix composites. *J. Am. Ceram. Soc.*, 1988, **71**, 1050–1061.
3. Shalek, P. D., Petrovic, J. J., Hurley, G. F. and Gac, F. D., Hot-pressed SiC whisker/Si_3N_4 matrix composites. *Am. Ceram. Soc. Bull.*, 1986, **65**, 351–356.
4. Li, J. B., Huang, Y., Zheng, L. L. and Wu, J. G., Effects of additives on mechanical properties of $SiC(w)/Si_3N_4$ ceramic composite. In *Proceedings of the 1st International Symposium on the Science of Engineering Ceramics*, ed. S. Kimura and K. Niihara. The Ceramic Society of Japan, Tokyo, 1991, pp. 333–338.
5. Becher, P. F., Lin, H. T. and Alexander, K. B., Development of toughening ceramics for elevated temperatures. In *Proceedings of the 1st International Symposium on the Science of Engineering Ceramics*, ed. S. Kimura and K. Niihara K. The Ceramic Society of Japan, Tokyo, 1991, pp. 307–324.
6. Zhai, H. X., Huang, Y., Guo, C. and Cheng, J., Strength distribution in toughened ceramics exhibiting *R*-curve behavior. In *Proceedings of 5th International Symposium Ceramic Materials and Components for Engines*, ed. D. S. Yan, X. R. Fu, and S. X. Shi, World Scientific, Singapore, 1995, pp. 522–527.
7. Evans, A. G., Perspective on the development of high-toughness ceramics. *J. Am. Ceram. Soc.*, 1990, **73**, 187–206.
8. Becher, P. F., Microstructural design of toughened ceramics.. *J. Am. Ceram. Soc.*, 1991, **74**, 255–269.
9. Mai, Y-W. and Lawn, B. R., Crack-interface grain bridging as a fracture resistance mechanism in ceramics: II, theoretical fracture model. *J. Am. Ceram. Soc.*, 1987, **70**, 289–294.
10. Warren, R. and Sarin, V. K., Fracture of whisker-reinforced ceramics. In *Application of Fracture Mechanics to Composite Materials*, ed. K. Friedrich. Elsevier Science, 1989, pp. 571-614.
11. Evans, A. G., Domergue, J.-M. and Vagaggini, E., Methodlogy for relating the tensile constitutive behavior of ceramic-matrix composites to constituent properties. *J. Am. Ceram. Soc.*, 1994, **77**, 1425–1435.
12. Zhai, H. X., Toughening behavior and reliability of multiphase ceramics. Report of Postdoctoral Research, Tsinghua University, China, 1994, pp. 5-44.
13. Zhai, H. X., Wang, C. A. and Huang, Y., Toughening effects of CMCs reinforced directionally with whiskers. *High Technology Letters*, 1997, **7**, 11–15 (in Chinese).
14. Zhai, H. X., Wang, C. A. and Huang, Y., Reinforcing mechanisms and their orientation effect of whiskers in ceramic matrix. *High Technology Letters*, 1997, **7**, 26–30 (in Chinese).
15. Goto, Y. and Tsuge, A., Mechanical properties of unidirectionally oriented SiC-whisker-reinforced Si_3N_4 fabricated by extrusion and hot-pressing. *J. Am. Ceram. Soc.*, 1993, **76**, 1420–1424.
16. Matsui, T., Komura, O. and Miyake, M., The effects of surface coating and orienting of whiskers on mechanical properties of $SiC(w)/Si_3N_4$. *J. Ceram. Soc. Jpn.*, 1991, **99**, 1103–1109..
17. Xu, Z., Solutions for the plane problem. In *Tanxing Lixue, Shangche (Elastic Mechanics)*, Vol. 1, ed. Z. Xu. The People's Education Publish Press, Beijing, 1980, pp. 94-99.