Ceramics International 33 (2007) 385–388

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

Improvement of mechanical properties of Al_2O_3/Ti_3SiC_2 multilayer ceramics by adding SiC whiskers into Al_2O_3 layers

Zan Qingfeng [a,b,*], Dong Limin [a], Wang Chen [a], Wang Chang-an [b], Huang Yong [b]

[a] *Institute of Nuclear and New Energy Technology, Tsinghua University, Beijing 100084, PR China*
[b] *State Key Laboratory of New Ceramics and Fine Processing, Department of Materials Science and Engineering, Tsinghua University, Beijing 100084, PR China*

Received 3 August 2005; received in revised form 18 August 2005; accepted 4 October 2005
Available online 28 February 2006

Abstract

Al_2O_3/Ti_3SiC_2 multilayer materials with high mechanical properties in terms of bending strength and work of fracture have been prepared by in situ hot-pressing at 1600 °C for 4 h. In order to further improving the mechanical properties, SiC whiskers were added to Al_2O_3 layers as the 2nd-level toughening agent. With the content of SiC whisker increasing, the bending strength and work of fracture showed a similar trend of increasing firstly and then decreasing. Hence, the optimal mechanical properties of 688 MPa for bending strength and 2583 J/m^2 for work of fracture were obtained when the added content of SiC whisker was 20 wt%. These excellent properties should be attributed to 1st-level toughening mechanism (multilayer structural toughening), 2nd-level toughening mechanism (whisker toughening), and the synergy effect between them. Hence, adding SiC whiskers was an effective approach to improving the mechanical properties.

Keywords: B. Whisker; D. Al_2O_3; Ti_3SiC_2; Toughening mechanism; Multilayer ceramics

1. Introduction

Ceramics are brittle and sometimes they fail catastrophically. This limits their applications in engineering. In order to overcome the problem, studying of toughening becomes a very important research area in ceramics, especially structural ceramics. As a result, many toughening methods had been devised, researched and used in ceramics, such as whisker toughening [1–3], long- and short-fiber toughening [4,5], dispersed rigid particle toughening [6], ZrO_2 phase transformation toughening [7,8], and so on. These toughening methods were all effective for some ceramics, hereinto whiskers were used very widely.

Otherwise, multilayer structural toughening was proved as an effective toughening mechanism [9–12]. Since the SiC/C multilayer ceramics was first developed by Clegg et al. [9], this toughening approach has been used in many systems, such as Si_3N_4/BN [10,11], SiC/C [9,12], Al_2O_3/ZrO_2 [13], etc. and several 10-folds of fracture toughness and several 100-folds of work of fracture were achieved compared to the monolithic ceramics. Al_2O_3/Ti_3SiC_2 multilayer ceramics is a typical multilayer material, which possess good mechanical properties of 659 MPa for bending strength and 2159 J/m^2 for work of fracture [14].

In recent years, synergy mechanism between two or more toughening methods was reported [15]. By this toughening mechanism, the toughness effect was usually larger than the additional effect brought by the individual toughening mechanisms. The all fore-mentioned toughening mechanisms could be used in synergy toughening design of ceramics by appropriate methods.

In this paper, the SiC whiskers as 2nd-level toughening agent were added into Al_2O_3 layers in Al_2O_3/Ti_3SiC_2 multilayer ceramics. The 2nd-level toughening mechanism in the multilayer ceramics, the synergy effect between the multilevel toughening mechanisms and the fractural behavior of the whole materials will be described in this paper.

* Corresponding author. Tel.: +86 10 89796112; fax: +86 10 89796118.
E-mail address: Zanqingfeng@tsinghua.org.cn (Zan Qingfeng).

0272-8842/$32.00
doi:10.1016/j.ceramint.2005.10.007

2. Experimental procedure

2.1. Materials and preparation

The Al_2O_3/Ti_3SiC_2 multilayer ceramics were prepared by in situ hot-pressing method according to a procedure described elsewhere [14]. The raw materials of Ti_3SiC_2 layers were Ti, SiC and activated carbon powders. Otherwise, the Al_2O_3 layers were sintered from Al_2O_3 powders with MgO as sintering aid and SiC whiskers as mechanical property adjusting agent.

The flow chart for the preparation of the multilayer materials is shown in Fig. 1. Firstly, powders of Al_2O_3 (99.5% pure, Jixin Ceramic Co., China) with 0.3 wt% MgO (99.9% pure) and given certain quantities of SiC whiskers for the Al_2O_3 layer and powders of Ti (99.9% pure), SiC (99.9% pure) and activated carbon with the atomic ratio of 3:1:1 for the Ti_3SiC_2 layer were milled for 24 h in alcohol medium, respectively. Secondly, the green tapes of Al_2O_3 and Ti-SiC-C with thickness of about 200 μm both were manufactured by rolling after adding PVA, glycerol and liquid paffix into the raw powders. Then these green tapes were dried in air and punched into rounds (Ø 50 mm). They were packed into a graphite die by turns of Al_2O_3 tapes and the Ti_3SiC_2 raw powders tapes. The green body underwent a conventional binder burnout through heating in flowing air. Finally, the multilayer ceramic was hot pressed at 1600 °C for 4 h in Ar atmosphere with a pressure of about 20 MPa.

2.2. Tests and characterization

The specimens were sliced into test bars with the dimensions of 4 mm × 3 mm × 36 mm for bending strength and 4 mm × 6 mm × 30 mm for fracture toughness and the fracture work. Three-point bending test was carried out at room temperature with a span of 30 mm and a crosshead rate of 0.5 mm/min. The work of fracture and fracture toughness were determined by the single-edge-notch-beam method (SENB) at room temperature with a span of 24 mm and a crosshead rate of 0.05 mm/min. All the tests were performed on A-2000 Shimadzu universal materials testing machine. The loads were applied perpendicular to the plane of laminates. The microstructure of the materials was observed by SEM (CSM900).

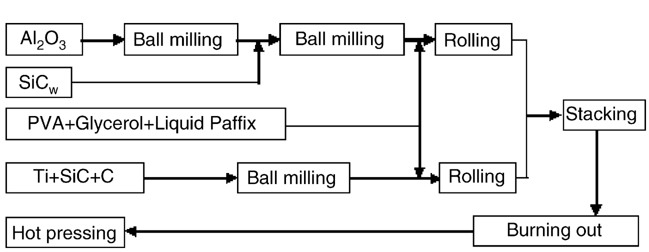

Fig. 1. Flow chart for the preparation of Al_2O_3/Ti_3SiC_2 multilayer materials with SiC whiskers.

3. Results and discussion

3.1. Mechanical properties

The mechanical properties of the Al_2O_3/Ti_3SiC_2 multilayer ceramics, including bending strength and work of fracture, are shown in Table 1 and Fig. 2. With increasing the content of SiC whisker, the bending strength of the materials slightly increased firstly, and then decreased when the content of SiC whisker exceeded 20 vol%. A similar trend was found for the work of fracture expected for the improved extent. Therefore, when whisker content was about 20 vol% (W20), the optimal mechanical properties of 688 MPa for bending strength and 2583 J/m^2 for work of fracture were obtained. Compared to the multilayer without whisker (W0), the gain in of strength and work of fracture for materials W20 were about 4.4 and 19.6%, respectively.

Otherwise, it was found that when whisker content was larger than 30 vol%, the specimens were broken after sintering and cooling, which looked likely because of the residual stresses (as described in the following section in detail).

3.2. Residual stresses analysis

For multilayer ceramics, especially with 'strong'-interface, the mechanical properties were reported to be influenced effectively by residual stress states [13], which was produced due to the thermal expansion mismatch between the interphase-layer and matrix-layer during cooling. The residual stresses would lead to cracking and three primary crack morphologies would be observed: transverse, longitudinal and debonding cracks if the residual stresses are large enough or the material loaded. For multilayer materials, the transverse and debonding cracks are favorable to improve the toughness.

The residual stresses states in Al_2O_3/Ti_3SiC_2 multilayer ceramics without SiC_W were already analyzed in our previous paper [16]. When SiC whiskers were added into Al_2O_3 layers, the residual stresses states were also changed. The residual stresses were calculated in the present work by the same method, the parameters adopted in the calculation method are listed in Table 2. The sintering temperature was 1600 °C, and the thermal expansion coefficients of the Al_2O_3, Ti_3SiC_2, TiC and SiC whisker were assumed to be constant from room temperature to sintering temperature. The thickness of Al_2O_3 layer and Ti_3SiC_2 layer were both about 100 μm. For the two layers both containing two components, these mechanical parameters of each layer all need to be calculated by the rule of

Table 1
The mechanical properties of the Al_2O_3/Ti_3SiC_2 multilayer ceramics with SiC_W

Samples	SiC_W% (vol%)	Bending strength (MPa)	Work of fracture (J/m^2)
W0	0	659.53 ± 47.3	2159 ± 153
W10	10	674.38 ± 25.6	2341 ± 146
W15	15	683.59 ± 34.1	2507 ± 143
W20	20	688.44 ± 35.2	2583 ± 142
W25	25	534.87 ± 68.2	2326 ± 135

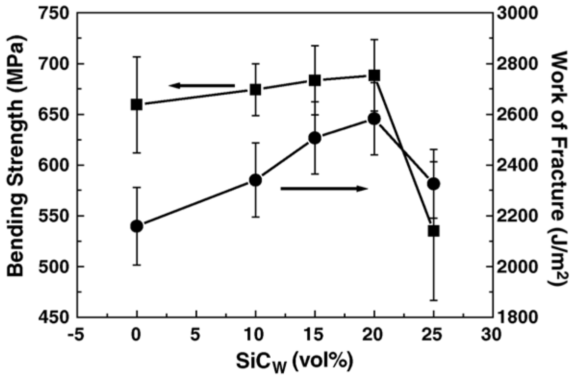

Fig. 2. Mechanical properties of the multilayer ceramics with SiC whiskers.

mixture. And the calculated results containing the mechanical parameters and residual stresses of each layer are shown in Table 3. With the content of whisker increasing, the residual stresses in Al_2O_3 layer changed from tensile to compressive. The residual compressive stresses could strengthen the Al_2O_3 layer, thereby the fracture behavior and the toughening mechanisms of the multilayer ceramics were also changed, as described in Section 3.3.

Otherwise, it is apparent from in Table 3 that when the content of SiC whisker reaches 30 vol%, the residual tensile stress in Ti_3SiC_2 layers is up to 372 MPa. According to the papers about Ti_3SiC_2 bulk ceramic [17,18], the bending strength was reported to be only about 260–320 MPa which was well-known higher than its tensile strength. Hence, the multilayer ceramics with 30 vol% or more SiC_W would be broken by the residual stresses during sintering and following cooling procedures.

3.3. Toughening mechanisms and synergy effect

The high toughness of multilayer ceramics should be attributed to different kinds of toughening mechanisms. In the Al_2O_3/Ti_3SiC_2 multilayer ceramics with SiC whiskers, these toughening mechanisms could be: multilayered structural toughening, whisker toughening and synergy effect by hereinbefore two mechanisms.

The toughening mechanisms induced by multilayered structure, defined as 1st-level toughening mechanisms in this paper, have been investigated in recent years [9–14], they include crack deflection mechanism, bridging mechanism of interlocking layers, frictional sliding mechanism, and so on.

Table 2
The mechanical parameters of Al_2O_3, Ti_3SiC_2, TiC and SiC whisker

	Materials			
	Al_2O_3	Ti_3SiC_2	TiC	SiC_W
E (GPa)	390	320	322	580
ν	0.30	0.20	0.20	0.20
CTE (10^{-6}/°C)[a]	8.8	9.2	7.74	4.5

[a] Assumed to be constant from room temperature to sintering temperature.

Table 3
Residual stresses of the Al_2O_3/Ti_3SiC_2 multilayer ceramics with SiC_W

Samples	E_A (GPa)	E_T (GPa)	ν_A	ν_T	α_A ($\times10^{-6}$/°C)	α_T ($\times10^{-6}$/°C)	σ_R[a] (MPa)
W0	390.0	321	0.300	0.2	8.800	8.47	−121.2
W10	409.0	321	0.290	0.2	8.370	8.47	37.8
W15	418.5	321	0.285	0.2	8.155	8.47	120.0
W20	428.0	321	0.280	0.2	7.940	8.47	203.1
W25	437.5	321	0.275	0.2	7.725	8.47	287.3
W30	447.0	321	0.270	0.2	7.510	8.47	372.3

[a] The residual compressive stresses in Al_2O_3 layer.

The toughening mechanisms induced by whisker in the multilayer ceramics, namely 2nd-level toughening mechanisms, are similar to the toughening mechanisms in whisker reinforced ceramic composites and have been studied ripely, they include crack deflection mechanism, crack bending mechanism, whisker pull-out mechanism, bridging mechanism, micro-crack mechanism, and so on [1–5]. Fig. 3 shows the microstructure of Al_2O_3 layer in the multilayer ceramics. The whiskers are observed homogeneously distributed and aligned directionally in two directions. The 2D texture of whiskers should be formed in the rolling procedure for green tape preparation, and this was very useful to the whisker toughening mechanisms. Otherwise, the pull-out mechanism selected as a representative one is shown in Fig. 4.

As we know, the toughening effects of the multilayered structural toughening mechanisms depend strongly on the crack propagation path and the mechanical properties of the matrix-layer and interphase-layer [10]. When the SiC whiskers are added into Al_2O_3 layers, the microstructure and mechanical properties of the Al_2O_3 layer is changed at first. The change of mechanical properties of Al_2O_3 layer can influence the location of crack deflection and crack propagated path. In other words, it influences the 1st-level mechanisms. Otherwise, with the addition of SiC whiskers the residual stresses are changed as already analyzed above. From Table 2, the layers carry the residual compressive stress when whiskers are added. It makes the Al_2O_3 layers difficult to fracture, and the crack is enforced

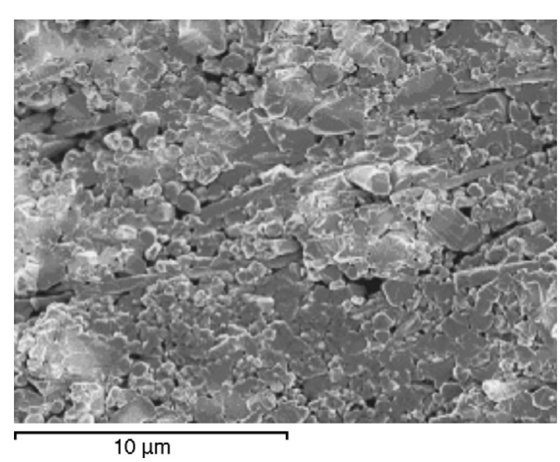

Fig. 3. Fracture surface of the Al_2O_3/Ti_3SiC_2 multilayer ceramics with 20 vol% SiC whiskers.

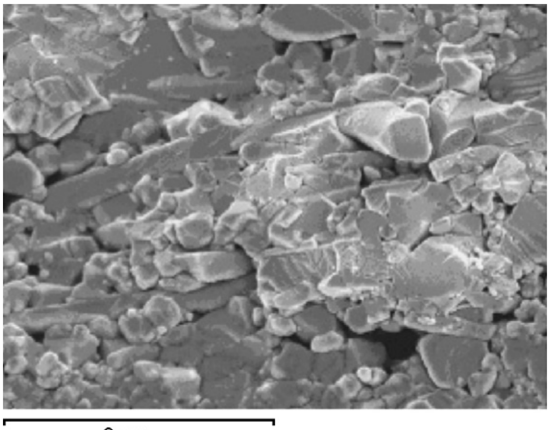

Fig. 4. Pull-out behavior of SiC whisker in Al_2O_3 layers.

to propagating along the interface instead of through Al_2O_3 layer, in other words, the resistance of longitudinal crack is improved and the crack path is prolonged. On the other hand, some whiskers insert adjacent Ti_3SiC_2 layers ineluctably. Some of the whiskers possibly act as a bridge between layers, and they can bridge the transverse crack propagating in the interface, improving the propagating resistance of transverse crack. The changes of the toughening mechanisms described above are different from the individual toughening mechanisms by multilayered structure or whisker toughening. Hence, there is a synergy effect between the 1st-level and 2nd-level toughening mechanisms.

According to the toughness data of the multilayer ceramics shown in Table 1, the greatly improvement of work of fracture reflects the interaction of the whisker toughening mechanisms, multilayer structural toughening mechanisms and their synergy effect.

4. Conclusions

By adding SiC whiskers into Al_2O_3 layers, the mechanical properties of multilayered ceramics is obviously improved, the optimal mechanical properties being obtained for 20 vol% SiC_W addition. Hence, the 2nd-level toughening by whiskers is an effective method to multilayer ceramics.The synergy effect between whisker toughening and multilayer structural toughening was induced in the multilayer ceramics with SiC whiskers, and the improvement of toughness was attributed to the interaction between whisker toughening, multilayer structural toughening and synergy effect.

Acknowledgement

This work was supported by the National Science Foundation of China (NSFC) (Grant no. 59982004).

References

[1] P.F. Becher, C. Hsueh, P. Angelini, T.N. Tiegs, Toughening behavior in whisker-reinforced ceramic matrix composites, J. Am. Ceram. Soc. 71 (1988) 1050–1061.
[2] P.F. Becher, G.C. Wei, Development of SiC-whisker-reinforced ceramics, Am. Ceram. Soc. Bull. 64 (1985) 298–304.
[3] R. Warren, V.K. Sarin, Fracture of whisker-reinforced ceramics, in: K. Friedrich (Ed.), Application of Fracture Mechanics to Composite Materials, Elsevier Science Publisher B.V., 1989, pp. 571–614.
[4] B. Wilshier, F. Carreno, Deformation and failure processes during tensile creep of fiber and whisker reinforced SiC/Al_2O_3 composites, Mater. Sci. Eng. A272 (1999) 38–44.
[5] P.F. Becher, in: K.S. Mazdiyasni (Ed.), Fiber Reinforced Ceramic Composites, Noyes Publication, Park Ridge, NJ, USA, 1990.
[6] A. Goldstein, A. Singurindi, Al_2O_3/TiC based metal cutting tools by microwave sintering followed by hot isostatic pressing, J. Am. Ceram. Soc. 83 (2000) 1530–1532.
[7] A.G. Evans, A.H. Heuer, Transformation toughening in ceramics: martensitic transformations in crack-tip stress fields, J. Am. Ceram. Soc. 63 (1980) 241–248.
[8] T.H.J. Hannink, P.M. Kelly, B.C. Muddle, Transformation toughening in zirconia-containing ceramics, J. Am. Ceram. Soc. 83 (2000) 461–487.
[9] W.J. Clegg, K. Kendall, N.M. Alford, T.W. Button, J.D. Birchall, A simple way to make tough ceramics, Nature 347 (1990) 455–457.
[10] D. Kovar, M.D. Thouless, J.W. Halloran, Crack deflection and propagation in layered silicon nitride/boron nitride ceramics, J. Am. Ceram. Soc. 81 (1998) 1004–1012.
[11] C.A. Wang, Y. Huang, Q.F. Zan, L.H. Zou, S.Y. Cai, Control of composition and structure in laminated silicon nitride/boron nitride composites, J. Am. Ceram. Soc. 85 (2002) 2457–2461.
[12] L. Zhang, V.D. Krstic, High toughness silicon carbide/graphite laminar composite by slip casting, Theo. Appl. Fract. Mech. 24 (1995) 13–19.
[13] B. Hatton, P.S. Nicholson, Design and fracture of layered Al_2O_3/TZ3Y composites produced by electrophoretic deposition, J. Am. Ceram. Soc. 84 (2001) 571–576.
[14] Q.F. Zan, C.A. Wang, Y. Huang, C.W. Li, S.Q. Li, Preparation of Al_2O_3/Ti_3SiC_2 multilayer materials by in-situ synthesis, Key Eng. Mater. 224–226 (2002) 409–412.
[15] W.H. Tuan, J.K. Guo, Toughening ceramics by adding two toughening agents, Key Eng. Mater. 224–226 (2002) 317–320.
[16] Q.F. Zan, C.A. Wang, Y. Huang, S.K. Zhao, C.W. Li, The interface and interface layer in the Al_2O_3/Ti_3SiC_2 multilayer composites by in-situ synthesis, Mater. Lett. 57 (2003) 3826–3832.
[17] M.W. Barsoum, T. El-Raghy, Synthesis and characterization of a remarkable ceramic: Ti_3SiC_2, J. Am. Ceram. Soc. 79 (1996) 1953–1956.
[18] T. El-Raghy, M.W. Barsoum, A. Zavaliangos, S.R. Kalidindi, Processing and mechanical properties of Ti_3SiC_2. II. Effect of grain size and deformation temperature, J. Am. Ceram. Soc. 82 (1999) 2855–2860.