Bull. Mater. Sci., Vol. 30, No. 4, August 2007, pp. 415–419. © Indian Academy of Sciences.

Interfacial microstructure and strength of diffusion brazed joint between Al_2O_3–TiC and 9Cr1MoV steel

WANG JUAN*, LI YAJIANG and S A GERASIMOV[†]
Key Lab of Liquid Structure and Heredity of Materials, School of Materials Science and Engineering, Shandong University (South Campus), Jing Shi Road 73, Jinan 250061, P.R. China
[†]Materials Science Department, Bauman Moscow State Technical University, Moscow 105005, Russia

MS received 5 February 2007; revised 13 July 2007

Abstract. Joining of composite, Al_2O_3–TiC, with heat-resistant 9Cr1MoV steel, was carried out by diffusion brazing technology, using a combination of Ti, Cu and Ti as multi-interlayer. The interfacial strength was measured by shear testing and the result was explained by the fracture morphology. Microstructural characterization of the Al_2O_3–TiC/9Cr1MoV joint was investigated by X-ray diffraction (XRD) and scanning electron microscope (SEM) with energy-dispersion spectroscopy (EDS). The results indicate that a Al_2O_3–TiC/9Cr1MoV joint with a shear strength of 122 MPa can be obtained by controlling heating temperature at 1130°C for 60 min with a pressure of 12 MPa. Multi-interlayer Ti/Cu/Ti was fused fully and diffusion occurred to produce interfacial layer between Al_2O_3–TiC and 9Cr1MoV steel. The total thickness of the interfacial layer is about 100 μm and Ti_3AlC_2, TiC, Cu and Fe_2Ti are found to occur in the interface layer.

Keywords. Al_2O_3–TiC; diffusion brazing; interfacial strength; microstructural characterization.

1. Introduction

Ceramic materials are promising candidates for high temperature structural and wear-resistant components such as cutting tools, seal rings, dies for drawing or extrusion and a variety of high temperature engine parts (Jahanmir *et al* 1989; Richerson 1992; Singh *et al* 2003). However, the low fracture toughness and the variability of their mechanical strength inhibit widespread use of these materials. However, considerable improvement in mechanical properties of the single-phase ceramic materials has been achieved by incorporating one or more components into the base material to form ceramic–matrix composites (Becher and Wei 1991; Steinbrech 1992). The composite, Al_2O_3–TiC, has been widely used in industries as cutting tools and wear resistance coating because of its high hardness, chemical stability, good strength and toughness at elevated temperatures and excellent wear resistance (Kong *et al* 1995; Deng *et al* 2005; Dobrzanski and Mikula 2005).

In some engineering applications, it is necessary for Al_2O_3–TiC to be joined with metal in order to decrease the brittleness. The development of techniques to join ceramic to metal makes possible this combination. Among these techniques, brazing and diffusion bonding are most suitable (Blugan *et al* 2004; Liu *et al* 2004; Wang *et al* 2006). Both processes are well known as far as ceramic/metal joining is concerned.

As for diffusion bonding, adhesion is attributed to two main mechanisms: (i) plastic deformation of the surface aspects and (ii) voids closure by diffusive mass transport. Once bonding is obtained, the strength of the joint is primarily dependent on the residual stress at the interface. When ceramics are involved, this stress is originated from the thermal expansion mismatch between the ceramic and the metallic substrates that occurs during cooling from the bonding temperature. By the use of a ductile and soft metallic interlayer and even multi-interlayer between ceramic and metal, the diffusion bonding process can be made easier. The method is sometimes called diffusion brazing.

The present work deals with the diffusion brazing of ceramic–matrix composite, Al_2O_3–TiC, with a heat resistant 9Cr1MoV steel using a combination of Ti, Cu, Ti as interlayer materials for the first time. The strength of the joint was measured by shear testing and the results were correlated with fracture morphology. Interface characterization was carried out by scanning electron microscope (SEM) with energy-dispersion spectroscopy (EDS). Finally, new phases formed in the joint were examined by X-ray diffraction (XRD).

2. Experimental

Cylindrical samples ($\Phi 55 \times 4$ mm) of hot pressure sintered Al_2O_3–TiC composite and heat resistant 9Cr1MoV steel of the same dimensions were diffusion bonded using

*Author for correspondence (jwang@sdu.edu.cn)

Ti/Cu/Ti multi-interlayer. The total thickness of the Ti/Cu/Ti multi-interlayer was 60 μm. Al_2O_3–TiC had a composition in wt.% of 43·8 Al_2O_3, 40·6 TiC, 8·9 molybdenum, 2·4 nickel, 3·8 chromium and 0·5 vanadium. 9Cr1MoV had a composition in wt.% of 8·7 chromium, 1·2 molybdenum, 0·5 vanadium and the rest iron.

Prior to bonding, substrates and interlayer were polished using a 0·25 μm diamond paste and cleaned by immersion in acetone with ultrasonic agitation. Diffusion brazing experiments were carried out in the temperature range 1080–1160°C for 60 min at each temperature under a vacuum of about 10^{-4} Pa. A pressure of 12 MPa was applied through a hydraulic ram inside the vacuum chamber.

The interfacial strength was measured by mechanical (shear) testing using a special device. The nominal strain rate was 5×10^{-3} min^{-1}. The shear strength was calculated by the load at the fracture divided by the nominal area of the joint. For metallographic examination, the bonded specimens were cut transversely through the bond and mounted in bakelite. The Al_2O_3–TiC/9Cr1MoV joints were etched with a solution consisting of 95% nitric acid and 5% alcohol.

Metallographic observations were performed with both light and scanning electron microscopies (SEM) and the chemical compositions across the joint were determined by energy-disperse spectrometry. Further, X-ray diffraction analysis was conducted using CuK_α radiation in order to obtain diffraction profiles across the bonds and to analyse the new phases in the Al_2O_3–TiC/9Cr1MoV joint.

3. Results and discussion

3.1 *Shear strength*

Figure 1a shows interfacial shear strength at the Al_2O_3–TiC/9Cr1MoV joint at various heating temperatures. The shear strength increases from 91–122 MPa with increase in heating temperature from 1080–1130°C. This is attributed to increased diffusion of Ti and Cu from interlayer into base metal to relieve the stress concentration in the interface. Above 1130°C, however, the interfacial shear strength decreases. This is because the microstructure becomes coarser when the temperature is increased. So, by controlling the heating temperature at 1130°C, an Al_2O_3–TiC/9Cr1MoV joint with a shear strength of 122 MPa can be obtained. The stress–strain curve of shearing joint obtained at a heating temperature of 1130°C is shown in figure 1b. In the first stage, stress and strain have linear relationship and stress increases quickly with strain from 0–0·4%. In the second stage, the stress increases slowly with strain above 0·4% and it is 122 MPa when strain is 2·2%. At this time, the Al_2O_3–TiC/9Cr1MoV joint was fractured. Compared with Al_2O_3–TiC composite, the toughness in Al_2O_3–TiC/9Cr1MoV joint increased greatly by the addition of Ti and Cu interlayers.

Figure 2a shows the fracture morphology at the shearing interface. All the fracture surfaces with different heating temperatures exhibit characteristics of light brittle rupture. EDS analysis has revealed that the fractured faces contain more of Al, Ti and O and less of Cr and Fe, as shown in figure 2b. This indicates that the shear fracture occurs in the joint near Al_2O_3–TiC side. Therefore, the joint near to Al_2O_3–TiC is the key to determine the quality of Al_2O_3–TiC/9Cr1MoV joint.

3.2 *Interface characterization*

Figure 3 shows the nature of the interface in the diffusion bonded Al_2O_3–TiC/9Cr1MoV joint. Ti/Cu/Ti multi-interlayer was fused fully and diffusion occurred to produce a continuous interfacial layer between Al_2O_3–TiC composite and 9Cr1MoV steel. The thickness of the interfacial layer

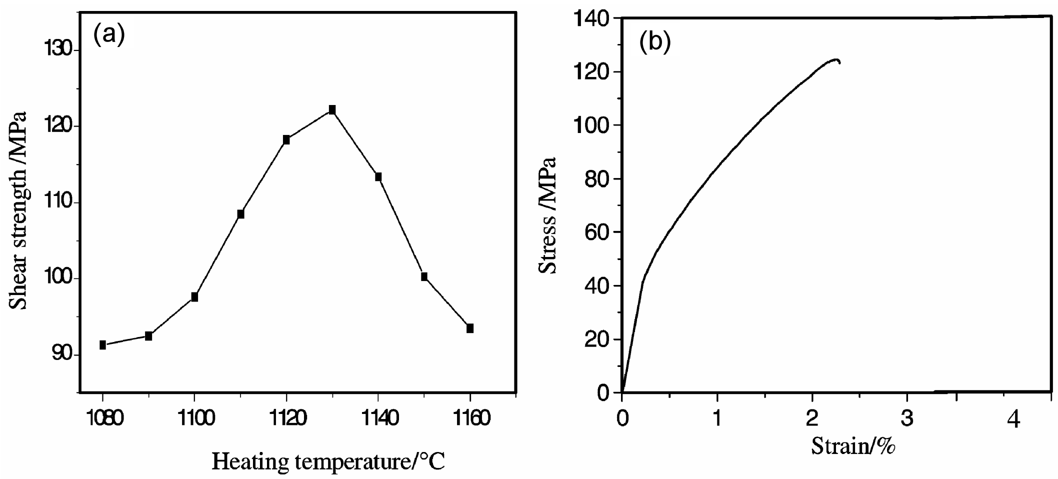

Figure 1. Interfacial strength at various heating temperatures and stress–strain curve: (**a**) strength and (**b**) stress–strain curve.

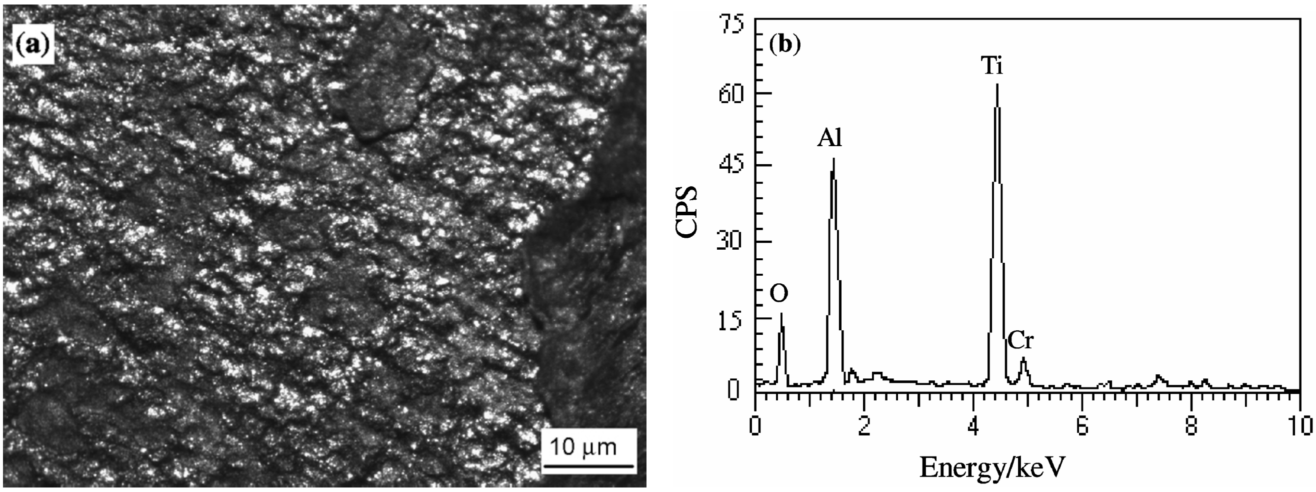

Figure 2. Fracture characterization at the shearing interface: (**a**) morphology and (**b**) EDS on fracture face.

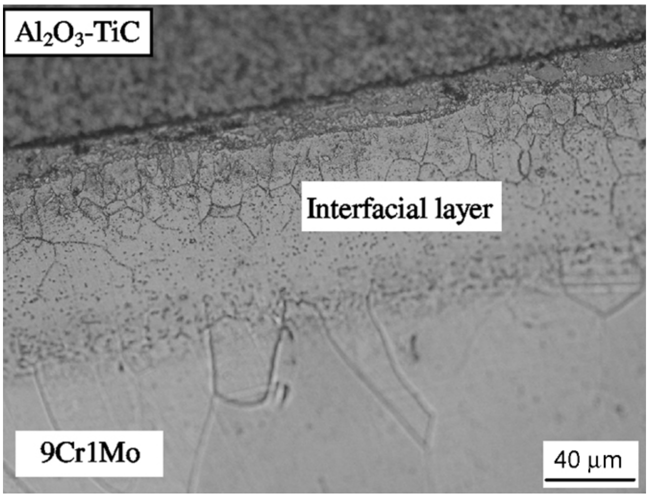

Figure 3. Microstructure characteristics in the diffusion bonded Al_2O_3–TiC/9Cr1MoV joint.

is about 100 μm measured by electron probe microanalysis (EPMA). The grain size in the interfacial layer is less than that in 9Cr1MoV steel. This is attributed to the fusion of Ti/Cu/Ti multi-interlayer during bonding and subsequent cooling.

The backscattered electron image of the interfacial layer is shown in figure 4. The microstructure of the interfacial layer exhibits a distribution of black particles. In order to clarify the microstructure characteristics in the layer, the composition at various positions in the interface, labeled as A, B, C, D and E, was determined by EDS. The results are shown in figure 4.

Near 9Cr1MoV side of the interface, the microstructure labeled A is rich in Ti, C and contains some Fe. The element distribution indicates that the phase A may be TiC with some Fe_3C. TiC is formed by a combination of Ti from interlayer with C from 9Cr1MoV steel. The microstructure labeled B is the matrix of the interfacial layer and the EDS shows that they contain Fe and Ti. The Ti is from interlayer and Fe is the diffusion from 9Cr1MoV. So the matrix of the interfacial layer is evaluated to be an Fe–Ti compound or α-Ti phase. In the black microstructure labeled C, Cu element is almost up to 90%. This is attributed to the fusion of Cu interlayer during diffusion brazing and it forms a solid solution during cooling. The microstructure labeled D is rich in Ti, C with some Fe. This indicates that it is a TiC phase. Near Al_2O_3–TiC side, there are Ti, Al and C in the microstructure labeled E. Therefore, some compounds with Ti, Al and C may have formed in this region.

3.3 *Phase composition*

X-ray diffraction analysis was carried out using CuK_α radiation on profiles across the bonds in order to analyse the phases in the Al_2O_3–TiC/9Cr1MoV joint. The XRD pattern of the joint is shown in figure 5. Figure 5a represents the joint near Al_2O_3–TiC side and figure 5b represents the joint near 9Cr1MoV side.

There are Ti_3AlC_2 and TiC due to the diffusion reaction of Al and C from composite, TiC–Al_2O_3, with Ti interlayer on Al_2O_3–TiC side at the interface. TiC is identified from the JCPDS card No. 32-1383. Ti_3AlC_2 is identified by the data from Nikolay (Nikolay and Barsoum 2000). On the 9Cr1MoV side of the interface, the phases Fe_2Ti and TiC were formed. Fe_2Ti is identified by JCPDS card No. 15-0336. This is because the Ti interlayer fused during diffusion brazing to form Fe_2Ti and TiC with Fe and C, which are from 9Cr1MoV steel. In addition, the single Cu phase was found. This indicates that Cu interlayer fused during brazing without interacting with any species.

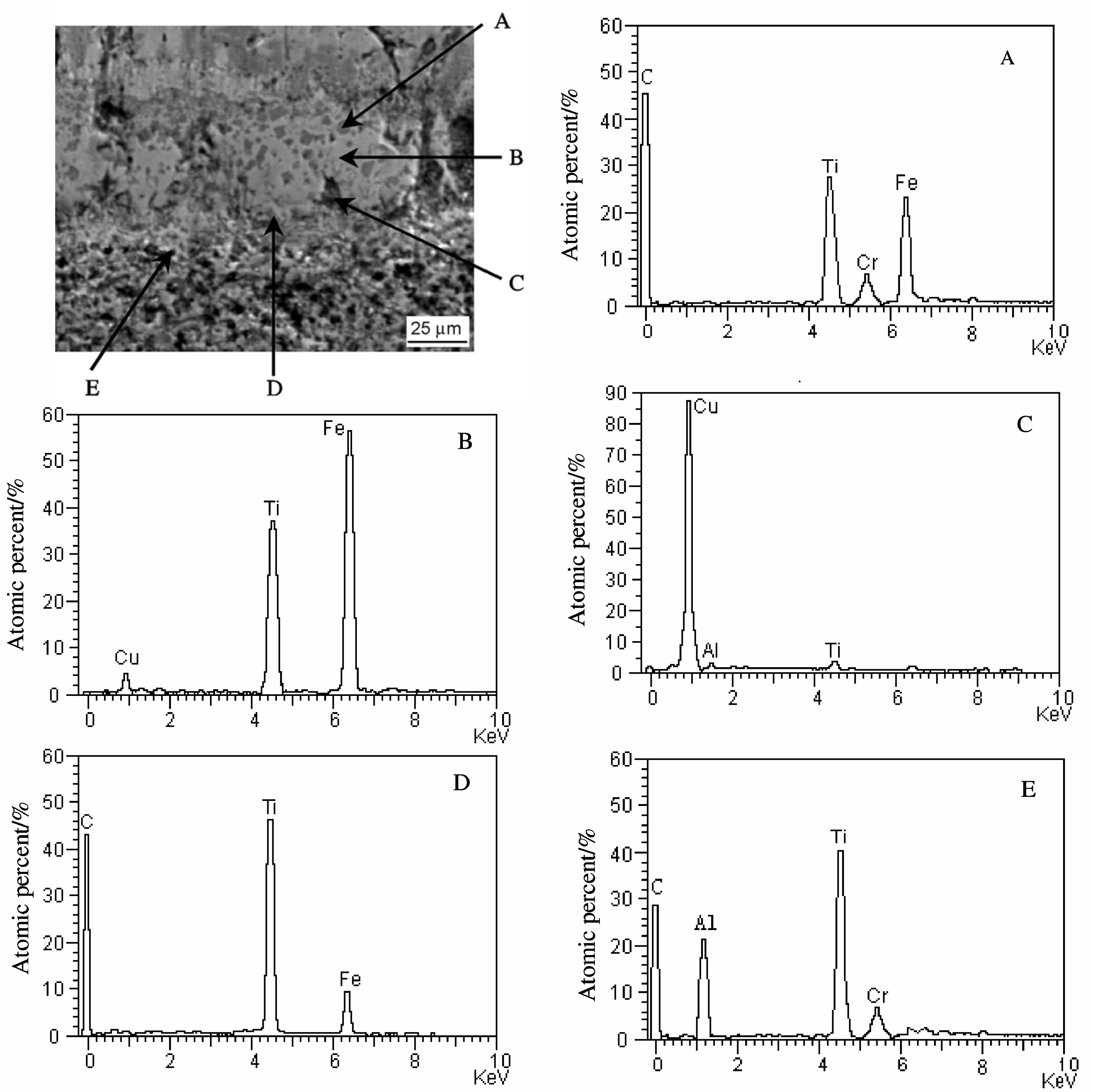

Figure 4. The backscattered electron image and EDS analysis on the interface layer.

Interlayer Cu plays an important role to promote the diffusion of Fe, C and Ti from substrates including TiC–Al_2O_3.

The X-ray diffraction evidence of the existence of Ti_3AlC_2, Fe_2Ti, Cu and TiC in the interfacial layer correlates well with the microstructural characteristics and with their compositions shown in figure 4. Correspondingly, the phases from TiC–Al_2O_3 composite to 9Cr1MoV steel across the interfacial layer exhibit a transition from Ti_3AlC_2 + TiC, Cu + Fe_2Ti, to TiC. This is the result of the diffusion of Al, Ti, Cu and C present in TiC–Al_2O_3, 9Cr1MoV steel and multi-interlayer.

4. Conclusions

(I) Controlling the heating temperature at 1130°C, a Al_2O_3–TiC/9Cr1MoV joint with a shear strength of 122 MPa can be obtained by diffusion brazing technology, using a combination of Ti, Cu and Ti as multi-interlayer.

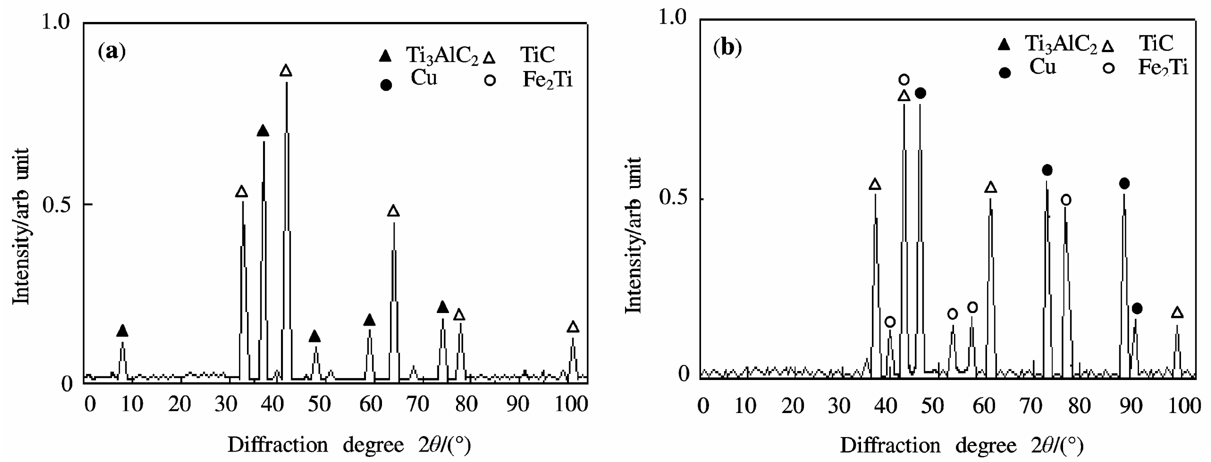

Figure 5. X-ray diffraction patterns: (**a**) near Al_2O_3–TiC and (**b**) near 9Cr1MoV.

The fracture morphology has a characteristic of light brittle rupture and EDS analysis indicates that the fracture during shearing occurs on Al_2O_3–TiC side of the interface.

(II) Ti/Cu/Ti multi-interlayer was fused fully during diffusion brazing and produced a continuous interfacial layer between the Al_2O_3–TiC composite and 9Cr1MoV steel. The diffusion reaction thickness of the interfacial layer is about 100 μm.

(III) The microstructure in the interfacial layer, EDS results and XRD analysis show that the new phases formed across the interfacial layer exhibit a transition from Ti_3AlC_2 + TiC, Cu + Fe_2Ti, to TiC.

Acknowledgements

This project was supported by the Doctoral Foundation of Shandong Province (2006BS04004), National Natural Science Foundation of China (50375088) and Key Lab of Liquid Structure and Heredity of Materials.

References

Becher P F and Wei G C 1991 *J. Am. Ceram. Soc.* **74** 255

Blugan G, Janczak R J and Kuebler J 2004 *Acta Mater.* **52** 4579

Deng J X, Can T K and Sun J L 2005 *Ceram. Int.* **31** 249

Dobrzanski L A and Mikula J 2005 *J. Mater. Process. Technol.* **164/165** 822

Jahanmir S, Deckmap D E, Ives L K and Feldman A 1989 *Mater. Res. Soc. Symp. Proc.* 285

Kong J B, Enoki M, Kishi T and Oh H K 1995 *Compos. Eng.* **5** 1275

Liu H J, Feng J C, Fujii H and Nogi K 2004 *Mater. Sci. Technol.* **20** 1069

Nikolay V T and Barsoum M W 2000 *J. Am. Ceram. Soc.* **83** 825

Richerson D W 1992 *Modern ceramic engineering* (New York: Marcel Dekker)

Singh J, Soni N C and Srivastava S L 2003 *Bull. Mater. Sci.* **26** 397

Steinbrech R W 1992 *J. Eur. Ceram. Soc.* **10** 131

Wang J, Li Y J and Yin Y S 2006 *Bull. Mater. Sci.* **29** 155