Fabrication and superplasticity of Al_2O_3/3Y-TZP laminated composite

Kaifeng Zhang*, Guofeng Wang, Zhenjie Wang, Changwen Wang, Wenbo Han

School of Material Science and Technology, Harbin Institute of Technology, Harbin, China

Abstract

Tape casting and hot-press sintering are used to fabricate an Al_2O_3/3Y-TZP laminated composite. The as-prepared material is deep drawn at high temperature to research its superplastic formability. It is found that the microstructure of the material sintered at 1550 °C is fine and no significant residual porosity was detected from SEM observations at the interfaces between the two types of layers. The superplastic forming experiment shows that, when the strain rate is constant, temperature has a great influence on the superplasticity of the Al_2O_3/3Y-TZP laminated composite. A hat-like part with the greatest deform height is obtained at 1500 °C. The processing will be less effective at higher or lower temperature.
© 2005 Elsevier Ltd. All rights reserved.

Keywords: Tape casting; Hot pressing; Laminated composite; Al_2O_3; TZP

1. Introduction

Since 1990, ceramic/ceramic laminated composites have emerged as promising candidates to overcome the inherent brittleness of ceramics for use in structural applications.[1–4] Al_2O_3/TZP laminated composites have been receiving growing attention, mainly due to their oxidation stability at high temperature and to the drastic increases in strength and especially in fracture toughness at room temperature because of the various crack-shielding phenomena related to the presence of the layers.[5,6]

Tomaszewski[7] fabricated Y-TZP/Al_2O_3 laminated composites and researched the effect of residual stress on the character of crack propagation. Chartier and Rouxel[8] made a significant improvement of both the fracture resistance and the toughness, from 380 to 560 MPa and from 3.7 to 8 MPa m$^{1/2}$, respectively, between the pressed alumina monolith and the tape cast Al_2O_3/ZrO_2 laminated composites.

Because of the great advantage of laminated composites, the plastic forming technology becomes very important.

Flacher and Blandin[9] found superplastic compressive properties in an Al_2O_3/ZrO_2 laminated composite at 1470 °C. Manuel and Clauss[10] examined the high temperature plastic deformation of an Al_2O_3/Al_2O_3-YTZP laminated composite in uniaxial compression testing. The composite exhibited creep properties at low strain rate. The fine grain sizes and the good interfacial adhesion of the layers impart creep resistance and ductility simultaneously to the laminates.[11] However, these studies are limited to compressive deformation. In the present work, superplastic deep drawing of Al_2O_3/3Y-TZP laminated composites is investigated in consideration of the importance of plastic deformation under tensile stress for engineering applications.

2. Experimental

An α-Al_2O_3 powder and a 3Y-TZP powder, with particle sizes of 50–200 nm and 15–40 nm, were used. TEM morphologies of the powders are shown in Fig. 1. It can be seen that the powders have no hard aggregate.

The tapes of Al_2O_3 and 3Y-TZP were prepared by tape casting, the details of which can be seen in the paper of Cui

---

* Corresponding author.
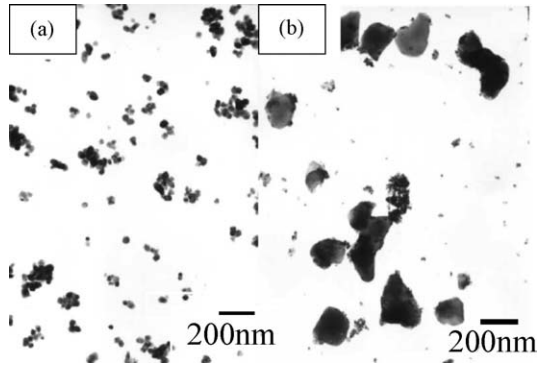*E-mail address:* kfzhang@hit.edu.cn (K. Zhang).

Fig. 1. TEM morphologies of the powders (a) 3Y-TZP; (b) Al_2O_3.

Fig. 2. Simultaneous TG-DSC analysis on green tape, up to 700 °C.

et al.[12] The dried tape was punched into ϕ 30 mm discs. The Al_2O_3 circles and 3Y-TZP discs were stacked in an alternation sequence to a total of 41 layers, with both of the outer surface layers being Al_2O_3.

The discs were placed in a low-temperature furnace at 800 °C for 2 h for binder burnout. Cooling was controlled at 3 °C/min.

The laminates were uniaxial pressure sintered in a cylindrical graphite die with diameter of 30 mm. The heating rate was 20 °C/min and the pressure was 25 MPa. The laminates were heated to the required temperature and held for 90 min. The thickness of all discs was 2 mm. Prior to the forming tests the samples were polished on one side (diamond paste 0.3 μm). The discs were placed on a cylindrical graphite ring with an inner diameter of 20 mm. A hemispherical graphite punch with radius of 8 mm was pushed on the non-polished side of the laminate using a velocity of 0.2 mm/min while the force was monitored. The tests were performed in vacuum at 1400, 1500 and 1600 °C. The axial displacement was measured externally during all tests. Microstructure examination of samples after hot pressing and superplastic forming was carried out using a scanning electron microscope (SEM, Philips XL 20) equipped with an image analyzer.

3. Results and discussion

3.1. Hot-pressing sintering

During the preparation procedure of the tapes, a lot of organic addition was used. This must be burned out completely before sintering. Otherwise, it will do harm to the properties of the laminated composite. The tapes were analyzed with TG-DSC and the results are shown in Fig. 2. It can be seen that the pyrolytic decomposition happened mainly between 200 and 400 °C and almost all of the organic addition was burned out when the temperature reached 700 °C.

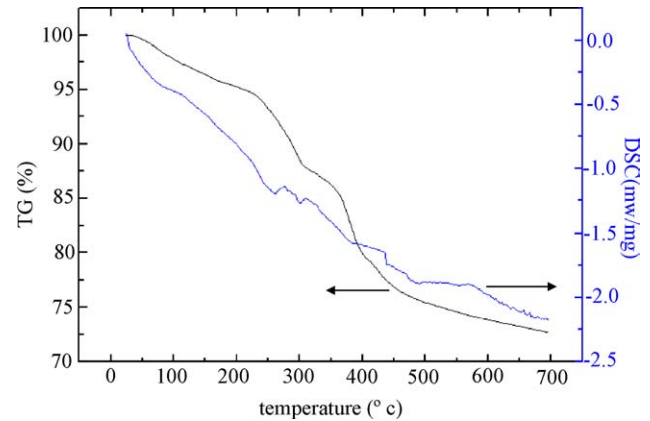Fig. 3 shows the cross section of the as-prepared laminated composite. The dark zones correspond to Al_2O_3 layers. The two types of layers are well defined with interfaces and no significant residual porosity was detected from SEM observations at the interfaces between the two types of layers. The thickness of each layer is about 90 μm. However, it must be mentioned that the interfaces are not so straight because the material was pressed during the sintering. This will advantage the strength of the interfaces.

In order to research the influence of temperature on the microstructure of laminated composite, different sintering temperatures, 1450, 1550 and 1650 °C, were chosen. Fig. 4 shows the cross section of the laminated composite sintered at different temperatures.

The grain size of the laminated composite sintered at 1450 °C is very fine. Mean grain sizes were determined to be 150 and 400 nm, respectively for the 3Y-TZP and Al_2O_3 layers. However, there are many cavities in the Al_2O_3 layer. The composite sintered at 1550 °C has a larger grain size. Mean grain sizes are 200 and 800 nm, respectively for the 3Y-TZP and Al_2O_3 layers. The relative density has great improvement. There is little porosity existing in the Al_2O_3 layer. When the temperature reaches 1650 °C, grains grow rapidly. They are 1 and 10 μm, respectively for the 3Y-TZP and Al_2O_3 layers (Fig. 4).
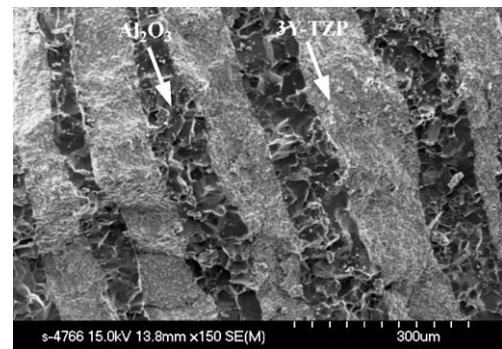

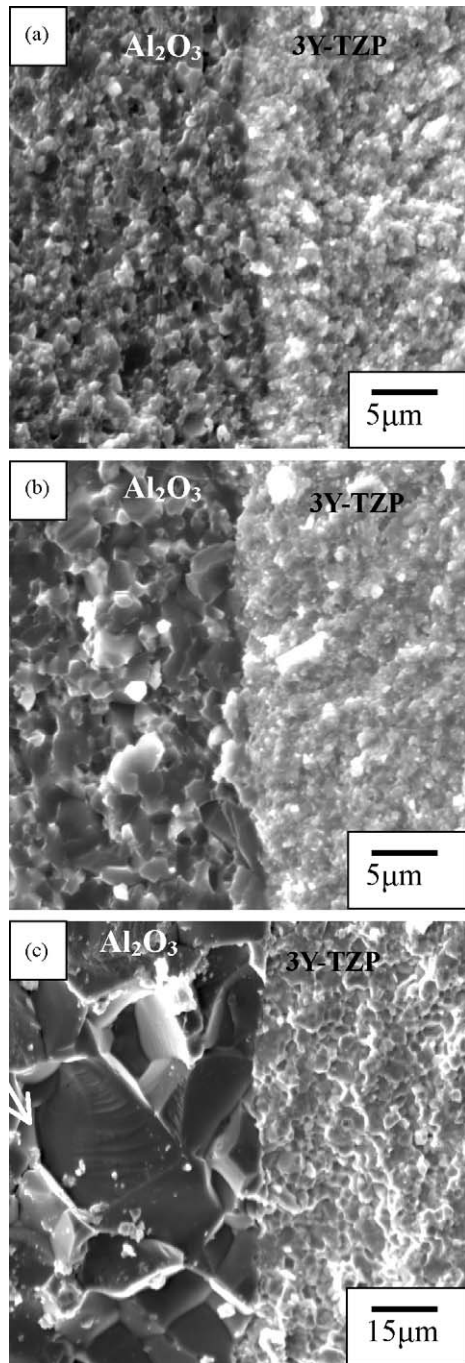Fig. 3. Morphologies of the cross section of laminated composite after sintering.
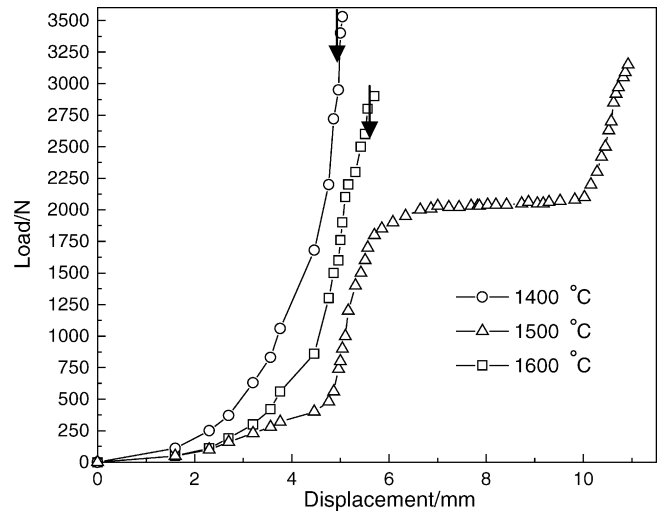
Fig. 5. Displacement vs. applied force during deep drawing at different deformation temperatures.

At 1500 °C the laminated composite can be stretched to a height of 11.1 mm without fracture. From Fig. 5 it is also clear that higher and lower forming temperature require more force for identical displacements.

This influence of deformation temperature on deformation can be explained in terms of the change of microstructures, which are shown in Fig. 6. A slightly grain growth occurs when the specimen was deep drawn at 1400 °C. Mean grain sizes are 3 μm and 500 nm for Al_2O_3 and 3Y-TZP, respectively, as shown in Fig. 6a. However, many cavities appear in the Al_2O_3 layer because of its bad deformability at lower temperature. It is widely accepted that cavity nucleation arises from stress concentrations generated at second phase particles or triple points as a result of grain boundary sliding. At a certain concentration and/or size the cavities are interlinking which is associated with the onset of tensile failure. Because of the stronger cavity growth at lower forming temperature, higher stresses are present in the ceramics. This explains the higher forces necessary for the same displacement at lower forming temperature.

Grains grow rapidly when deep drawing at 1600 °C. Mean grain sizes are 8 and 1 μm for Al_2O_3 and 3Y-TZP, respectively, as shown in Fig. 6c.

The grain-size effect on superplasticity has been often observed in the literature and can be explained by the fact that grain boundary sliding is the dominant deformation mode. High-temperature deformation behavior in fine-grained ceramics is usually described by:[13]

$$\dot{\varepsilon} = A \frac{\sigma^n}{d^p} \exp\left(-\frac{Q}{RT}\right) \tag{1}$$

where A is a numerical constant, σ the flow stress and n its exponent, d the grain size with exponent p, Q the apparent activation energy and T is the temperature.

Generally, superplastic deformation occurs in ceramics due to grain boundary sliding. Accompanying the grain

Fig. 4. SEM morphologies of the cross section of laminated composites after sintering at different temperatures: (a) 1450 °C; (b) 1550 °C; (c) 1650 °C.

3.2. Superplastic deep drawing

In Fig. 5 the displacement of the punch and the resulting force on the sample are given during deep drawing at 1400, 1500 and 1600 °C using the laminated composite sintered at 1550 °C. Here the influence of deep-drawing temperature is clearly visible. If a higher or lower forming temperature is used, the chance of fracturing during deep drawing increases (fracturing is indicated in the figure by an arrow downwards).
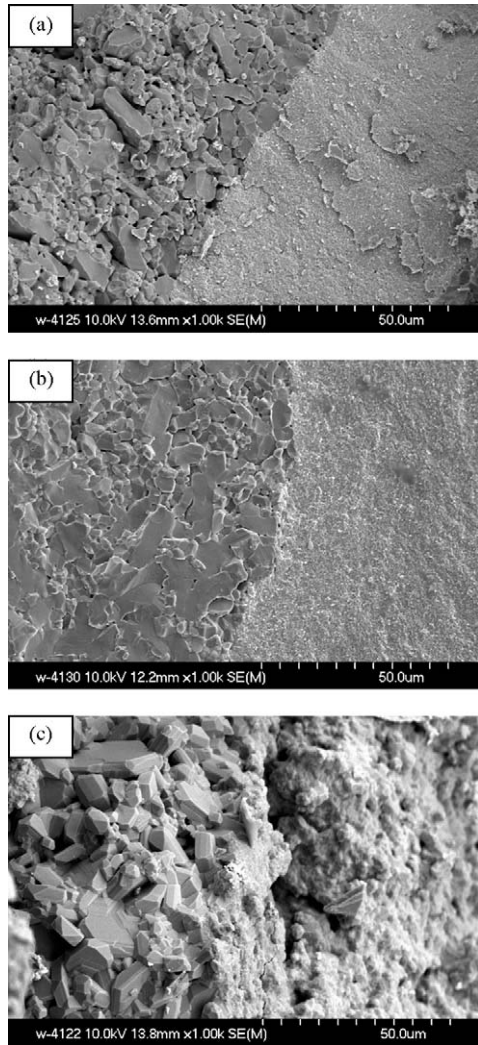
Fig. 6. SEM morphologies of cross section of laminated composites after superplastic forming at different temperatures: (a) 1400 °C; (b) 1500 °C; (c) 1600 °C.

boundary sliding the general understanding is that accommodation involving diffusion and/or dislocation movement is required to maintain grain boundary cohesion. If accommodation is incomplete or slower than the grain displacement rate then cavities will occur. The effect of grain growth is to make the accommodation process more difficult by increasing the unit path length for the operative mechanism.

Grain sizes are fine and few cavities appear after deep drawing at 1500 °C. So, the best superplastic deep-drawing temperature for the Al_2O_3/3Y-TZP laminated composite is 1500 °C.

An Al_2O_3/3Y-TZP laminated composite part has been formed by superplastic deep drawing at 1500 °C; the part and its cross section are shown in Fig. 7. The diameter of the flange reduces from 30 to 25 mm and the mean thickness reduces from 2 to 1.7 mm. This shows that the shape of hemisphere is achieved by the shrinkage of the diameter and the reduction of thickness.
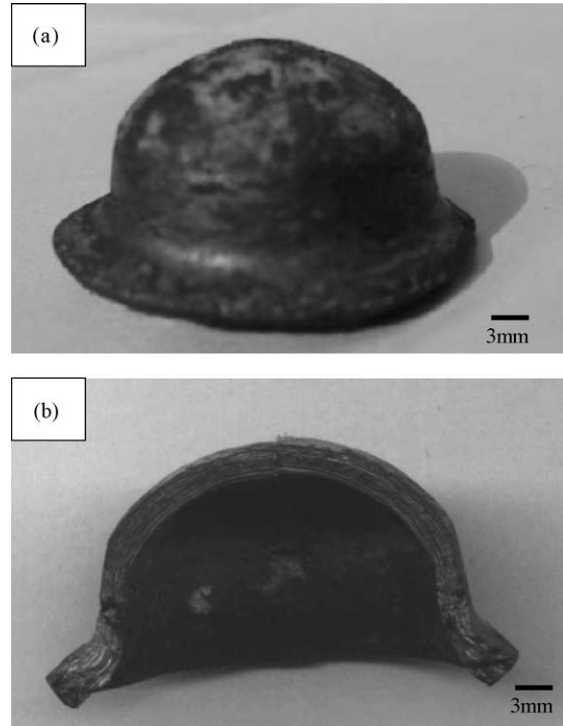

Fig. 7. Al_2O_3/3Y-TZP laminated composite part formed by superplastic deep drawing: (a) the part; (b) the cross section.

It is also shown that the 3Y-TZP layers, which have good superplastic forming ability, help the Al_2O_3 layers, which have poor superplastic forming ability, when the laminated composite is deep drawn at a suitable temperature. However, if we want to improve further the superplastic forming ability of the Al_2O_3/3Y-TZP laminated composite, grain growth of Al_2O_3 must be controlled. Park et al.[14] reported that the addition of SiO_2 or ZrO_2 can effectively inhibited the growth of Al_2O_3. This will be researched in the future.

4. Concluding remarks

It is shown that tape casting and hot pressing can be applied to the fabrication of an Al_2O_3/3Y-TZP laminated composite with fine grain size and crack-free microstructure at 1550 °C. The as-prepared laminated composite has good superplasticity. At 1500 °C it can be deformed to form a hemisphere without fracture. However, the superplastic forming ability will decreases rapidly at higher or lower temperature. If we want to improve further the superplastic forming ability of an Al_2O_3/3Y-TZP laminated composite, grain growth of Al_2O_3 must be controlled.

Acknowledgements

References

1. Clegg, W. J. *et al.*, A simple way to toughen ceramic. *Nature*, 1990, **347**(4), 455–461.
2. Zhang, L. and Krstic, V. D., High toughness silicon carbide/graphite laminar composite by slip casting. *Theor. Appl. Fract. Mech.*, 1995, **24**(1), 13–19.
3. Wang, C., Huang, Y. and Zan, Q. F., Biomimetic structure design—a possible approach to change the brittleness of ceramics in nature. *Mater. Sci. Eng.*, 2000, **11**(1), 9–12.
4. Ohji, T., Shigegaki, Y., Kondo, N. *et al.*, Fracture toughness of multilayer silicon nitride with crack deflection. *Mater. Lett.*, 1999, **40**(6), 280–284.
5. She, J. and Inoue, T., Damage resistance and *R*-curve behavior of multilayer Al₂O₃/SiC ceramics. *Ceram. Int.*, 2000, **26**(8), 801–805.
6. Chen, I. W. and Xue, L. A., Development of superplastic structural ceramics. *J. Am. Ceram. Soc.*, 1990, **73**, 2585–2609.
7. Tomaszewski, H., Residual stresses in layered ceramic composites. *J. Eur. Ceram. Soc.*, 1999, **19**(6–7), 1329–1331.
8. Chartier, T. and Rouxel, T., Tape-cast alumina–zirconia laminates: processing and mechanical properties. *J. Eur. Ceram. Soc.*, 1997, **17**, 299–308.
9. Flacher, O. and Blandin, J. J., Microstructural aspects of superplastic deformation of Al₂O₃/ZrO₂ laminate composites. *Mater. Sci. Eng. A*, 1996, **219**, 148–155.
10. Manuel, J. M. and Clauss, C., Microstructure and high-temperature mechanical behavior of alumina/alumina-yttria-stabilized tetragonal zirconia multilayer composites. *J. Am. Ceram. Soc.*, 1997, **80**(8), 2126–2130.
11. Manuel, J. M. and Gutierrez Mora, F., Effect of layer interfaces on the high-temperature mechanical properties of alumina/zirconia laminate composites. *Acta Mater.*, 2000, **48**, 4715–4720.
12. Cui, X. M., Ouyang, S. X. *et al.*, A study on green tapes for LOM with water based tape casting processing. *Mater. Lett.*, 2003, **57**, 1300–1304.
13. Langdon, T. G., Superplastic forming of structural alloys. *Metall. Soc. AIME*, 1982, 27–40.
14. Park, S. Y., Saruhan, B. and Schneider, H., Mullite/zirconia laminate composites for high temperature application. *J. Eur. Ceram. Soc.*, 2000, **20**, 2463–2468.