Journal of Ceramic Processing Research. Vol. 9, No. 2, pp. 131~134 (2008)

JOURNAL OF
Ceramic Processing Research

Preparation and characterization of Y-TZP powders coated with alumina

Chih-Cheng Chen[a], Hsing-I Hsiang[b,]* and Shu-Wei Hsu[b]
[a]*Department of Mechanical Engineering, Far East College, Tainan, Taiwan, R.O.C.*
[b]*Department of Resources Engineering, National Cheng Kung University, Tainan, Taiwan, R.O.C.*

In this study, alumina was coated onto yttria tetragonally stabilized zirconia (Y-TZP)core powder using aluminum nitrate with urea in an aqueous solution. Boehmite coating onto the surface of Y-TZP powders was confirmed using transmission electron microscopy (TEM), energy dispersive spectroscopy (EDS), Fourier transform infrared (FTIR) spectroscopy and zeta potential measurements. The sintering behaviors of Y-TZP powders with and without an alumina coating were investigated using a dilatometer and scanning electron microscopy (SEM). Boehmite coating onto the surface of Y-TZP powders significantly improved the densification of the resulting ceramic bodies. The relative density of the Y-TZP with a boehmite coating sintered at 1350°C can reach 97.3%.

Key words: Y-TZP, coating, boehmite

Introduction

Yttria tetragonally stabilized zirconia (Y-TZP) is of great interest due to its high strength, high toughness, chemical stability, high melting temperature, ionic, electrical and optical properties in structural ceramics [1]. Y-TZP applications are widespread, such as wear-resistant components and optical fiber connectors, like ferrules. The ferrule materials should satisfy a set of conditions, among which the most important are stability during environmental changes, thermal expansion coefficient and hardness close that of SiO_2 glass, low Young's modulus (easy deformation), and a good-quality polished surface. Y-TZP has been the material of choice for ferrules because it satisfied all of the above requirements [2]. Y-TZP is stable up to 2370°C, but aging at temperatures between 100-300°C in the presence of water could produce severe property degradation [3].

The densification and degradation resistance of Y-TZP can be improved by doping with trace quantities of alumina. However, it is difficult to solve the heterogeneity of alumina trace quantities mixed in the Y-TZP matrix due to the strong agglomeration of the dry nanopowders [4]. The uniformity of the incorporation of a minor constituent into the matrix can be ameliorated using coating processing, such as colloidal processing, a sol-gel route, solution coating, or precipitating coating [5-8]. Mei et al.[9] developed a controlled aqueous heterogeneous precipitation method by which a thin layer of boehmite was deposited onto the surface of cordierite-glass particles by the slow release of OH- from the hydrolysis reaction of urea. In this study, boehmite was coated onto Y-TZP core powder using the aluminum nitrate reaction with urea in an aqueous solution. Boehmite coating onto the surface of Y-TZP powders was confirmed using TEM, FTIR spectroscopy and zeta potential measurements. The sintering behaviors of Y-TZP powders with and without an alumina coating were investigated using a dilatometer and scanning electron microscopy (SEM).

Experimental procedure

A commercially available 3-mole%-yttria-stabilized tetragonal zirconia (Y-TZP) (TP-3Y, Teamcera, Taiwan) with a specific surface area of 16 ± 2 m^2/g was used in this study. The addition of alumina into Y-TZP powders was 0.25 wt% which is the calculated alumina content for converting boehmite to alumina after sintering. An aqueous dispersion containing 20 g of Y-TZP powder in 200 ml of deionized water was agitated ultrasonically for 30 minutes followed by the addition of 0.3688 g of $Al(NO_3)_3 \cdot 9H_2O$ and 0.8 g urea. The pH of the starting mixture was about 2.9. The mixed solution was mixed and heated slowly from 30°C to 90¢J. The mixture, with a final pH value of 6.0±0.2 was kept at 90°C for 5 h, then dried at 100°C. The crystalline phases and chemical composition of the specimens were characterized using an X-ray diffractometer with Cu-K_α (Siemens, D5000) and an FTIR spectrometer (Bruker, EQUINOX 55), respectively. The boehmite coating onto the Y-TZP powder surfaces was observed using TEM (Jeol, JEM 2010) and EDS (Noran, Voyager 2.0).

Electrophoretic measurements of coated and uncoated powders under different pH values were performed on

*Corresponding author:
Tel : +886 6 2757575x62821
Fax: +886 6 2380421
E-mail: hsingi@mail.ncku.edu.tw

a zeta potentiometer (Malvern, Zetasizer Nano ZS). The disk-shaped Y-TZP was formed by die-pressing at 80MPa and then cold isostatic pressed at 200MPa. The green compacts were sintered in air at 1350°C for 2 h, and then furnace cooled. After it was sintered, the disk was polished using diamond paste and then annealed for 30 minutes at 1200°C. The apparent density was measured using the Archimedes method. The relative density was calculated using 6.079 g/cm^3 as the theoretical density. The microstructures of the thermally etched cross-sections were investigated by SEM (Hitachi, S-4100). The sintering studies were performed by a dilatometer (Netzsch, DIL 402C) in which the samples were heated at a constant rate of 10 K minute^{-1} to 1500°C.

Results and discussion

The FTIR spectra of Y-TZP powders before and after coating with boehmite are shown in Fig. 1. Comparing the spectra of uncoated and boehmite coated Y-TZP powders, new absorption bands at 3090 and 1040 cm^1 were observed in the Y-TZP powders coated with boehmite. The absorption bands at 3090 and 1040 cm^1 are related to the O-H stretching vibration and O-H bending vibration of the boehmite respectively [10]. Figure 2 shows a TEM micrograph of the Y-TZP powder coated with boehmite. The boehmite coating did not form a continuous shell layer on the surface of the Y-TZP powder, which may result from the minor addition of boehmite. The existence and composition of the boehmite surface layer were confirmed using EDS. It is interesting to note that boehmite crystallites were formed mostly on the surface of the Y-TZP powders and no individual boehmite crystallites were observed, showing that the boehmite crystallites were formed by heterogeneous nucleation and growth. Electrophoretic measurements were performed on coated and uncoated Y-TZP powders to evaluate the effects of the coating layer on the surface charge properties. Figure 3 shows the results of the zeta potential as a function of pH for the Y-TZP powders with and without the boehmite coating.

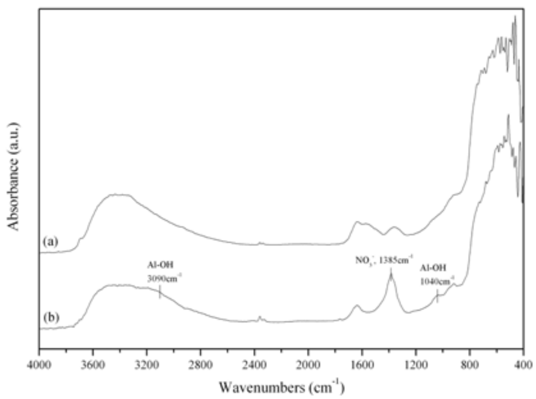

Fig. 1. FTIR spectra of Y-TZP powders before (a) and after (b) coating with boehmite.

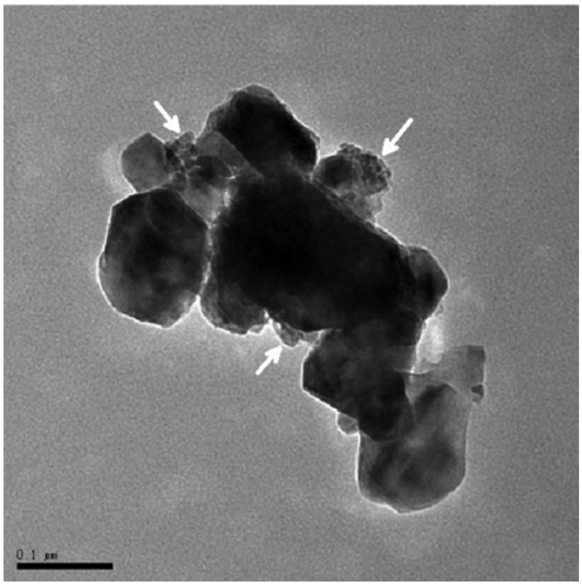

Fig. 2. TEM micrograph of Y-TZP powder coated with boehmite (the arrows: boehmite).

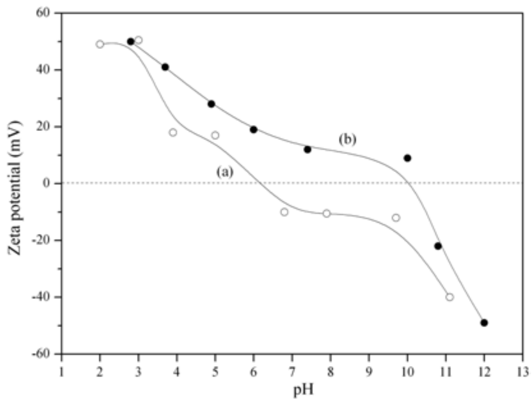

Fig. 3. Zeta potential as a function of pH for the Y-TZP powders without (a) and with (b) boehmite coating.

It can be seen that the isoelectric point of the uncoated Y-TZP powder is located at pH~6. However, the IEP was shifted to pH~10 which is similar to the IEP of boehmite (pH 10) [11] for the Y-TZP powders coated with boehmite. This result confirms that the boehmite crystallites were formed on the surfaces of the Y-TZP powders via heterogeneous nucleation.

In this experiment, the hydrolysis of urea proceeded and released NH^{4+} slowly, because the mixed solution was heated slowly. Therefore, the precipitant was always maintained at a low value, which resulted in a low supersaturation ratio. Consequently, the boehmite crystallites can form on the surface of Y-TZP powders by heterogeneous nucleation and growth in a low supersaturated solution, as evidenced by TEM and zeta potential measurements.

The densification behaviors of Y-TZP powders with and without a boehmite coating were obtained using a dilatometer. The densification curves are plotted in Fig. 4. It shows that the densification curve of Y-TZP coated

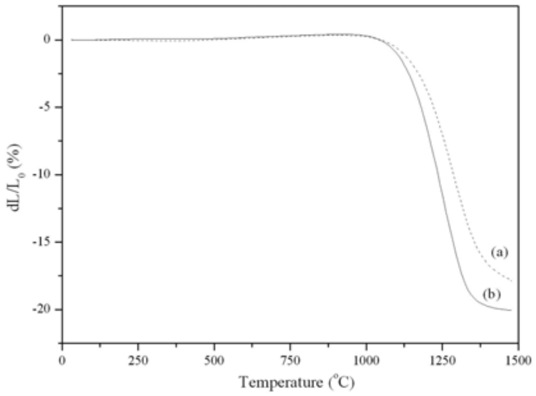

Fig. 4. The densification curves of Y-TZP powders without (a) and with (b) a boehmite coating.

with boehmite was shifted to a lower temperature and shrank more than that of theY-TZP without the coating. The densities of the Y-TZP with and without the boehmite coating sintered at 1350°C are 97.3 and 90.6% respectively. The final microstructures in Fig. 5 show the samples coated with boehmite compared with uncoated samples after sintering at 1350°C. The mean grain sizes of the pure Y-TZP and the Y-TZP coated with boehmite are similar. However, the sample coated with boehmite was highly dense and the uncoated sample was characterized by rather large pores. Yang et al. [12] investigated the effects of alumina on the densification of Y-TZP and observed that the sintered density of Y-TZP can be improved due to the accelerated materials transport by oxygen vacancies as the addition of alumina is within the solubility limit (about 0.2 wt%). A boehmite coating onto the surface of Y-TZP powders significantly improved the densification of the resulting ceramic bodies. The uniformity of the alumina incorporation into the Y-TZP using the boehmite coating process can enhance Al_2O_3 dissolution into the Y-TZP structure and the creation of oxygen vacancies, which can promote Y-TZP densification.

Conclusions

(1) The surface of Y-TZP powders can be coated with boehmite using an aluminum nitrate reaction with urea in an aqueous solution.

(2) A boehmite coating onto the surface of Y-TZP powders was confirmed using TEM, EDS, FTIR spectroscopy and zeta potential measurements.

(3) The uniformity of the alumina incorporation into the Y-TZP using the boehmite coating process can enhance Al_2O_3 dissolution into the Y-TZP structure and the creation of oxygen vacancies, which can promote Y-TZP densification. The relative density of the Y-TZP with a boehmite coating sintered at 1350°C can reach 97.3 %.

Acknowledgement

This work was supported by the Ministry of Economic Affairs (92-EC-17-A-08-S1-023) through the Particulate Materials Research Center of National Cheng Kung University.

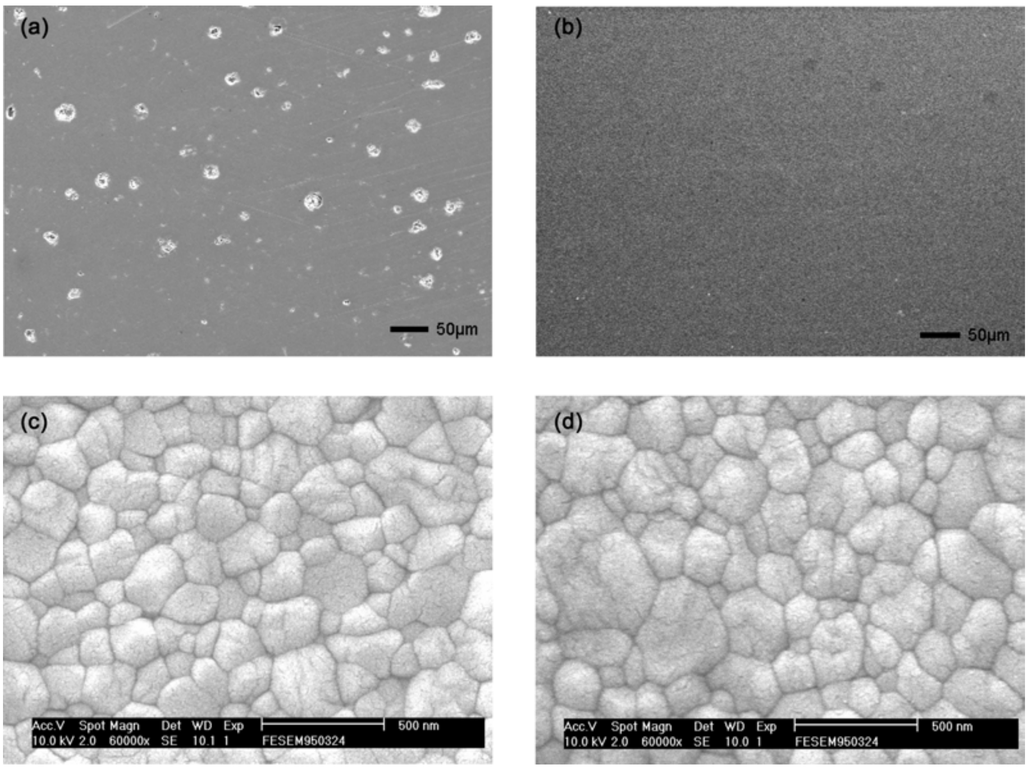

Fig. 5. The final microstructures of the samples without (a) (c) and with (b) (d) boehmite coating after sintering at 1350°C.

References

1. S. P. S. Badwal, J. Mater. Sci. 19 (1984) 1967-1776.
2. S. Lawson, J. Eur. Ceram. Soc. 15 (1995) 485-502.
3. C. L. Yang, H. I Hsiang, C. C. Chen, Ceram. Int. 31 (2005) 297-303.
4. S. Mullens, J. Cooymans, C. Smolders, J. Luyten, Am. Ceram. Soc. Bull. 1 (2005) 9101-9103.
5. Y. F. Tang, Z. D. Ling, Y.N. Lu, A. D. Li, H. Q. Ling, Y. J. Wang, Q.Y. Shao, Mater. Lett. 56 (2002) 446-449.
6. K. Prabhakaran, J. James, C. Pavithran, J. Europ. Ceram. Soc. 23 (2003) 379-385.
7. S. F. Wang, Y. F. Hsu, T. C. K. Yang, C. M. Chang, Y. Chen, C.Y. Huang, F. S. Yen, Mater. Sci. Eng. A 395 (2005) 148-152.
8. W. H Shih, L. L. Pwu, A. A. Tseng, J. Am. Ceram. Soc. 78 (1995) 1252-60.
9. S. Mei, J. Yang, R. Monteiro, R. Martins, J.M.F. Ferreira, J. Am. Ceram. Soc. 85 (2002) 155-60.
10. I. H. Joe, A. K. Vasudevan, G. Aruldhas, A. D. Damodaran, K. G. K. Warrier, J. Solid State Chem. 131 (1997) 181-184.
11. C. Y. Yang, W. Y. Shih, W. H. Shih, J. Am. Ceram. Soc. 84 (2001) 2834-40.
12. S. Y Yang, J. H. Lee, J. J. Kim, J. S. Lee, Solid State Ionics 172 (2004) 413-416.