Journal of Ceramic Processing Research. Vol. 11, No. 5, pp. 566~569 (2010)

J O U R N A L O F
Ceramic Processing Research

Factors in forming a junction by glass infiltration between different materials for a LTCC application

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The need for embedding the passive components into a LTCC substrate also has been increasingly focused on. In a junction between different materials, there have been two main problems such as shrinkage difference and interaction. In this study, zero-shrinkage glass infiltration technology was adopted for the junction. Two types of commercial glasses were examined for their glass transition temperatures, viscosities at high temperature and wetting angles to ceramic powders. The wetting angle was the most dominant in the infiltration behavior. 'Pb-B-Si-O'glass which has a low wetting angle was adequate for the junction for it infiltrated completely up to 28ß¦ into both alumina and barium titanate layers.

Key word: LTCC, Glass infiltration, Wetting angle.

Introduction

Material demand of radio frequency communication has led to the development of low temperature cofired ceramic (LTCC) materials which have dielectric constants less than 10. In order to achieve high integrity, the embedding of passive components into LTCC modules has been focused on recently. Some passive components, especially those capacitors needing a very high capacitance could not be made by LTCC with a limited thickness of substrate. Research, however, has been focused on the development of materials that are sinterable at low temperature and there have been fewer studies to solve the problem arising from forming junctions between different materials. There are two main problems. First, warpage and delamination occur due to shrinkage differences between different materials. Second, a chemical reaction may occur at the sintering temperature. It is necessary to solve these two main problems in a junction between different materials [1-3].

To solve these problems, laminated green sheets with a structure of 'ceramic/glass/ceramic' were sintered to make a ceramic-glass composite and it was reported that a glass was infiltrated into $BaTiO_3$ to make a glass/$BaTiO_3$ composite below 900. A junction with zero-shrinkage in the x-y directions between alumina and barium titanate was thought to be possible in these studies despite delamination among the layers [4-6].

In this study as an extension of previous studies, the main factors affecting infiltration in a junction between different materials using common glass have been investigated. Using two commercial glass powders, the high temperature viscosity and glass transition temperature (T_g) of the glasses were evaluated and wetting angles between the glasses and ceramic powders were measured.

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Experimental Procedure

The ceramic materials were $BaTiO_3$ (BT045, Samsung Fine Chemical, Korea) and Al_2O_3 (AES-23, Sumitomo Chemical, Japan), Pb-B-Si(here after LB) glass (NEG, Japan, denoted as 'LB' henceforth), Pb-B-Zn-Si(here after LBZ) glass (Phoenix PDE, Korea, denoted as 'LBZ' henceforth). Polyvinyl butyral (PVB, BM-SZ, Sekisui, Japan) was used as a binder, esteric surfactant (SN-Dispersant 9228, San Nopco, Japan) as a dispersant, and dibutyl phthalate (DBP, Daejung Chemical & Metals, Korea) as a plasticizer. As a solvent, a mixed solution of ethanol (95%, Daejung Chemical & Metals, Korea) and toluene (99.5%, Daejung Chemical & Metals, Korea) was used.

Ceramic powders such as the glass, alumina and barium titanate were dispersed in a dispersant-added (0.5-1.5 wt%) solvent for 24 hours, using zirconia balls. Dispersed slurries were well-mixed with the binder (8-14 wt%) and plasticizer. Mixed slurries were filtered with a #200 mesh, degassed and aged for 12 hours. Prepared slurries were cast with a doctor blade. The casting speed was 2.4 m·minute^{-1}, and the cast sheet-roll was dried at 80 °C. Sheet thicknesses were in the range of 25-50 μm.

As shown in Fig. 1, green sheets with structure of B (barium titanate)/G(glass)/B, A(alumina)/G/A, and A/G/A/B/G/B, were warm-pressed for 300 seconds at 60 °C, 50 MPa. And then, they were warm isostatically pressed for 30 minutes at 80 °C, 255 MPa. Prepared sheets were fired at 450 °C for 1 hour for binder-burnout. Pre-fired

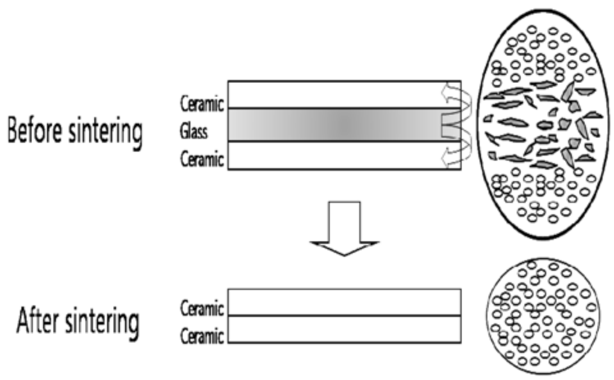

Fig. 1. Schematic of junction with zero-shrinkage ceramic/glass/ceramic structure.

specimens were sintered at 650 °C, 700 °C, 750 °C, 800 °C, 820 °C, 850 °C, 900 °C for 15 minutes. Sintered samples were observed with a field-emission scanning electron microscope (JSM-6700, JEOL, Japan) to measure the infiltration depth and to identify the state of the junction and microstructures [7].

High temperature viscosities of two commercial glass powders were measured. When the amount of glass frit is exceeded, it does not good affect on the electrical property and strength of ceramic body. Consequently we used a 5 Ø pellet considering the sufficient amount of glass in infiltrating and the wetting angel in ceramic body. So to determine the wetting angle between the glasses and ceramic powders (LB, LBZ vs. alumina, barium titanate), ceramic powders were formed into 10Ø pellets and glass powders were formed into 5Ø pellets, and then glass pellets were put on ceramic pellets, followed by high temperature optical observation [8, 9].

Result and Discussion

Fig. 1 shows a schematic of the glass infiltration method for zero-shrinkage a LTCC substrate presented in preliminary studies [4-6]. Glass powder which is non-reactive with ceramics was cast into a single sheet and placed between two ceramic sheets that were of the same material or of different materials. This 'ceramic-glass-ceramic' structured LTCC substrate showed x-y shrinkage below 0.2% when sintered.

Fig. 2(a) shows high temperature viscosity plots of two commercial glasses. The 'LBZ' glass appeared to have a lower viscosity than the 'LB' glass. So it could be thought that the 'LBZ' glass would infiltrate into the packed ceramic layers more easily than the 'LB' glass due to its lower viscosity during sintering.

Fig. 2(b) shows wetting angles of the two glasses to the two ceramic powders. The wetting angle of 'LB' to 'alumina' was lower than that of 'LBZ' by 10° at 900 °C. Glass wetting angles to ceramic powders indicate how a glass could be coated on the surface of ceramic powder in a packed layer and the lower the wetting angle, the better the wettability [10]. From this figure, it could be thought that 'LB' would be more easily coated on the surface of 'alumina' powder than 'LBZ' when sintered. And this coating occurred macroscopically as the infiltration of glass into the ceramic layer. The wetting angle of 'LB' to 'barium titanate' was lower than that of 'LBZ' by 50° at 900 °C. From these results, it can be seen that the LBZ glass had a bad wettability on barium titanate and the LB glass has a better wettability than that to alumina. The glass transition temperatures of the two glasses were 460.4 °C for 'LB' and 454.8 °C for 'LBZ', and did not show much difference.

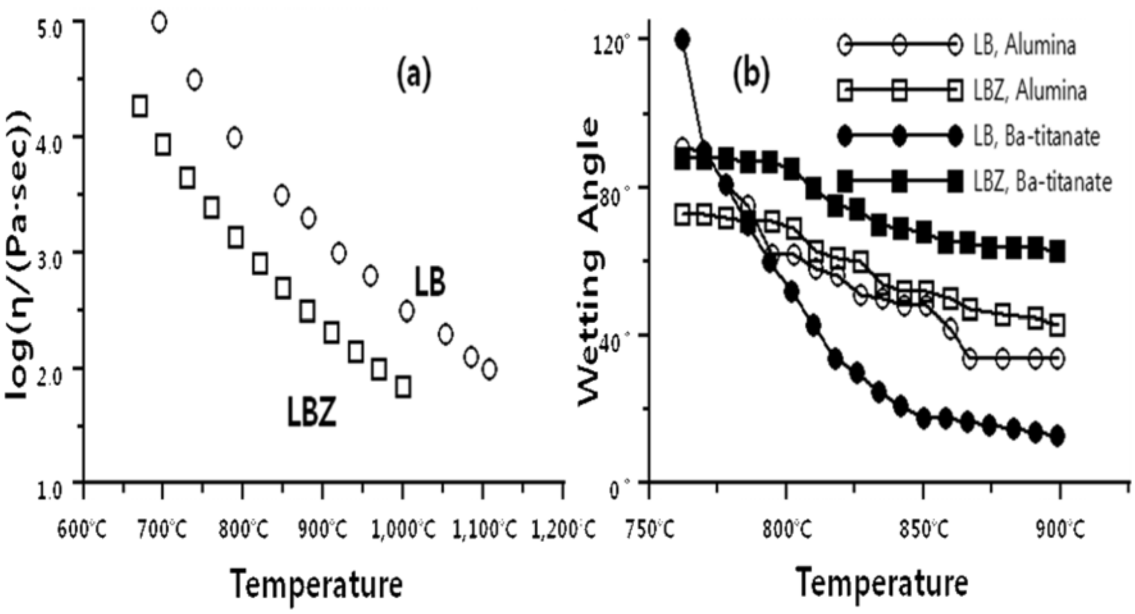

Fig. 2. High temperature characteristic of two glasses with (a) viscosities in the high temperature range, (b) wetting angles to 'alumina' and 'barium titanate'.

Fig. 3 shows infiltration depths of ‘LB’ to ceramic powders. (a) shows that ‘LB’ infiltrated into ‘barium titanate’ up to 54 μm and b) into ‘alumina’ up to 39 μm. In (b), however, the depth of full infiltration was about 20 μm.

Fig. 4 shows infiltration depths of ‘LBZ’ to ceramic powders. (a) ‘LBZ’ infiltrated into ‘alumina’ up to 24 μm at 900 °C a similar extent to about 20 μm of ‘LB’, but (b) shows that ‘LBZ’ hardy infiltration at all into ‘barium titanate’. In (b), infiltration into ‘barium titanate’ did not occur regardless of the temperature.

With a comparison of infiltration depth versus wetting angle and viscosity at high temperature, ‘LB’ with a high viscosity and a low wetting angle infiltrated into both ‘alumina’ and ‘barium titanate’ deeper than ‘LBZ’ which had a lower viscosity but a higher wetting angle. From this, it can be seen that the wetting angle is more important to infiltration than the viscosity at high temperature.

Fig. 5 shows a microstructural profile of a substrate with the structure of ‘A/G/A/B/G/B’ (here, ‘A’ means ‘alumina’, ‘B’ means ‘barium titanate’, and ‘G’ means ‘LB glass’) sintered at 820 °C where infiltrations into both layers were good enough. Fig. 5(b) shows the upper region of the ‘alumina’ layer where ‘LB’ glass infiltrated incompletely up to 28 μm and Fig. 5(c) shows the contact region between the ‘alumina’ and ‘barium titanate’ where the ‘LB’ glass in the ‘barium titanate’ region infiltrated over to the ‘alumina’ region. In Fig. 5(d) of the lower region of the ‘barium titanate’ layer, full infiltration may be seen. There also remained glass layers both between the ‘alumina’ layers and ‘barium titanate’ layers and large pores could be seen. Despite these glass layers and pores, a junction between different materials is thought to be possible from the microstructure observation, where the thickness of a ceramic layer was below 28 μm [11-12].

Conclusions

In this study, factors affecting infiltration for a junction

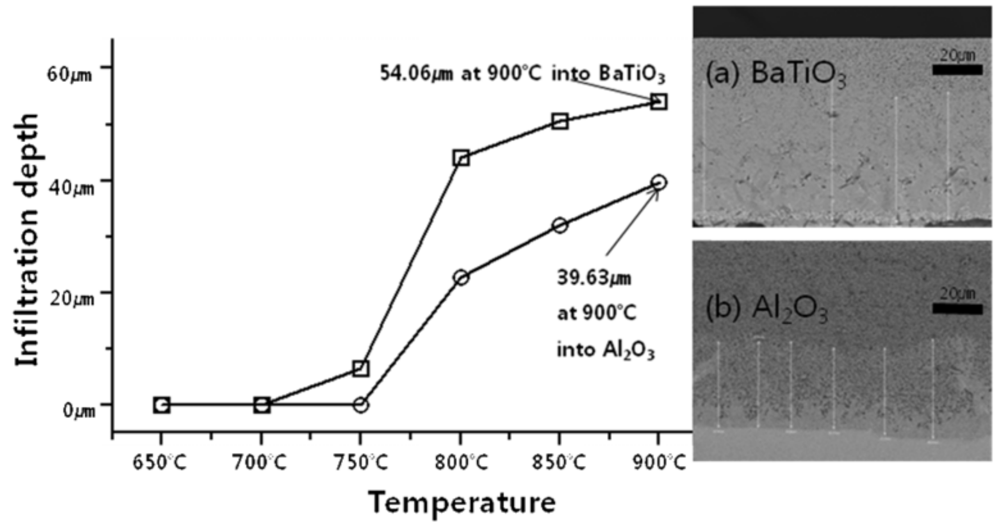

Fig. 3. Infiltration depth of ‘LB’ glass to ‘alumina’ and ‘barium titanate’.

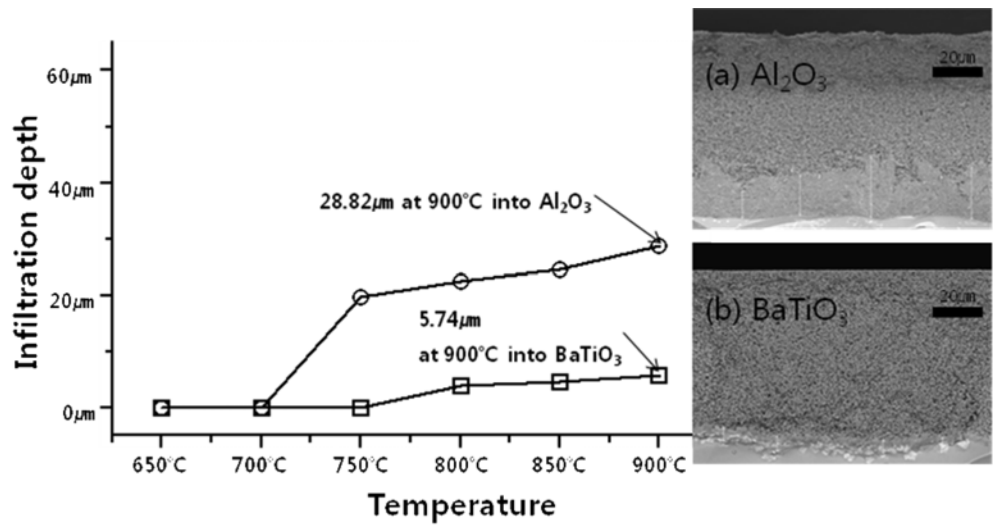

Fig. 4. Infiltration depth of ‘LBZ’ glass to ‘alumina’ and ‘barium titanate’.

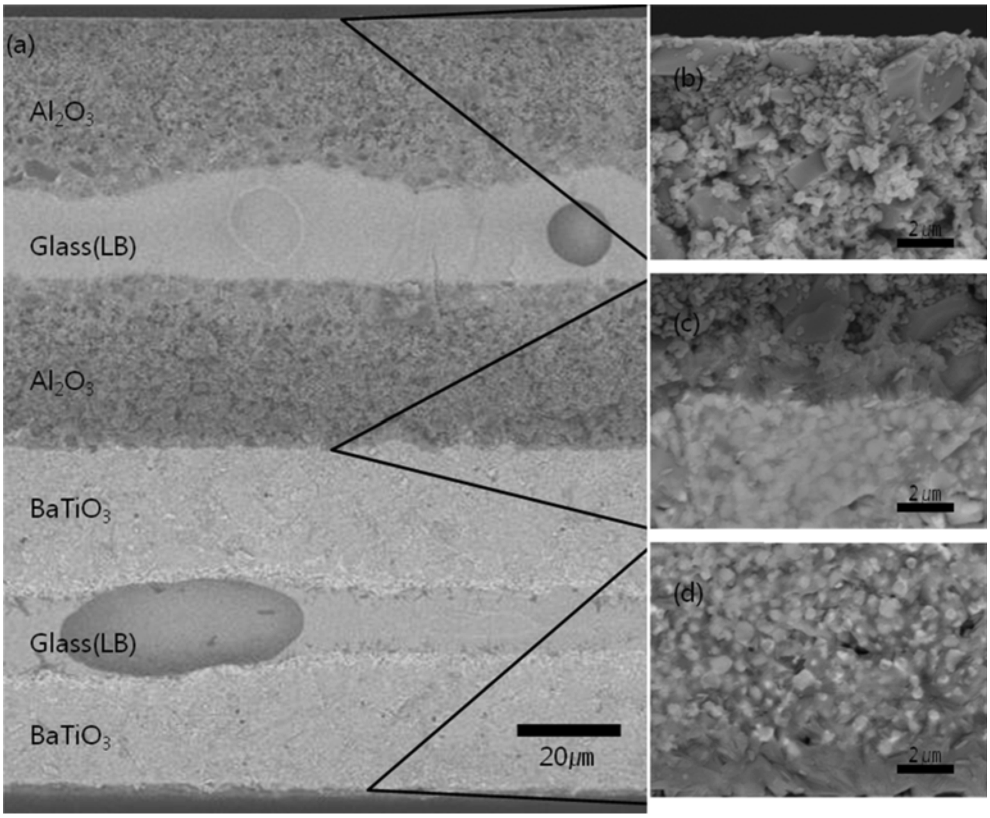

Fig. 5. Microstructural profile with the use of 'LB' glass for a junction between different materials sintered at 820 °C.

between different materials were investigated. Two types of commercial glasses were examined for their glass transition temperatures, viscosities at high temperature and wetting angles to ceramic powders. High temperature viscosities and wetting angles were thought to be important in the infiltration behavior. 'LB' glass which had a low wetting angle was adequate for the junction as it infiltrated completely up to 28 μm into both alumina and barium titanate layers. From this, it could be seen that the wetting angle of a glass to a ceramic is the most important in controlling infiltration depth.

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