

Silicon radical effect coated on alumina powder layer in junction between different materials for LTCC

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Received: 29 May 2007 / Accepted: 16 December 2007 / Published online: 19 January 2008
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Abstract Several studies are reported on the possibility of reliable bonding alumina and barium titanate with derivation from the glass infiltration method for zero-shrinkage substrates, but infiltration into alumina was not satisfactory, compared with the case of barium titanate. In this study, silicon radical, which is main component of glass, is coated on alumina powder in order to improve glass infiltration effect in bonding alumina and barium titanate with use of common glass in low temperature cofired ceramic composite. As a result, it is verified that infiltration effect on large-sized alumina powder layer was improved to the level of barium titanate which is fully glass infiltrated and bonding barium titanate and alumina was possible.

Keywords LTCC · Glass infiltration · Bonding

1 Introduction

Recent years, electronic devices with small size used in high frequency range necessarily required with advance of mobile communication. Multi-chip module is the most-favored form of those devices with this trend. Low temperature cofired ceramic (LTCC) substrate with low dielectric constant below ten is developed to be made into highly integrated module form embedded with some

passive components, replacing commonly used printed circuit board or alumina substrates.

Some passive components, especially requiring high dielectric constant or many layers, cannot be made to work enough with only LTCC which has low dielectric constant with limited thickness. With this problem, embedding of passive components into LTCC is required.

Multiple sheets with different dielectric constants are difficult to be cofired after laminated into single substrate, for two main problems exist. First, warping and delamination occurs due to difference of shrinkage rate among those materials during sintering. It is reported that warping increases significantly even with 1~2% of shrinkage differences. And though shrinkage rates are same for two materials, there could be limitation of sintering with different shrinkage behavior and insufficient shrinkage resulted. Second problem is chemical reactivity between each material. Minimizing this reactivity, reliable bonding has to be obtained [1].

Some studies on bonding of different materials with commercial glasses in LTCC application has been reported in order to solve these problems. According to the results, GA-1 glass infiltrates sufficiently in the range of 750 °C to 800 °C as glass/ceramic composite, confirmed by X-ray diffraction results. This may suggest the possibility of bonding different materials with zero-shrinkage in x-y plane. But, there is no experimental result if infiltration rate is same in alumina and barium titanate [2, 3, 5].

In this study, infiltration depths were compared with sintering temperature and infiltration rates with particle size were also investigated. To improve infiltration depth and rate, silica coating on ceramic particles were introduced.

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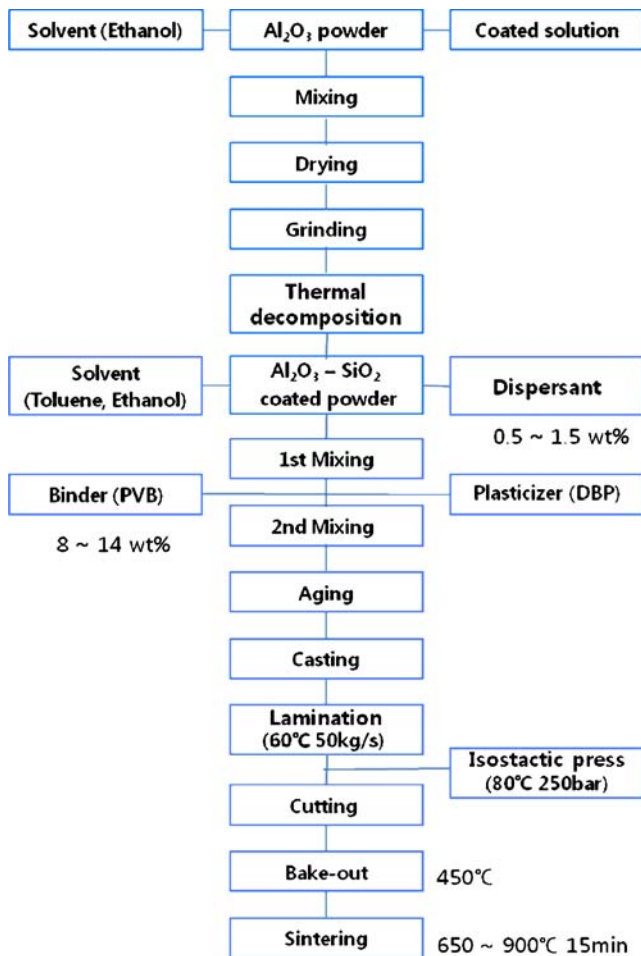


Fig. 1 Experimental procedure

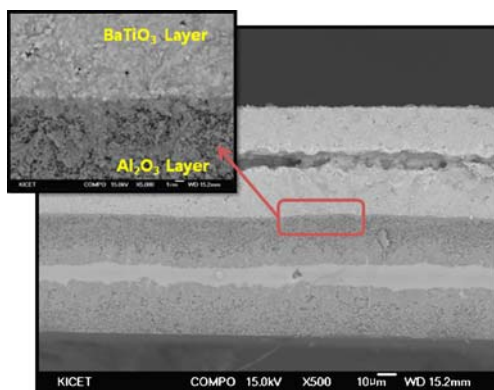


Fig. 2 Microstructure of sintered sheets at 800 °C with use of GA-1 glass of controlled amount

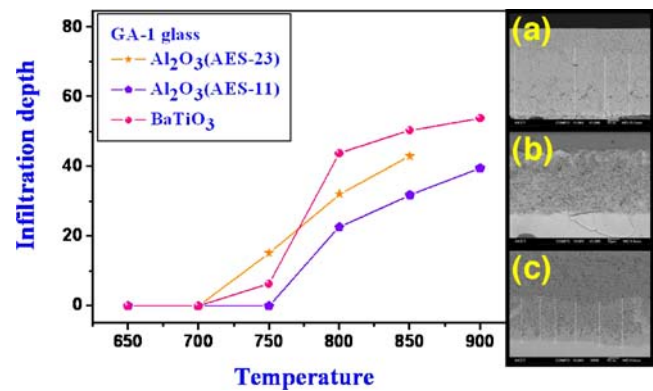


Fig. 3 Infiltration depth of GA-1 with temperature. (a) Al₂O₃ (AES-23), (b) Al₂O₃ (AES-11), (c) BaTiO₃

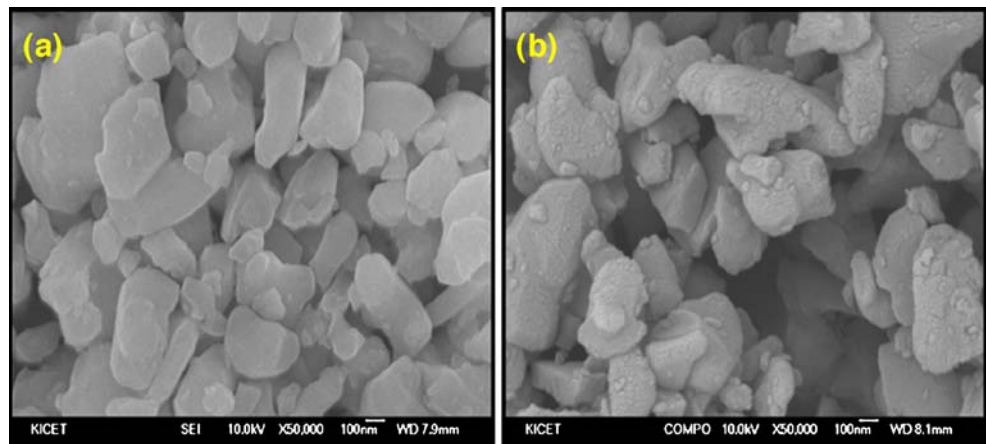
2 Experimental procedure

As raw materials, barium titanate (SBT-03B, Samsung Chemical, Korea), alumina (AES-23, AES-11, Sumitomo Chemical, Japan) and PbO-B₂O₃-SiO₂ glass (GA-1, NEG, Japan) for LTCC application were used.

Schematic of experimental procedure is shown in Fig. 1. Si-containing chemical (silane, Z-6040, Dow Corning®, USA) was added with weight ratio of 5~15% into alumina-ethanol dispersion for coating on alumina powder and mixtures were ball-milled for 2 h. Well-mixed slurries were dried, ground and then decomposed at 600 °C for 2 h. Ceramic powders including coated powder and glass powder were formed into sheets by tape-casting respectively. During preparation of slurry for tape-casting, polyvinyl butyral (BM-SZ, Sekisui, Japan), dibutyl phthalate (Daejung Chemicals & Metals, Korea) were used as binder and plasticizer respectively with dispersant (SN-9228, Sanopco, Japan). Mixed solvent of toluene and ethanol with azeotropic ratio was used for ball-mill. Powders, solvent and dispersant were ball-milled for 24 h with zirconia beads for dispersion. And then, binder and plasticizer were added into each dispersed slurry. This dispersed slurry was ball-milled again for 24 h for mixing. Prepared slurry was filtered with 200-mesh screen and aged for 12 h. Slurry was tape-casted, with film-feeding speed of 2.4 m/min, drying temperature of 80 °C, into green sheet with uniform thickness of 25~50 µm. Fabricated sheets were laminated at 250 bar for 300 s with hot-press. And then cut into certain size. Laminated sheets were fired at 450 °C for 1 h for binder burn-out. Out-burned sheets were sintered for 15 min at 650 °C, 700 °C, 750 °C, 800 °C, 850 °C, 900 °C respectively.

Sintered specimens were observed by field emission scanning electron microscope (JEOL, Japan) for bonding state and microstructure.

Fig. 4 Powder microstructures of alumina (a) before and (b) after coating



3 Result and discussions

Figure 2 shows profile microstructure of glass-amount controlled sheets sintered at 800 °C with use of GA-1. As shown in inset) it can be known that glass infiltrated into barium titanate deep sufficiently, while into alumina, insufficiently. Though glass quantity was estimated precisely, glass site for barium titanate was delaminated while that for alumina had remnant glass uninfiltreated. This suggests that infiltration rate was different in each material.

Glass infiltration depths into each powder were plotted with temperature in Fig. 3.

Infiltration depth into barium titanate was deeper than those into alumina with different particle size.

Into barium titanate, infiltration was completed deep as 54 μm, shown in a). In (b), depth was about 43 μm at 850 °C into 1.5 μm-sized alumina and in (c) depth was about 39 μm

at 900 °C into 0.5 μm-sized alumina. However, in alumina, infiltrations were insufficient and full infiltration depths were about 20 μm. At this, improvement of infiltration depth into alumina was required to level of barium titanate for reliable bonding between barium titanate and alumina [6–9].

Silicon, network former of glass structure, was coated on alumina particles to improve infiltration of glass.

Figure 4 shows powder microstructures of alumina before and after coating of SiO₂. There can be seen small particles of SiO₂ on surface of large alumina particles.

Alumina powders before and after Si-coating were made into pellets with use of ø10 mm mold. GA-1 glass powder was also made into pellets with use of ø5 mm mold. As temperature increases, GA-1 pellets on various alumina pellets were observed by high temperature microscope for comparison of glass wettability on both coated alumina and uncoated alumina and results are shown in Fig. 5 [10–13].

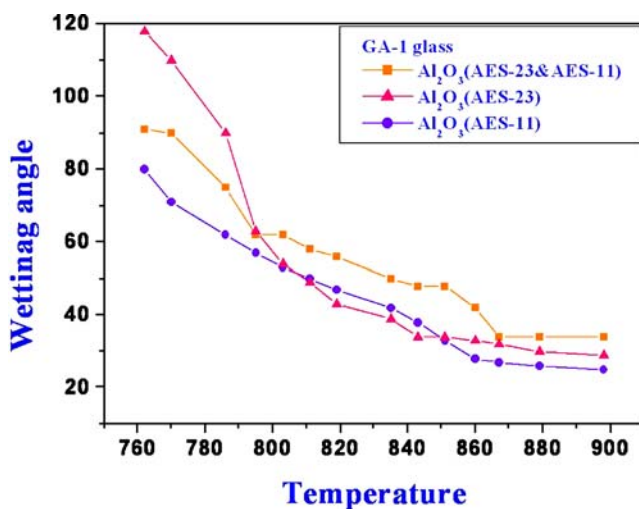


Fig. 5 Wetting angles of GA-1 glass with temperature on surfaces of alumina before and after coating

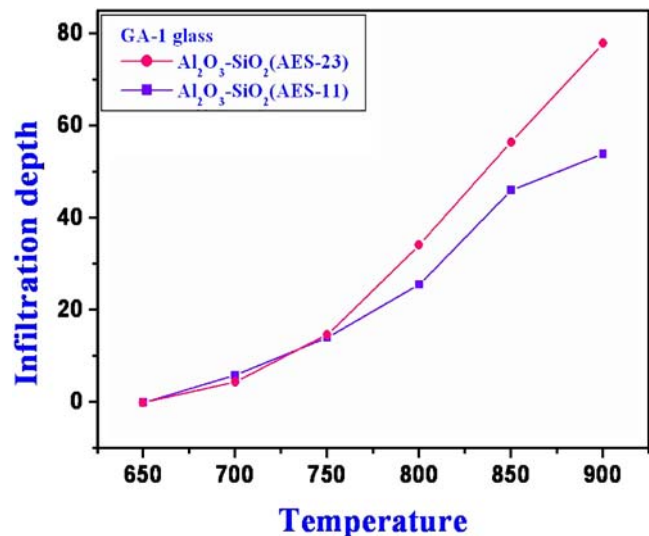
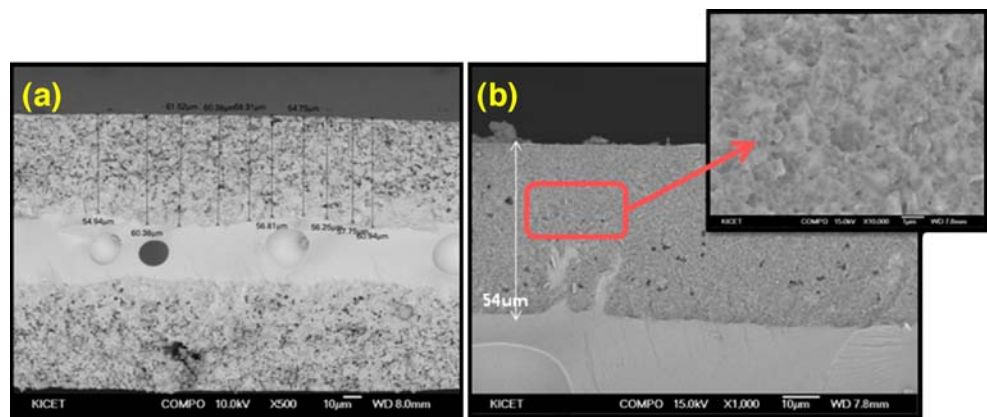


Fig. 6 GA-1 glass infiltration depth into coated alumina of different particle size with temperature

Fig. 7 Microstructures of infiltrated sheets with different particle size of coated alumina sintered at 900 °C. (a) AES-23, (b) AES-11



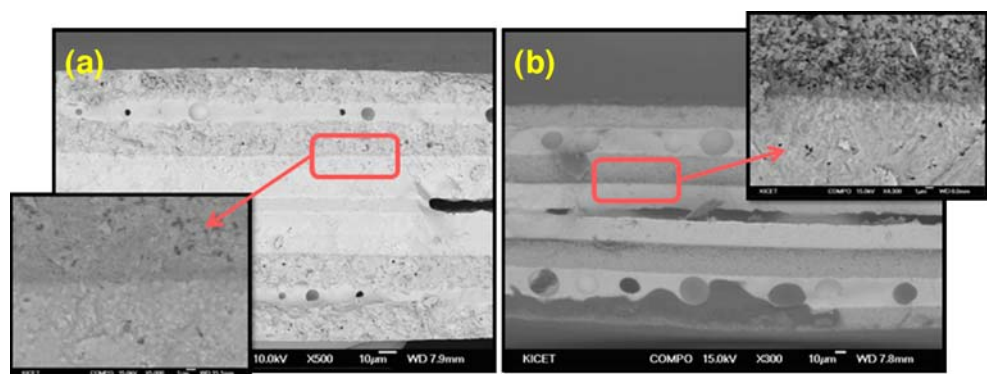
After coating, wetting angle is appeared to reduce as many as 10°. In preceding study, wetting angle is known to improve infiltration of glass largely, while high temperature viscosity, and T_g also affect that of glass (U.K. Jang et al., submitted in Materials Letters).

Figure 6 is plots of GA-1 glass infiltration depth into coated alumina with temperature. Glass infiltrated as deep as into barium titanate. And large-sized alumina showed deeper infiltration. As suggested by Erik [4] this is thought to be result of more fluent glass flow due to large pore size.

Figure 7 shows microstructures of laminated sheets sintered at 900 °C. In (a) of large-sized alumina, infiltration depth was about 78 μm, while large pores were observed, but in (b) of small-sized alumina, infiltration depth was 54 μm without noticeable pore. It is thought that pore distribution in the green sheet was kept in the infiltrated body.

Figure 8 shows microstructures of laminated sheets with different materials bonded and sintered at 800 °C. Coated alumina powders with different particle size were casted to thickness of 27 μm and laminated sheets were sintered at 800 °C.

Fig. 8 Microstructures of bonded substrates with different particle size of coated alumina and barium titanate cofired at 800 °C. (a) AES-23, (b) AES-11



In (a) of large-sized alumina, bonding was successful and infiltration was completed though there was remnant glass uninfiltrated in the alumina layer.

In (b) of small-sized alumina, infiltration into alumina was insufficient and large amount of remnant glass was observed in alumina layer and barium titanate layer showed delamination, due to lack of glass because glass in the barium titanate region infiltrated over boundary between alumina and barium titanate. It is thought that infiltration rate difference with different particle size of alumina made different bonding behavior [4].

4 Conclusion

To improve infiltration of common glass, alumina particles were coated with Si-chemical.

Large-sized alumina particle, which was coated, showed sufficient infiltration and good bonding state though further study is required to optimize glass layer thickness. And this suggests that bonding barium titanate and alumina can be possible with use of large-sized alumina coated through thickness control.

Acknowledgement This work was supported by the IT R&D program of MIC/IITA, Korea [2006-S005-02, “Ceramic Material and Process for High Integrated Module”].

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