Glass Infilteration in Bonding of $BaTiO_3$ and Al_2O_3 Layers

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Abstract

A novel sintering process is proposed for bonding of $BaTiO_3$ and Al_2O_3 layers. Common commercial glass was used and infilterated among filler particles. As the kind of commercial glass, the phenomenon of the infilteration is different. Although Sud-1140 glass forms a glass/filler composite, it is not completely infilterated into the filler particles at 900 ℃. However as the increase of sintering temperature the infilteration of glass was improved. In this study, GA-1 and GA-12 glasses were infilterated the more than Sud-1140 glass. However, they are reacted by $BaTiO_3$ layer. The results of the experiment show that constrained sintering and the co-firing of the different materials were possible for glass infilteration using Sud-1140 glass at 1000 ℃.

Keywords : LTCC, infilteration, bonding, heterostructure

1. Introduction

The free sintered LTCC in the x-y direction usually experience tape shrinkage of between 12%-16%, and slightly more in the thickness direction. The shrinkage and shrinkage variation limit the size of the substrates that can be processed, hence imposing some limitations on embedded passive components and introducing complexities in the processing of boards with cavities.[1] Especially, the high permittivity material with different chemical composition tend to display different shrinkage profiles with substrate material at low temperature sintering. Therefore, the difference of the sintering profile in the heterostructure is a fundamental problem in capacitor embedding. The proposed study comprising a self-constrained sintering and embedding technology is needed to enable more flexibility in module design and to develop suitable embedding materials.

In this study, therefore, a novel sintering process is proposed for bonding of $BaTiO_3$ and Al_2O_3 layers. For low temperature process, a common glass layer is used. The glass layers comprise ZnO-B_2O_3-PbO-SiO_2(Sud-1140), PbO-B_2O_3-SiO_2(GA-1), Na_2O-ZnO-B_2O_3(GA-12), and MgO-B_2O_3-SiO_2(GA-60). Each glass and filler was used as commercial powder. In a multilayer structure of $BaTiO_3$, common glass and Al_2O_3, microstructures, the densities and their defects were investigated at various sintering temperatures and glass compositions.

2. Experimental and Results

The properties of the materials are listed on Table1. Each powder and glass was milled with binder, plasticizer and dispersant for 24 hours. Each slurry was casted at a speed of 4m/mim and dried at 80℃. The thickness of the casted tapes was about 30μm. Each tape was baked at 450℃ for one day and its green density was measured. The relative green densities for each material were about 60%.

The laminates were prepared by sandwich structure comntaining $BaTiO_3$/glass/Al_2O_3 for each glass sheet. They were prelaminated by uniaxial press and finally pressed by warm isostatic press at 250bar. Green laminates were cut into square by 15×15mm. They were baked at 450℃ for 3hours and sintered at 900℃ and at 1000℃ for 15 minutes. The shrinkage of green and sintered laminates was measured by dimensional method. Shapes of sintered sample were observed by eyes and optical microscope. The microstructures of the fractured sample were observed by FE-SEM (Field Emission Scanning Electron Microscope, JEOL, Japan). The variation of the crystal structure was measured by x-ray diffraction analyzer.

LTCC materials are the mixture of the glass powder to be sintered at low temperature and the filler powder to keep electric properties. Gongora-Rubio et al.[2] showed a sintering model of the glass filler mixture in overview of meso-system technology as shown in Fig. 1(a). Glass was melted and the pores among the powders were filled. During the sintering process, alumina using filler powder rearranges in the melting glass. There may some times be some chemical reactions occurring as the composition of the glass and filler, such as phase transition and glass crystallization. Fig. 1(b) shows a model of constrained sintering by glass infilteration. In this method, the sintering shrinkage of laminate is constrained in the x-y direction by the separation of the glass and filler layer in the z-direction. During the sintering process, although the glass fills the pore, alumina powders are kept in the layer, and this makes it impossible for shrinking to occur in the x-y direction by sintering.[3] Fig. 1(c) shows the binding of the hetero-structure between alumina and $BaTiO_3$ filler layer, using the constrained sintering method as shown in Fig. 1(b). The core of LTCC technology is the cofiring between dielectric material and Ag electrode. Different materials shrink in different ways, and many papers have reported about them.[4] In addition to electrode, bonding between different dielectric materials is more important, increasing the concern for the development of a passive embedding technology. Fig. 1(c) shows the model for coupling of the constrained sintering and the bonding between different dielectric materials.

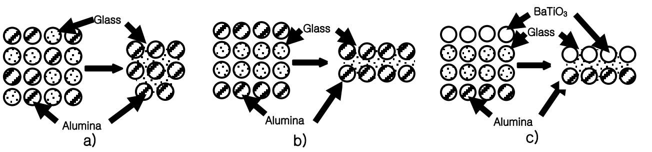

Fig. 1. Sintering model of the different materials using common glass.

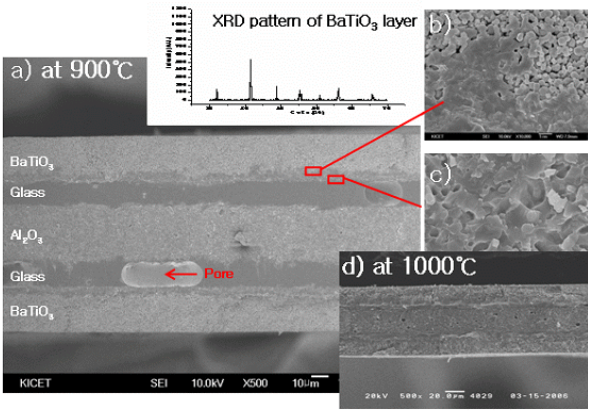

Fig. 2. Fractured microstructures and XRD pattern of sintered laminate using Sud-1140 glass.

Fig. 2 shows the fractured microstructure of the bonding layer using Sud-1140 glass. The thickness of the glass layer is about 13μm, and it can be seen that the glass layer does not disappear completely. Sud-1140 glass is infiltered into the alumina and $BaTiO_3$ layer. In Fig. 2(a), the transparent ellipse in the glass layer looks like a captured pore in a liquefied glass during sintering. The interface between the glass and $BaTiO_3$ powder is shown in Fig. 2(b). The microstructure of the glass/ceramic composite typically appears in the area where is put in the pore by glass, as shown in Fig. 2(c). However, when glass is not enough infiltered into $BaTiO_3$ particles, the layer appears as a powder packing. In the case of Fig. 2(d) which sintered at 1000℃, the thickness of glass layer is decreased until 5μm. Although sintering temperature is higher, it is shown the possibility of the infilteration sintering. Because the XRD pattern of $BaTiO_3$ layer isn't changed, glass don't effect to the $BaTiO_3$ phase.

3. Summary

To enhance bonding between different materials, glass infilteration in LTCC sintering process was studied. Sud-1140 glass was infilterated among the filler particles at 1000℃, and GA-1 and GA-12 glass were reacted with $BaTiO_3$ layer. In addition, GA-60 glass did not melt at 900℃. Sud-1140 glass formed a composite of glass/filler and the thickness of the glass layer decreased with increasing sintering temperature. Although infilteration is not enough at 900℃, in the case of Sud-1140, the possibility of the constrained sintering is shown in the sintering process which glass is infilterated in the filler at 1000℃.

4. References

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