Tape Casting System for ULTCCs to Fabricate Multilayer and Multimaterial 3D Electronic Packages with Embedded Electrodes

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A 3D multilayer structure built by two ultra-low temperature co-fired ceramic (ULTCC) compositions with silver embedded electrodes are co-fired at a temperature of 450°C. The 3D multilayer module is prepared by laminating the ULTCC green tapes with a new binder system, which organics can be completely burned out at temperature of 250°C before the sintering of the ULTCC 3D modulus. High-density microstructures are achieved for the sintered module. In this study, the ULTCC feasible binder system is introduced. Also, ULTCC multilayers and multimaterial structures with surface and embedded silver electrodes are fabricated. This research opens up a new horizon for fabrication of electroceramic devices with embedded electrodes in multimaterial devices.

Keywords: *ultra-low sintering temperature ceramics; tape casting; multilayers; glass-ceramics*

I. Introduction

LOW-TEMPERATURE co-fired ceramic (LTCC) technology is currently widely used in industry for high-frequency devices and electronics packaging. The advantage of this technology is that multilayer ceramic structures with embedded conductive lines and vias can be fabricated in co-firing process.[1] The process starts with casting ceramic tapes using slurry composed of desired ceramic powder, organic additives, and solvents. The tapes with printed circuits and vias are stacked, laminated, and co-fired at about 850°C to fabricate a three-dimensional monolithic package.[2] In last 10 years, ultra-low temperature co-fired ceramics (ULTCCs) with sintering temperatures less than 700°C are investigated enabling low energy consumption and integration with plastic or semiconductors to create diverse applications.[3–6] However, there are only two papers in open literature where tape casting has been performed for ULTCCs. Honkamo et al.[5] using polyvinyl butyral (PVB) binder system cast TiO_2-doped $Zn_2Te_3O_8$ tapes and sintered at 660°C. Ju et al.[6] studied the dielectric properties of SiO_2 filled $3ZnO–2B_2O_3$ glass tapes, which were sintered at temperature of 650°C. Also, in this case, the binder system was based on PVB. However, the tape casting for ULTCCs with sintering temperature approaching 500°C or less has not been reported. All well-known binder system for LTCC,[2] like the one based on PVB, is not suitable for ULTCC due to its high burn-out temperature (~400°C).

In this research, a new binder system for ULTCCs is introduced enabling sintering temperature even close to 300°C. Using this binder system, a low-permittivity Al_2O_3–BBSZ tape[7] and a high-permittivity $BaTiO_3$–BBSZ tapes[8] are cast, laminated, and co-fired at 450°C. Furthermore, co-firing of these tapes with commercial silver paste to form electrodes on top and inside the multilayer structures was studied. The procedure, densification, and microstructures of these two tapes and multimaterial structures are presented and discussed.

II. Experiments

The fabrication processes of $BaTiO_3$–BBSZ and Al_2O_3–BBSZ glass-ceramics were performed as earlier.[7,8] The slurry was prepared by mixing the glass ceramic powder with solvent, plasticizers, and a binder for 15 h. Dimethyl carbonate (DMC; Sigma-Aldrich, St Louis, Missouri, USA) was used as solvent. The binder was polypropylene carbonate (QPAC40; EMPOWER MATERIALS, New Castle, Delaware, USA), and the plasticizers were butyl benzyl phthalate (S160; Richard E. Mistler, Yardley, Pennsylvania) and polyalkylene glycol (UCON 50HB2000; Richard E. Mistler). The tapes were cast on silicone coated Mylar™ using a laboratory caster (Unicaster 2000, Leeds, U.K.) with a single 400-mm wide doctor blade and a casting speed of 0.8 m/min. Before releasing and further handing, the tapes were dried for 5 h using a metallic lid cover. Differential scanning calorimetric measurement (DSC) and thermogravimetric analysis (TGA) for green tapes were performed (Netzsch 404 F3, Selb, Germany) with a heating rate of 2°C/min for samples of 15.4 mg. Silver electrodes (599-E; ESL Europe, Berkshire, UK) were screen printed on the surface of the green tapes and dried 24 h at room temperature. Five to 10 layer tapes were stacked and hot isostatic laminated at 75°C and 80 MPa for 10 min. Silver paste printed tapes were placed on the top and in the middle of the stack. Laminated multimaterial modules of 6–10 layers with $BaTiO_3$–BBSZ and Al_2O_3–BBSZ were then fabricated. The laminated modules were debindered and sintered with porous alumina plate on the top. The bulk densities of the sintered samples were measured by the Archimedes method. The microstructures of multilayer structures were studied by field emission scanning electron microscope (FESEM) (Zeiss Ultra Plus, Oberkochen, Germany) with an energy-dispersive spectrometer (EDS). The thicknesses of the fired electrodes were around 10–18 µm.

Table I. Slurry Compositions for Tape Casting of $BaTiO_3$-BBSZ and Al_2O_3-BBSZ Composites

Glass ceramic		Powder	QPAC40 1.26 g/cm^3	S160 1.12 g/cm^3	UCON 1.06 g/cm^3	DMC 1.07 g/cm^3
Al_2O_3–BBSZ	wt%	89.4	7.4	1.6	1.6	71.5
(5.13 g/cm^3)	vol%	66.5	22.6	5.3	5.7	255
$BaTiO_3$–BBSZ	wt%	89.5	7.0	1.7	1.7	69.8
(6.33 g/cm^3)	vol%	61.8	24.2	6.8	7.2	285

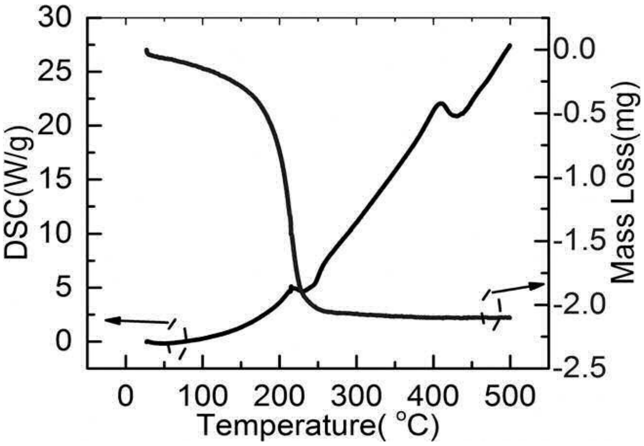

Fig. 1. Typical thermogravimetric analysis and differential scanning calorimetry (TGA/DSC) diagram of a green tape.

Table II. Shrinkage Rates of $BaTiO_3$-BBSZ, Al_2O_3-BBSZ. and Multimaterial Stacks in *x* & *y*- and *z*-Directions and Volume Shrinkage Rates and Bulk Densities

Bulk density of sintered tape	Al_2O_3–BBSZ	$BaTiO_3$–BBSZ	Multilayers
x & y shrinkage rate (%)	26	24	25
z shrinkage rate (%)	28	29	31
Volume shrinkage rate (%)	61	59	62
Bulk density (g/cm^3)	5.62	5.97	5.76

III. Results and Discussion

Table I lists the slurry compositions in volume and weight ratios of powders, QPAC40 binder system and plasticizers. The amount of solvent is also separately calculated and adjusted taking account the whole binder system. The thickness of the tape casted green tapes is 110 µm. The green density of the laminated stack is about 2.5 g/cm^3. The TGA/DSC measurements for the green tapes (Fig. 1) show that the organics content in the green tapes are burned out at temperature of 250°C. This organic system might be feasible for ULTCC materials with sintering temperature even close to 300°C. The DSC also reveals that the BBSZ glass melted and dielectric composites with $BaTiO_3$ or Al_2O_3 based on BBSZ glass are fully sintered at 450°C as reported earlier for the bulk samples.[7,8] According to the TGA/DSC results, the sintering profile can be proposed as increasing temperature with 3°C/min heating rate from room temperature to 250°C, dwelling at 250°C for 1 h to burn out the organics completely, and increasing temperature to 450°C with 1°C/min heating rate, dwelled at 450°C for 1 h.

Table II shows the linear and volume shrinkages and bulk density of $BaTiO_3$–BBSZ and Al_2O_3–BBSZ, and multimaterials stacks sintered at 450°C for 1 h. These results indicate that different glass-ceramics based on BBSZ glass have similar shrinkage rate with only 2% difference. The formation of defects between different glass ceramic layers during sintering is not found. Both sintered modules exhibit high densities (Table II), which are consistent with the values of the bulk sample (6.36 g/cm^3 for $BaTiO_3$–BBSZ and 6.03 g/cm^3 for Al_2O_3–BBSZ).[7,8] For the multimaterial module, the density is presented as the average of the density value of both tapes.

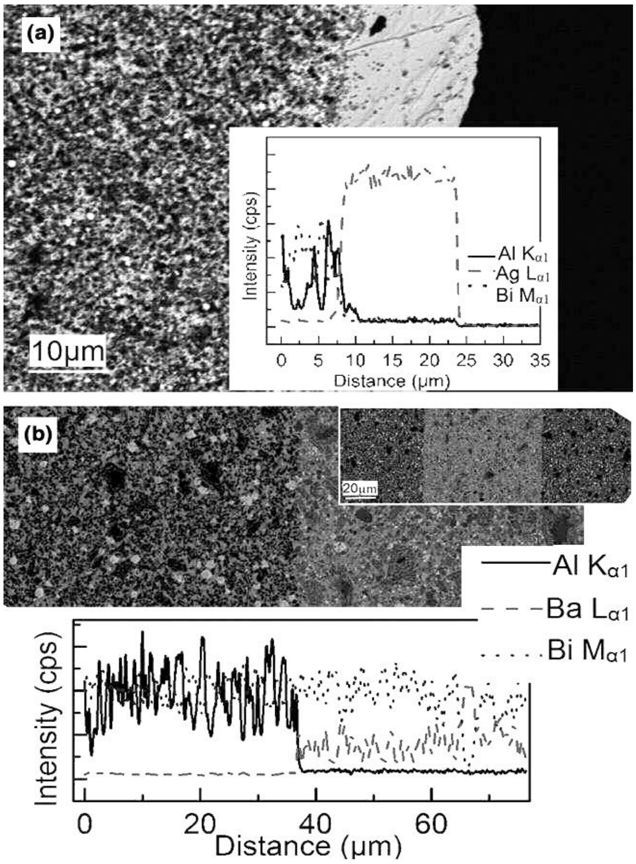

Fig. 2. (a) Backscattered electron image and linear energy-dispersive spectrometer analysis of the Al_2O_3–BBSZ tape with sliver electrode and (b) multimaterial stacks after sintered at 450°C for 1 h with insert showing a three-layers structure.

The FESEM backscattered electron images in Fig. 2 show dense microstructures for sintered modules which are similar with the phase structures as the sintered bulk samples reported in the previous investigations.[7,8] The cross section of the Al_2O_3–BBSZ tape and silver electrode co-fired at 450°C is shown in Fig. 2(a), where no chemical reaction or diffusion detected, which is proven by the linear EDS analysis (inset figure). Also, no diffusion or cracks observed at interface of the multimaterial stack is observed (Fig. 2(b)).

IV. Conclusions

In this paper, a new binder system for ULTCC with sintering temperature less than 500°C is introduced. The thermal analysis (DSC/TGA) shows that the organic additives are burned out at temperature around 250°C. The dielectrics of BBSZ glass with Al_2O_3 and $BaTiO_3$ fillers having various relative permittivity can be co-fired at 450°C with sliver electrodes without cracking and obvious diffusion. The developed binder system opens up a new horizon to for electroceramics to fabricate multilayers, multimaterial 3D electronics packages, and high-frequency devices in lower temperature with ultimate low fabrication costs.

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