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Fabrication of a high-performance film based borosilicate glass/Al_2O_3 ceramics for LTCC application

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ABSTRACT

The present work was initiated aiming at developing a high-performance film for LTCC application using a kind of polymeric dispersant in tape casting process of borosilicate-based glass/Al_2O_3 ceramics. The dispersion mechanism of the polymeric dispersant with the powders was analyzed in detail and the optimal amount of the dispersant addition was also determined through the sedimentation experiment. Its effects on the compactness, tensile strength and surface roughness of the films were analyzed. In addition, the sintering and microwave properties of the composite fired at 850 °C were also determined. For the application of tape technology, the co-firing compatibility between the composite and Ag electrodes at 850 °C was also verified. After testing and verification with different qualitative and quantitative instruments, a flexible film optimized by 1.5 wt% polymeric dispersant achieved the best overall performance.

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1. Introduction

The recent rapid developments in the microelectronic industry demand miniature microwave devices with high processing speed [1]. To meet this requirement, Low Temperature Co-fired Ceramic (LTCC) technology, which combines the use of low permittivity dielectrics with low loss glasses, has been developing rapidly [2]. Recently, glass/Al_2O_3 composites have received much attention because they can meet a lot of requirements of physical, electrical and thermal properties [3,4]. Different glass systems are designed to cater to the specific purpose, such as B_2O_3-Bi_2O_3-SiO_2-ZnO(BBSZ) [5], Li_2O-B_2O_3-SiO_2(LBS) [6], Li_2O–SiO_2–ZrO_2(LSZ) [7], CaO–B_2O_3–SiO_2-Al_2O_3(CABS) [8], etc. Among the different LTCC glass-ceramics systems, CABS glass/Al_2O_3 has been reported as a promising material for using in microelectronic packaging because of its low firing temperature and low dielectric loss [9].

In our previous study [10], the CABS glass/Al_2O_3 sintered at 850 °C showed $\varepsilon_r = 7.87$ and tan $\delta = 20 \times 10^{-4}$ at 7 GHz. The composite is physically and chemically compatible with the common electrode material, silver. For practical applications, the ceramic should be in the form of thin sheets through tape casting. The development of such flexible LTCC films with smooth surface, high density and high performance is the most important step in the fabrication of multilayer ceramic modules [11].

In this work, we introduced a kind of polymeric dispersant into slurry system instead of the common castor oil which dispersion mechanism was found to be steric hindrance [12–14]. This kind of polymeric dispersant belongs to the type of weakly cationic polymeric amine (WCPA). The physicochemical property of polymeric dispersant and castor oil are listed in Table 1. It is expected to disperse primary particles and to hold them in a homogeneous suspension by two methods, ionic repulsion and steric hindrance [15]. So the deflocculation of polymeric dispersant is better than that of castor oil in theory. We investigated the optimization results of the polymeric dispersant on the tape casting process of CABS glass/Al_2O_3 composite, expecting to develop a smooth surface, high density and strong mechanical films for further tape processing of LTCC application.

2. Experimental procedures

CABS glass with composition 15CaO:5Al_2O_3:16B_2O_3:64SiO_2 was prepared by conventional glass quenching method. After being equally mixed, raw materials were melted in a platinum crucible at 1500 °C for about 1 h. To prevent the occurrence of any crystallization, the melt was quenched into deionized water to form frits. Then, the glass frits were dried and milled in high speed agate mill, resulting in fine glass powders. The resulting glass powder ($D_{50} = 3.8\ \mu m$) was mixed with the Al_2O_3 ceramic pow-

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Table 1
Physicochemical property of polymeric dispersant and castor oil.

Dispersant	Ingredient	Molecular weight	Decomposition temperature
Weakly cationic polymeric amine(WCPA)	Substituted amino-terminal (-NR_3^+) group polycaprolactone	12000	500 °C
Castor oil	Ricinoleic acid, glyceride	933.26	265 °C

der (D_{50} = 3.0 μm) at the mass ratio [m(Al_2O_3):m(glass) = 45:55] for about 20 h. According to our previous study[8], the melting point of CABS glass was 710.7 °C with ε_r = 5.99 and tanδ = 1.4 × 10^{-3} at 7 GHz.

Due to the solid state of the polymeric dispersant, it should, firstly, be dissolved in solvent. So the tape casting slurry was prepared in a three stage process. In the first stage, polymeric dispersant was ball-milled for 5 h in the solvent (isopropanol/xylene/ethanol) ensuring it was fully dissolved. In the second stage, CABS glass/Al_2O_3 ceramic powder was added and ball-milled for 24 h. In the third stage, binder (polyvinyl butyral) and plasticizer (dibutyl phthalate) were introduced and ball-milled for another 24 h. The final slurry was then degassed in a vacuum desiccator for 10 min to remove air bubbles and cast using a tape casting machine (BHLY-011A). After that, the tapes were dried at room temperature. The thickness of the green tape was 160 ± 5 μm and the green tape was cut into circular sheets with the diameter of 21.46 mm. For the microwave test, 60 layers of the green tape were stacked and laminated together to obtain the final cylindrical sample at 21 MPa for 10 min at 70 °C. The thickness of the laminated sheets were about 6 mm. After binder burnout and sintering, dense LTCC composite could be obtained.

Zeta potentials of CABS glass/Al_2O_3 suspensions were calculated from the measured electrophoretic mobility using ZetaPALS (Brookhaven Instruments Corporation, USA). The chemical information of the powders and dispersant was measured by means of Fourier Transformation Infrared Spectrometer (Nexus-670, Nicolet, USA). Density of the film was the average of five sample datum through the density formula-mass divided by volume. Mechanical property of films was defined through electronic universal material testing machine (CSS-2205). Surface roughness of the film was measured using an Atomic Force Microscope (AFM) (Dimension Edge, Bruker, Germany). The microstructure was investigated by scanning electron microscopy (SEM) analysis (JSM-5900).The bulk densities of the sintered samples were evaluated by Archimedean immersion method using water as medium with the accuracy of ±0.001 g/cm^3. The microwave properties (dielectric constant ε_r and quality factor value) were determined by the cavity method with a network analyzer (HP 8722ET, Agilent, U.S.A.) operating at 7 GHz at room temperature. The temperature dependence of dielectric constant was also measured by the same method by changing temperature from −5 °C to 45 °C. The coefficient of thermal expansion (CTE) measurements from 23 °C to 300 °C were carried out by dilatometer Netzsch 402E. The heating rates for dilatometric measurements were 5 K/min.

3. Results and discussion

3.1. Surface features and dispersion mechanism

To identify the glass/Al_2O_3 powder surface properties (surface electrical behavior and surface chemical groups), Zeta potentials of CABS glass/Al_2O_3 suspensions with different pH values and infrared absorption spectrum of glass/Al_2O_3 powder were measured. Fig. 1a showed the zeta potentials of the powder suspensions with different WCPA content at different pH values. The zeta potential was noticeably decreased with the higher content of polymeric dispersant. The isoelectric point (IEP) of the powder was also affected by addition of polymeric dispersant WCPA, but all within the pH range from 2 to 4. The used powder could be determined to carry negative charges on the surface. Moreover, distinct changes of surface charge in dispersed glass/Al_2O_3 suspensions were due to adsorption of dispersant on the surface of the particles, inducing more negative charges to the particle double layer [16]. Infrared absorption spectrum of polymeric dispersant WCPA and CABS glass/Al_2O_3 was depicted in Fig. 1b. After detailed analysis, the determination of functional group was as follows: for WCPA, a typical spectrum showed the —NH group stretching vibration absorption bond at 2495.47 cm^{-1}. The –NH group bending vibration absorption occurred at 1552.27 cm^{-1} and 1648.80 cm^{-1}. The peak at 1724 cm^{-1} indicated the existence of carbonyl (i.e. C=O) in amide groups (i.e. –$CONH_2$). The peak at 3436.89 cm^{-1} indicated

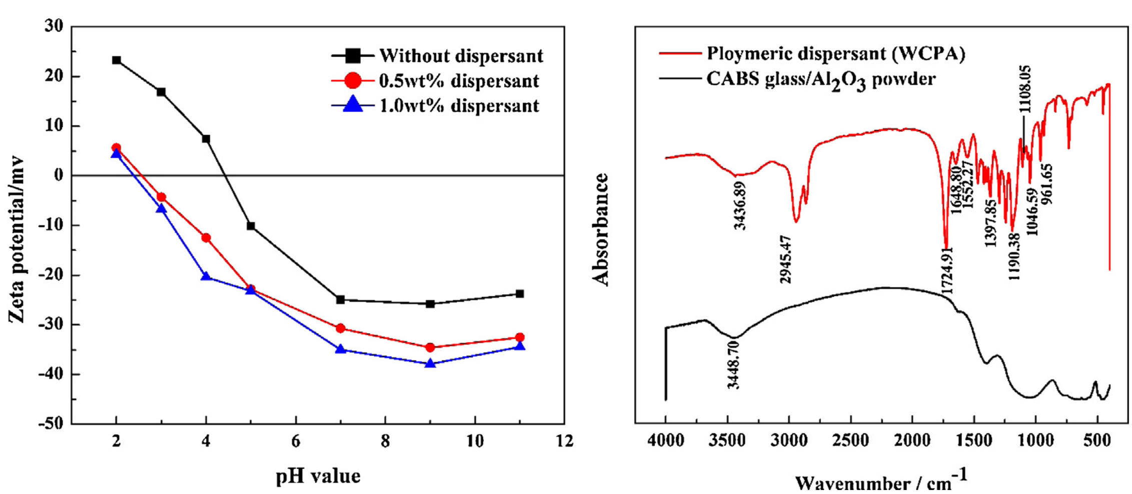

Fig. 1. Powder surface properties. a. Zeta potential as a function of pH value (surface electrical behavior). b. Infrared absorption spectrum of the dispersant and powder (surface chemical groups).

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