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Bimodal distribution of filler on viscosity and thermal expansion of glass composites

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Abstract

This paper investigates the bimodal oxide filler system to study the viscous behavior and thermal expansion properties of glass composites. Zinc oxide and cordierite, which are two types of filler, with different average diameters (10 μm and 1 μm, respectively), were considered in a Bi2O3 containing glass with various volume fractions (up to 40 vol%). The experimental results for the composites with the bimodal filler distribution show a reduced viscosity. The viscosity increased from fine particles to coarse particles with an increase in the volume fraction of the composite. Both viscosity and coefficient of thermal expansion (CTE) decreased significantly in the composite with the cordierite filler. The CTE is determined from the volume fraction with respect to particle size and distribution. On the other hand, viscosity is dependent on the particle distribution, particle size, and volume fraction of the composite.

Keywords: B. Composite; D. Glass; Oxide filler; Viscosity

1. Introduction

Currently, glass-filler composites are used for applications such as semiconductor packaging, low temperature co-fired ceramics, and display panels [1]. The preparation of suitable glass-filler composites is controlled by glass flow and viscosity. The study of glass viscosity from the transformation stage to the working temperature stage involves crucial steps that relate to the behavior of the seal [2]. Fig. 1 displays a schematic presentation of the sealing behavior of a glass-filler composite. The first phase consists of the firing stage of the glass-filler composite, and the second stage represents sealing with the filler distribution in the matrix.

Some researchers have implicated various properties, such as dielectric, thermal, and coefficient of thermal expansion (CTE) of the composite, by using bimodal distribution in polymer composites [3,4]. Previous researchers [5] studied the co-addition of oxide filler and various other types of filler to the glass composite. However, the filler of the same type with bimodal distribution in glass composites has rarely been investigated. This paper is an extension of previous work on an earlier approach that uses filler with two mean size distributions [6].

The present investigation focuses on the bimodal filler size distribution in glass composites and the effects of filler size and distribution of the viscous and CTE behavior of these glass composites. The composites were formed by adding one or more fillers to a glass matrix in various volume fractions to control the viscous and thermal expansion properties. The filler arrangement in the liquid phase of the composite, as a function of the firing temperature with respect to viscosity, was observed. Thus, the relationship between particle size, distribution, volume fraction, viscosity, and CTE allows for key physical parameters of the composite to meet product specifications.

2. Experimental procedure

A glass composition was prepared with 44 Bi_2O_3, 34 ZnO, and 22 B_2O_3. To achieve such a glass batch, the appropriate amounts of Bi_2O_3 (99.9%, Aldrich), ZnO (99.9% Aldrich) and B_2O_3 (99.98% Aldrich), were mixed and melted in air in an electric furnace at 1500 °C using an

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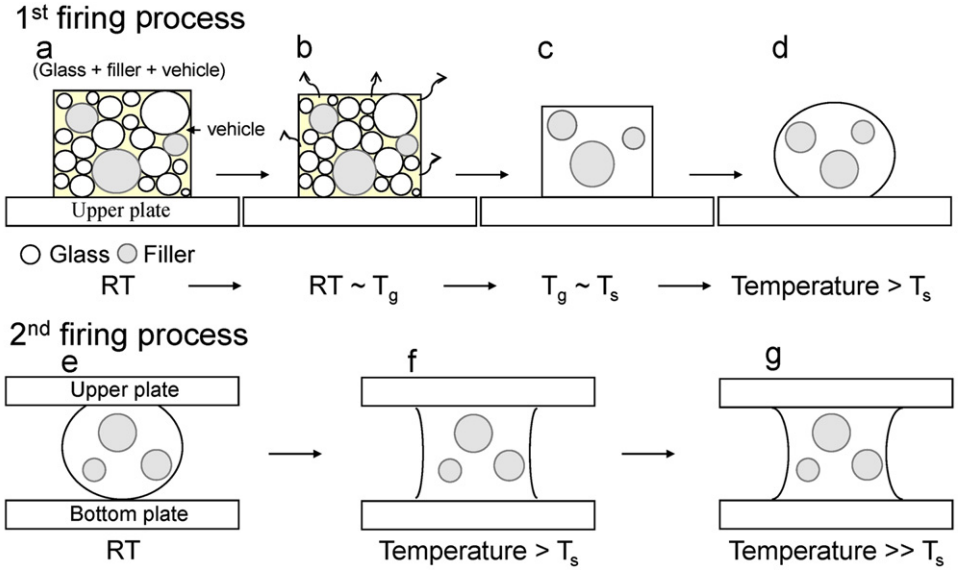

Fig. 1. Schematic diagram of the glass-filler composite that is used for sealing and the distribution of fillers in the glass matrix with respect to temperature. Sealing is used for a plasma display panel, which has a space between the two glass substrates bonded by glass powder. The sealed paste is fired at a softening temperature (a)–(d), cooled down to room temperature and then heated again for glass viscous flow (e)–(f): (a) green composite (b) after binder burn out (c) sintering of glass (d) viscous flow (e) upside down of plate (f) wetting of molten glass and (g) ideal shape of sealed.

alumina crucible. The quenched glass cullet was ball-milled to an average particle size of 100 μm frit. Then, the frit was pulverized with a jet mill to an average particle size of 5 μm. Commercially available cordierite and zinc oxide (Sigma-Aldrich Inc.) fillers were also considered. For both of the filler, the particle size was distributed over a wide range from coarse (10 μm) to fine (1 μm).

The glass-filler composites were prepared by adding the two fillers described above (zinc oxide and cordierite) with two broad ranges of particle sizes. The mixtures were prepared in various proportions, with 10, 20, 30, and 40 vol% fraction. The coarse and fine filler sizes were added to the glass powder with coarse to fine proportions of unity (50:50), in a bimodal filler mixtures. Zirconia balls with 3-mm diameters were added to the mixture and mixed well in a container inside a tubular mixer (Model T2 F, Glenmills, Basel, Switzerland). The arrangement of the fillers in the liquid phase of the composite were studied at various temperature ranges, from 480 °C to 580 °C, inside a sintering furnace, during a firing cycle with a 30-min holding time.

The glass transition temperature and exothermic and endothermic peaks of the glass-filler mixtures were analyzed using a differential thermal analyzer (DSC, NETZSCH, STA 449 F3). The measurements were performed in air at a heating rate of 10 °C/min. The samples were prepared by cold pressing the composite mixture. The surface morphology of the composites, after firing inside the furnace (Ajeon, S-18, sintering furnace) at 480 °C for a 10-min holding time, was analyzed with a scanning electron microscope (SEM; Hitachi S-4300). The densities of the green composites were calculated by using a pycnometer (Micromeritics, Accupyc II 1340). The crystallization phases of the green and fired composites were analyzed using XRD techniques (X-ray diffractometer, DMAX 2500) at 4 kV with a scan speed of 2°/min.

3. Results

The results of the particle size analysis (PSA) for the glass are shown with average particle size of 5 μm (Fig. 2a). Fig. 2(c) shows the fine particle size of cordierite in the submicron range as being similar to the fine particle size of zinc oxide in Fig. 2(b). In terms of bimodal distribution, filler with coarse and fine particle sizes were chosen with broad distributions in the composite [7]. The fraction that tended toward unity showed the best results among the distributions that were tested [8,9].

In the case of bimodal filler distribution, it has been observed that fine fillers arrange themselves within the interstitial gaps of the coarse filler (Fig. 3a–d). The composite with a 40 vol% fraction of the cordierite filler exhibits an excess amount of filler within the glass matrix, which created voids in the composite (Fig. 3c). The interphases are observed in the zinc oxide-glass composite (Fig. 3d). This implicates that interphase layer results due to interaction between zinc oxide filler dissolution in the glass matrix. The zinc oxide-glass composite represent the reactive system with lighter region represents the glass matrix, darker region represents filler and the medium region represents the interphase layer. The characterizations of the composites were carried out by the XRD (Fig. 4). The composite with cordierite-glass shows amorphous nature in both green and fired conditions. On the other hand the sharp and distinct peaks are observed in zinc oxide-glass composite in both green and fired composites. Partial dissolution of the zinc oxide filler within the glass matrix was confirmed from the XRD analysis