



Influence of diamond particle size on the thermal and mechanical properties of glass-diamond composites



Dandan Feng, Zhihong Li*, Yumei Zhu, Huanli Ji

Key Laboratory for Advanced Ceramics and Machining Technology of Ministry of Education, School of Materials Science and Engineering, Tianjin University, Tianjin 300350, People's Republic of China

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ABSTRACT

In this study, the particle size of diamond was used as a variable to investigate the properties of glass-diamond composites. A low melting point borosilicate glass was selected and its thermal analysis was performed by TG-DSC. The phase composition of the glass-diamond composites was analyzed by XRD, and SEM images showed the microstructure of the composites. Thermal properties of the composites were characterized by thermal conductivity and coefficient of thermal expansion (CTE). The bending strength was measured and the results showed that the glass-diamond composites with 30 μm diamond had the lowest CTE and the highest thermal conductivity and bending strength.

1. Introduction

With the miniaturization, multi-functionalization and high frequency of electronic components, the relative heat flow and calorific value have become increasingly higher, which increases the requirement of heat dissipation of the device. The key problem of electronic cooling lies in efficient thermal conductivity, and it can be effectively solved by studies on thermal management [1]. At the same time, electronic packaging materials have become the main part of thermal management materials, because of the development of electronic packaging. Low temperature co-fired ceramics (LTCC) technology as an effective approach to the miniaturization and multi-functionalization of the electronic components has become the mainstream technique of passive integration [2,3]. However, despite the processing cost of traditional LTCC materials being reduced, its thermal conductivity performance is poor and does not meet the increasingly high dissipating heat demand [4–6]. Therefore improving thermal conductivity of the LTCC materials has become a high-priority research topic.

The most important properties of determining heat conduction in materials are thermal conductivity and diffusion. Electronic packaging materials should possess appropriate coefficient of thermal expansions (CTE) to match with substrate materials as well as high thermal conductivity to meet the cooling requirements [7,8]. It is well known that diamond monocrystals have a very high thermal conductivity value and that their thermal expansivity matches well with Si [9]. However, diamond monocrystals are small in size, and the processing is extremely difficult. However, the advantages of a diamond composite are

obvious—simple preparation technology and adjustable CTE to match with the substrate and reduce the harmful effect of thermal stress [10]. Research on diamond composites as electronic packaging materials mainly focus on diamond films and metal matrix diamond composites [11–13]. The preparation methods are mostly by chemical vapor deposition (CVD), high temperature-pressure and spark plasma sintering. These methods are relatively restrictive and have high preparation costs, and their usage limitation limits its development.

The LTCC materials developed can be divided into two categories—glass/ceramic and glass-ceramic systems [2]. This study used diamond as a high thermal conductivity ceramic material charge and borosilicate glass as glass material as the amorphous matrix, exploring the thermal and mechanical properties of the glass-diamond composites prepared at low temperature and normal pressure. According to practical experience and numerous studies [14,15], borosilicate glass with appropriate composition has a low melting point and good thermal stability, which benefits the sintering of diamond composites. In a substrate material, the particle size and morphology of the ceramic grains play important roles on its properties. Too large or too small particle size reduces the sintering densification and then affects the other performances of the composites. Consequently, for the same system and sintering technology, reasonable selection of the ceramic particle size becomes the key factor affecting the final performance of composites. Thus, this paper investigated systematically the influence of diamond particle size on the properties of glass-diamond composites.

* Corresponding author at: No. 135, Yaguan Road, Jinnan District, Tianjin, People's Republic of China.
E-mail address: lizhihong@tju.edu.cn (Z. Li).

Table 1
Composition of the glass used in this study (mass fraction).

Component	SiO ₂	Al ₂ O ₃	B ₂ O ₃	Li ₂ O	Na ₂ O	CaO
wt%	51	7	34	3	6	5

2. Material and methods

2.1. Sample preparation

The borosilicate glass used in this study was based on a SiO₂-Al₂O₃-B₂O₃-Li₂O-CaO-Na₂O glass system, and Table 1 lists its composition.

Components were introduced in the form of silica, alumina, boric acid, lithium carbonate, calcium oxide and sodium carbonate, respectively. The raw glass powders were dry ground for 6 h in a ball mill (ball:powder = 2:1). The batch was melted at 1300 °C for 2 h and then quenched in cold water. The glass frit was dried, milled, and sieved using a 250# mesh (63 μm).

The glass powder and diamond (monocrystal, Henan Funik Ultrahard Material Co. Ltd, China) of different particle sizes (3 μm, 10 μm, 20 μm, 30 μm, and 40 μm) were mixed in proportion and dry pressed to form the glass/diamond green bodies. The composites samples were then pressed into circular pellets of Φ 30 × 2 mm and rectangular bars with dimensions of 20 mm × 6 mm × 4 mm and 30 mm × 6 mm × 4 mm. Some green bodies of 20 mm × 6 mm × 4 mm size were directly measured for sintering shrinkage. The rest of samples were sintered in an electric furnace with a highest constant temperature of 750 °C. The sintered composite samples were then studied to measure their thermal conductivity, coefficient of thermal expansion, and bending strength.

2.2. Sample characterization

The TG-DSC curves of the glass were measured using thermal analyzer (449C, NETZSCH STA, Germany) at a continuous heating rate of 5 °C/min. The phase composition of the glass-diamond composites was analyzed by X-ray diffraction (D/MAX-2500, Japan). The sintering shrinkage and thermal expansion of the samples were measured using a thermal dilatometer (DIL-402C, NETZSCH STA, Germany). The density of the obtained glass-diamond composites were tested by the Archimedes drainage method. The microstructure of the glass-diamond composites was observed by a scanning electron microscope (SEM, TDCLs4800, Hitachi Ltd, Japan), and the thermal conductivity of the glass-diamond composites was determined on the Laser Flash Apparatus (LFA 427, NETZSCH Geratebau GmbH, Germany). In addition, the bending strength of the glass-diamond composites was measured by the three-point test method with a universal tester (XWW, Beijing Jinshengxin Detecting Instrument Co., Ltd., China) at a span length of 20 mm and crosshead speed of 0.5 mm/min.

3. Results and discussions

3.1. Thermal analysis of the glass

Thermal analysis of the glass was performed using TG-DSC curves. In Fig. 1, the vertical axis represents the weight loss and the heat absorption/release during the heating process of glass.

The left vertical axis represents weight loss during the heating process of glass. Fig. 1 shows that there are two sharp exothermic peaks at about 226 °C and 264 °C with a remarkable weight loss that corresponds to the loss of crystal water and impurity in materials. Because glass is an amorphous material and has no constant phase transformation point, the glass transition from solid to liquid occurred gradually as the temperature increased; therefore, there was no obvious endothermic peak in the DSC curve instead, heat was absorbed during the course of heating. With temperature increasing, the glass absorbed heat

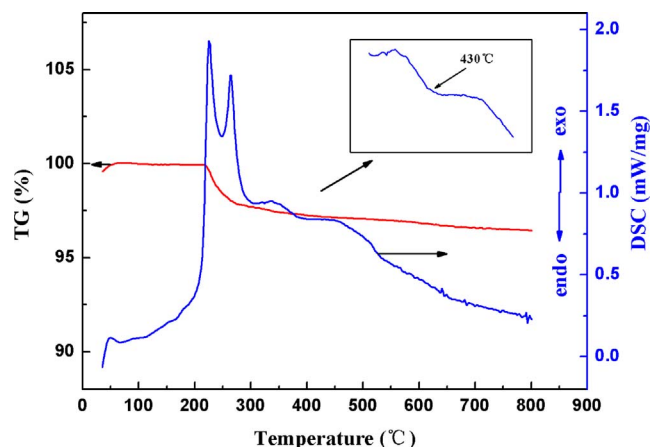


Fig. 1. TG-DSC curves of the glass.

constantly and the DSC curve declined. Fig. 1 shows that when the temperature reached about 350 °C, the DSC curve had an inflection point of bate endothermic, suggesting an obvious heat absorption of the glass. At this temperature, the glass went through a sudden change—the viscosity of the glass decreased rapidly and began a viscous flow deformation. The TG-DSC curves suggest that the glass used in this experiment had a low softening point. The formation of the glass melt at lower temperature allowed for a low sintering temperature of the glass-diamond composites [14].

3.2. Phase analysis of the glass-diamond composites

To establish the phase composition of the glass-diamond composites, XRD analysis of the samples was performed (Fig. 2) with diamond particles of different size.

The composites showed two crystalline phases: the main phase was diamond and the secondary phase was quartz. The crystalline SiO₂ was separated from the glass melt during the heating process, and a small amount of it can enhance the glass strength. The XRD patterns showed that the crystal type was constant and that diamond had no graphitization during the process of sintering, which guaranteed the good performances of the glass-diamond composites.

3.3. Sintering shrinkage of the glass-diamond composites

The sintering shrinkage of the glass-diamond composites with 3 μm and 30 μm diamond particulates were characterized by sintering

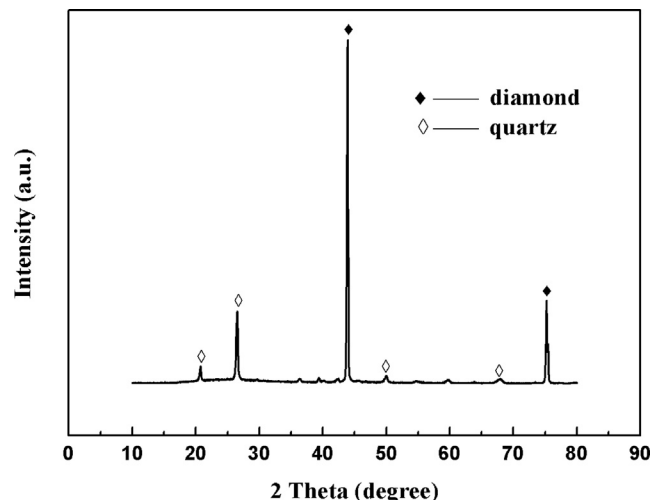


Fig. 2. XRD patterns of the glass-diamond composites.

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