

Liquid-phase sintering of lead borosilicate glass-alumina composite

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In the past few years there has been a search for suitable low-temperature-sinterable alumina-based compositions which could be cofired with other dielectric materials and pastes for use in multilayer hybrid microcircuit substrate applications [1]. Glass-ceramic materials having low dielectric constants and which could be sintered at 900°C in air or in a neutral atmosphere have been suggested [2]. The advantages of sintering in the presence of a liquid phase have long been recognized in both powder metallurgy and ceramics [3-7].

Glass powder of the following composition was used for the present study: (wt %) 65.18 PbO, 20.13 B₂O₃, 13.02 SiO₂ and 1.67 Al₂O₃. Details of glass powder preparation are reported elsewhere [8]. The composite was prepared as follows. A mixture of 55 wt% alumina and 45 wt% glass was first ball milled for 10 h in water medium using zirconia balls in a polyethylene container. The dried composition was mixed with 3% PVA as binder and granulated. Pellets were made from -60 to +200 mesh fraction of these granules. Discs of 15 mm of 15 mm diameter and 4 mm thickness were pressed with a pressure of 100 MPa. The discs were sintered in air for a duration of 210 min at temperatures ranging from 900 to 1100°C. A typical sintering schedule is given in Fig. 1. Diametrical shrinkages of more than 250 samples were measured. XRD and SEM studies were carried out in a Philips Diffractometer Model No. PW 1710 and Camscan Cambridge System, respectively.

The XRD patterns of the glass-alumina composites sintered at different temperatures are shown in Fig. 2. Peaks marked E do not appear in pure alumina. They

even appear in compositions heated to 900°C and are due to the reaction between the glass and alumina. Intensity of these new peaks, E, gradually increases up to 1000°C indicating the extent of the reaction. Above 950°C, considerable peak shift can be noticed. At 1050°C the intensities of all the peaks have drastically decreased showing reduction in the crystalline phase.

The variation of diametrical shrinkage as a function of sintering temperature is shown in Fig. 3. It can be observed that, initially, shrinkage increases with sintering temperature and reaches a maximum around 1000°C, and then decreases. The process is essentially liquid-phase sintering. The viscosity of the liquid glass formed decreases with increasing temperature, which in turn enhances densification due to the surface tensional rearrangement [3].

Figs 4a and c show a schematic representation of the green compact and the composite sintered at 1000°C, respectively. The densification in this case is mainly due to particle rearrangement. The average particle size of the alumina powder (RC 172, Renold's Co., USA) was 0.65 μm. Each particle therefore consisted of only one crystallite and hence the secondary rearrangement through grain boundary penetration by the liquid and particle disintegration is nonexistent in this case [9]. The following are the factors contributing to the decrease in shrinkage after 1000°C:

(a) The increase in temperature which increases alumina dissolution in glass [10]; this can be seen from the XRD pattern (Fig. 2). As a result of this, the viscosity of the liquid glass increases, which slows down the initial rearrangement due to capillary pull.

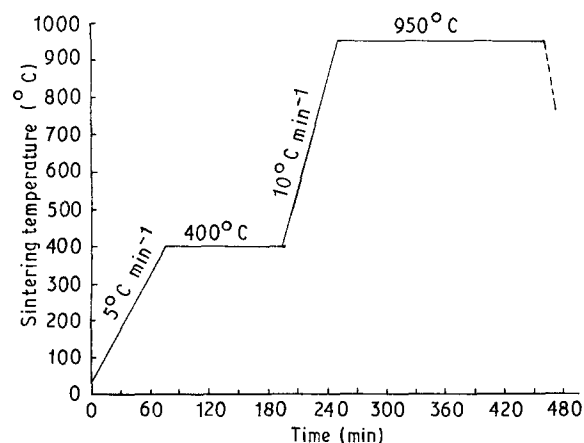


Figure 1 A typical sintering schedule for the glass-alumina composites.

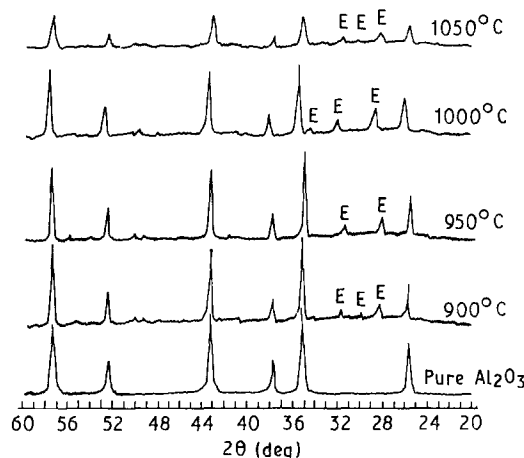


Figure 2 XRD patterns of pure alumina and glass alumina composites heated at different temperatures.

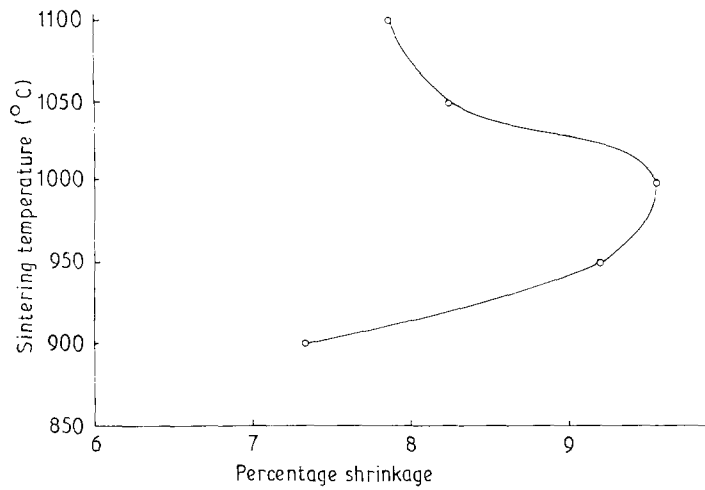


Figure 3 Diametrical shrinkage plotted against sintering temperature for glass-alumina composites.

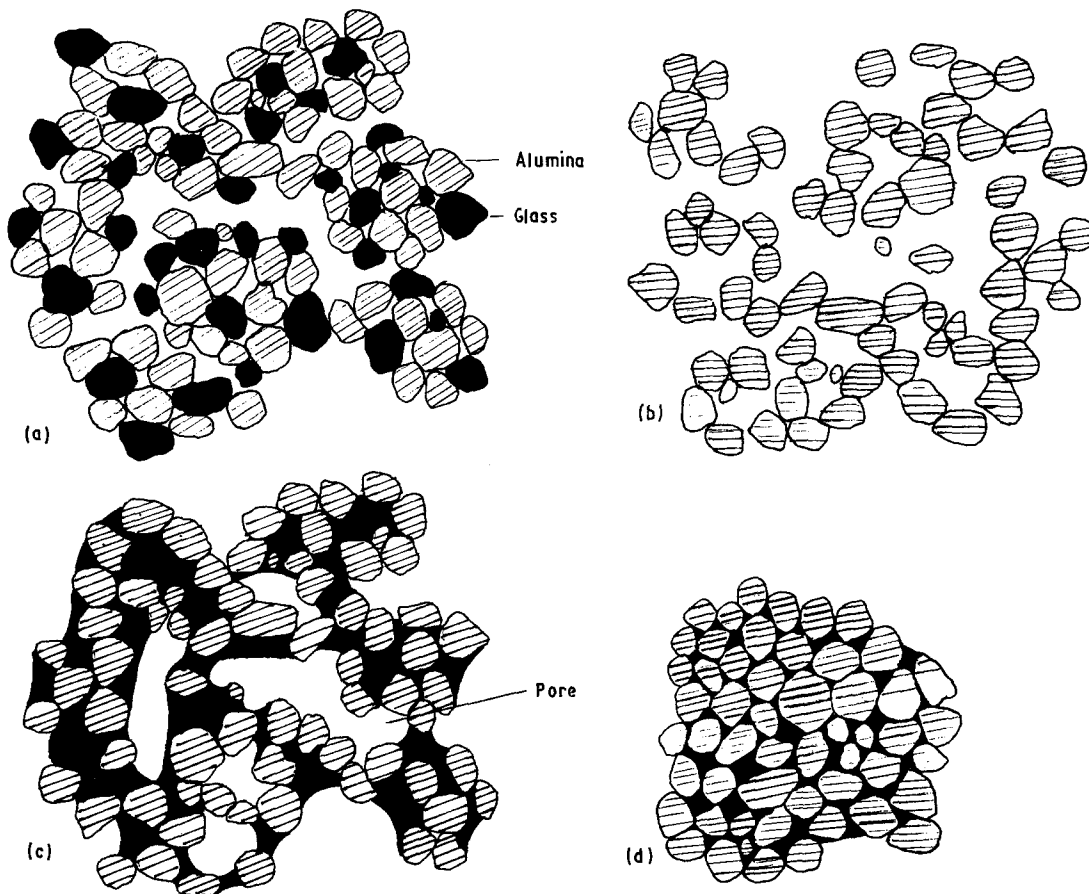


Figure 4 Schematic representation of the sintering process: (a) green glass-alumina composite, (b) skeleton structure of alumina without glass particles, (c) less densified structure, partially due to early onset of solid solid neck formation, and (d) well-densified structure.

(b) The early onset of the solid-solid contact, which forms the alumina network (Fig. 4b), at higher temperatures, slows down the densification due to rearrangement (Fig. 4d) [11]. Moreover, solid skeleton fragmentation is difficult due to the very high viscosity of the alumina-rich, liquid-glass phase.

There is a greater amount of glassy phase above 1000°C (Fig. 2) and the glassy phase is usually less dense than the crystalline phase. But in this case the contribution of the above factor to the lowering of densification above 1000°C is not appreciable because of the higher density of the glass (4.2 g cm^{-3}) compared to that of alumina (2.9 g cm^{-3}).

Detailed microstructural investigations are in

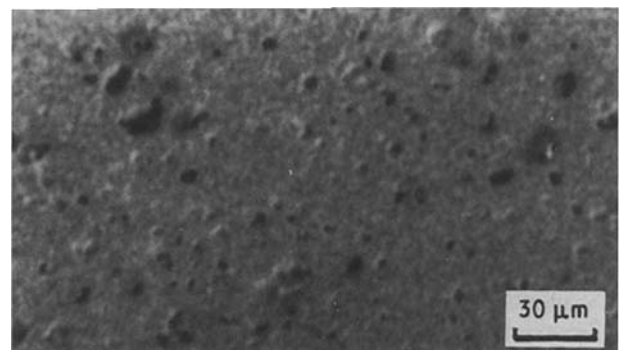


Figure 5 SEM micrograph of polished surface of the glass-alumina composite sintered at 1000°C.

progress with aluminas of different particle size in order to find out the mechanism of secondary rearrangement due to grain boundary penetration and particle disintegration. A typical polished-surface SEM micrograph of a sample sintered at 1000°C is given in Fig. 5.

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*Received 7 July
and accepted 14 September 1989*