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Ceramics International 33 (2007) 911–914

CERAMICS
INTERNATIONAL

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Low-temperature fabrication of steatite ceramics with boron oxide addition

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Received 15 December 2005; received in revised form 17 January 2006; accepted 14 February 2006

Abstract

Steatite ceramics have been fabricated by using coarse starting materials such as talc, clay, and barium carbonate with addition of boron oxide (B_2O_3). B_2O_3 has been found to be a useful flux to densify MgO–Al_2O_3–SiO_2–BaO (MASB) powders. The steatite ceramic with a relative density of 97% was obtained at a sintering temperature of 1200 °C. The temperatures needed to densify MASB powders decrease with increase of B_2O_3 addition. For the same sintering temperature, as the amount of B_2O_3 addition increases, the crystallization rate of protoenstatite increase. Large pores and visible cracks were seen in B_2O_3-free samples, while pore elimination and enhancement of densification for B_2O_3-added steatites were promoted by the viscous flow due to B_2O_3. Hence, nearly dense ceramics were obtained for use in high temperature electrical applications.

Keywords: Steatite; Talc; Boron oxide; Densification

1. Introduction

As a general class of dielectrics, steatite ceramics are inorganic materials made from natural raw materials containing predominantly magnesium silicate and can be considered in place of alumina as a cost-effective way to meet performance requirements. They are easier to form and fire at lower temperatures, making them more economical to produce. The steatite ceramics are widely used in electronics, electrical engineering, radio electronics, and other fields owing to their low dielectric losses, high temperature resistance, and high mechanical strength [1,2]. On the other hand, steatite has been proven in innumerable electro-technical applications such as halogen bulb holders, NH-fuses, element formers and casings for thermostats. Other applications include heating element holders, stand off insulators, interlocking insulating beads, split bush insulators, lamp bases and caps, regulator parts [3].

The steatite ceramics are formulated from a natural raw material containing magnesium silicate, known as talc together with additions of clay and flux. The type of flux influences the electrical characteristics and leads to the following differentiation: steatite for low-frequency, normal steatite, as well as special steatite with low loss factor. Typical steatite materials have the following analysis: 58–65% SiO_2, 26–32% MgO, 3–6% Al_2O_3, 1.3% Na_2O (normal steatite) to 7–9% BaO (low-loss steatite). Normal steatite is especially well suited for manufacturing complicated pressed pieces. Seven to ten percent BaO is added for low-loss steatite. Its structure is mainly crystalline magnesium silicate ($MgSiO_3$) with a glassy portion of 25–45%. It has an impervious material structure that has good mechanical strength, and low dielectric loss. It makes excellent insulators for radio frequency applications, and exhibits high volume resistivity up to 1000 °C [4,5].

The steatite ceramics are produced by dry pressing, extrusion, casting and semi wet pressing using talc (about 80–90%), plastic clay (about 5–10%) and a flux melting agent (about 5–10%). During the preparation of the mass, low quantities of clay and/or barium carbonate are added. The clay serves to improve the formability of the mass and its processability. The barium carbonate acts as a flux-melting agent during the sintering process, i.e. it supports the development of a melting phase. The material is fired at around 1400 °C and it is formed by crystallization, fusion and dissolution during the vitrification. At the end of sintering, the material consists of about 70% of crystalline $MgSiO_3$ (so-called protoenstatite) and of about 30% of glass phase [6].

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doi:10.1016/j.ceramint.2006.02.001

A method of improving the sinterability of steatite ceramics made from natural coarse materials is the addition of sintering aids. In the present study, it was aimed to obtain dense low-loss steatite ceramics at lower sintering temperatures from talc, clay and barium carbonate without the necessity of very fine particles. For this purpose, boron oxide (B_2O_3) was used as a sintering aid due to its low melting point and less harmful effect on the insulating characteristics than other sintering aids [7,8].

2. Experimental procedure

Talc, clay, and barium carbonate (99.5%) were used as starting materials. The chemical analysis of starting materials, performed by X-ray fluorescence spectrophotometer (ARL, XRF-8660), is given in Table 1. The particle size distribution of starting materials determined with a Mastersizer 2000 particle sizer (Malvern Instruments Ltd.) indicated that the mean particle sizes of the talc, clay, and barium carbonate were 40.75, 12.11, and 10.42 μm, respectively (Fig. 1).

Talc (84 wt.%), clay (8 wt.%), and barium carbonate (8 wt.%) were weighed according to a commercial grade low-loss steatite composition of a domestic producer. 0.25, 0.75, and 1.5 wt.% of B_2O_3 were added to the mixture in the form of boric acid (H_3BO_3), respectively, and indicated as MASB5, MASB10, and MASB20. These powders were wet mixed and milled in deionized water for 2 h using alumina balls in a plastic container. In order to compare results, a steatite powder (MASB0) was also produced without B_2O_3 addition using the same raw materials. After drying the resulting slurry at 80 °C for 24 h and re-milling of the dried mixture, it was uniaxially pressed at 75 MPa into pellets approximately 15 mm diameter. Afterwards, the green compacts were sintered at a temperature in the range 1000–1200 °C for 2 h with a heating and cooling rate of 300 °C/h in an electrical furnace (Alser Teknik).

The crystalline structures of sintered MASB steatite ceramics were investigated using X-ray diffraction (XRD) patterns. Microstructural studies on metallographically prepared samples were carried out using a JEOL 6500 scanning electron microscope (SEM). The densities of sintered samples were measured as a function of sintering temperature and B_2O_3 addition by the liquid displacement method based on Archimedes's principle.

Table 1
Chemical analysis of talc and clay

	Composition (wt.%)	
	Talc	Clay
Oxide		
MgO	32.0	–
Al_2O_3	–	26.2
SiO_2	61.5	64.5
CaO	0.8	–
Fe_2O_3	0.5	0.3
Na_2O	–	0.1
K_2O	–	0.1
Ignition loss[a]	5.1	8.5

[a] Ignition loss was determined at 1100 °C.

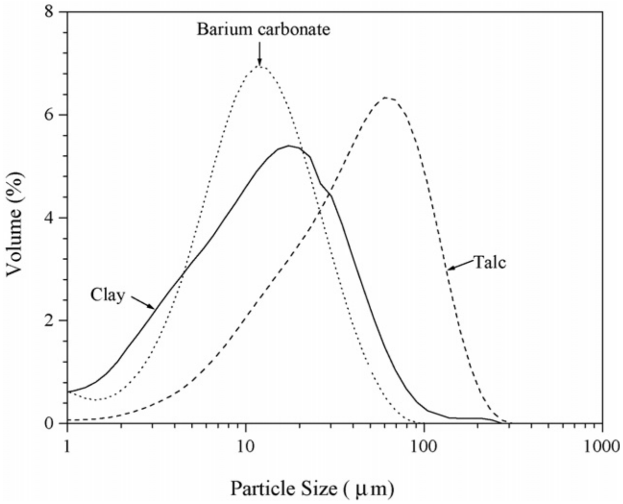

Fig. 1. Particle size distribution of raw materials used in this study.

3. Results and discussion

The X-ray diffraction patterns of MASB steatite powders with different amounts of B_2O_3 addition and sintered at temperatures between 1000 and 1200 °C are shown in Fig. 2. After sintering at 1000 °C, protoenstatite phase was observed with quartz, aluminum silicate, and barium aluminum silicate phases for the samples (Fig. 2a). The crystallization of protoenstatite phase increased with amount of B_2O_3 addition. On heating, talc decomposes and gives a mixture of fine-grained protoenstatite crystals in a silica matrix at about 1000 °C. As similar to talc, clay decomposes at 980 °C to form fine-grained mullite in a silica matrix. Addition of clay to talc decreases the liquidus temperature. For 90% talc–10% clay composition, about 30% liquid is formed abruptly at the liquidus temperature of 1345 °C [1]. The steatite compositions with clay and barium carbonate as an added flux frequently precipitate some crystalline phases such as aluminum silicate and barium aluminum silicate on cooling. There is usually a residual glassy matrix (viscous silicate liquid) which is required for firing to form a vitreous densified ceramic [1]. Upon heating, in dehydroxylated talc grains form micropores, which appear as the temperature increase. This process is accompanied by production of free silica or quartz, as indicated in Fig. 2a. The quartz grains are undesirable constituents of steatite and must be eliminated [11].

The crystallization rate of protoenstatite phase and appearance of other crystalline phases were also dependent on sintering temperature (Fig. 2b). Highest crystallization rate of protoenstatite was obtained with MASB20 samples. After sintering at 1100 °C for MASB20 samples, aluminum silicate and barium aluminum silicate dissolved into the glassy matrix and disappeared. Although the peak intensities of protoenstatite phase increased, there was still a minor quartz phase. However, the crystallization rate of protoenstatite reached a maximum

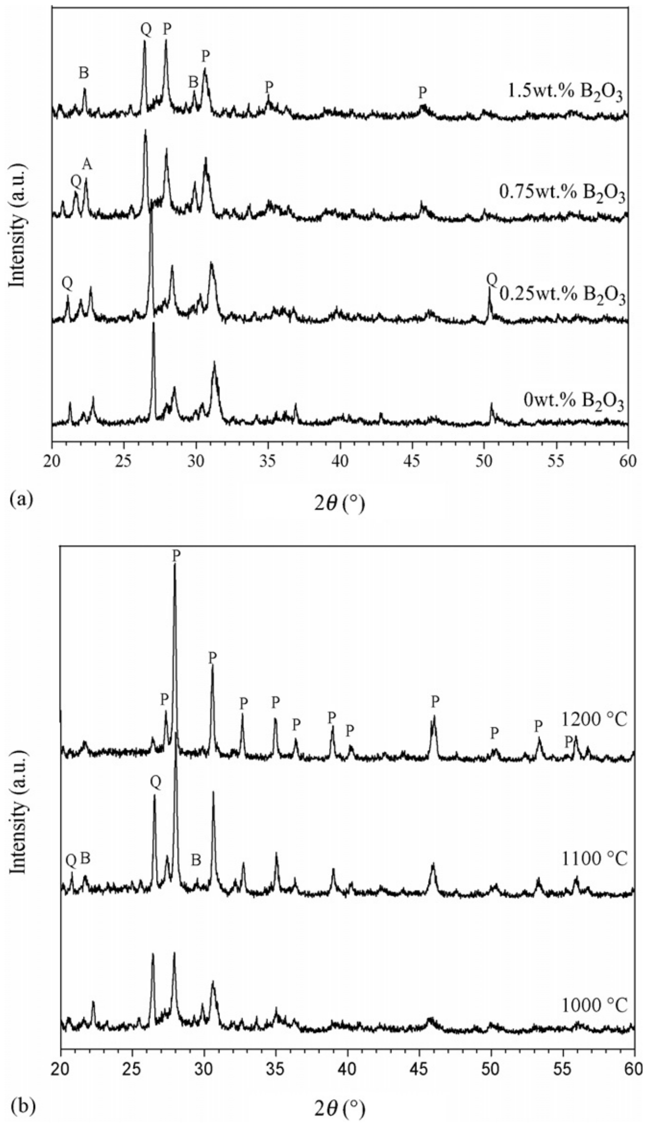

Fig. 2. X-ray diffraction patterns of MASB steatite ceramics: effects of (a) B_2O_3 addition, and (b) sintering temperature (P, protoenstatite; Q, quartz; B, barium aluminum silicate; and A, aluminum silicate).

value at a sintering temperature of 1200 °C. At this temperature, quartz phase disappeared and only a single protoenstatite crystalline phase was obtained. The process of protoenstatite crystallization in talc generally starts at temperatures above 1250 °C and fully develops at 1420 °C [11]. It is clear that the addition of B_2O_3 accelerates the crystallization of protoenstatite.

Densification of MASB steatite ceramics, added with different amounts of B_2O_3 and sintered at different temperatures is shown in Fig. 3. The sinterability of MASB0 steatite was poorer than those of B_2O_3-added ceramics. The density increased with sintering temperature, as expected. As the content of B_2O_3 increases, a much easier densification of the steatite compositions was evidenced in Fig. 3. MASB0 specimens had a density of around 75% of theoretical density at 1000 °C but the density of the specimens did not change significantly with sintering temperature even at 1200 °C.

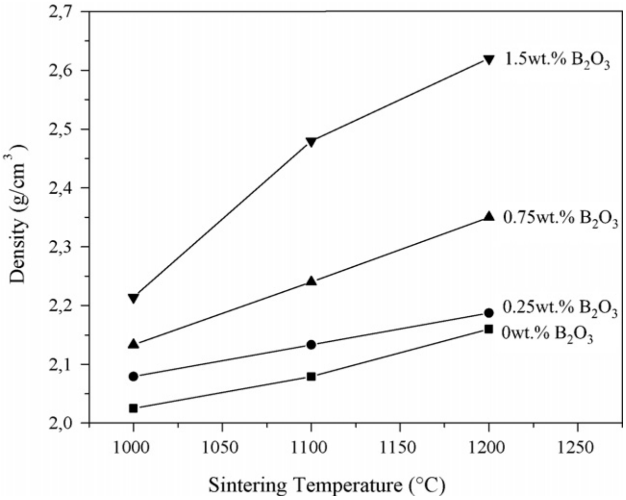

Fig. 3. The densities of MASB steatite ceramics as a function of sintering temperature and B_2O_3 amount.

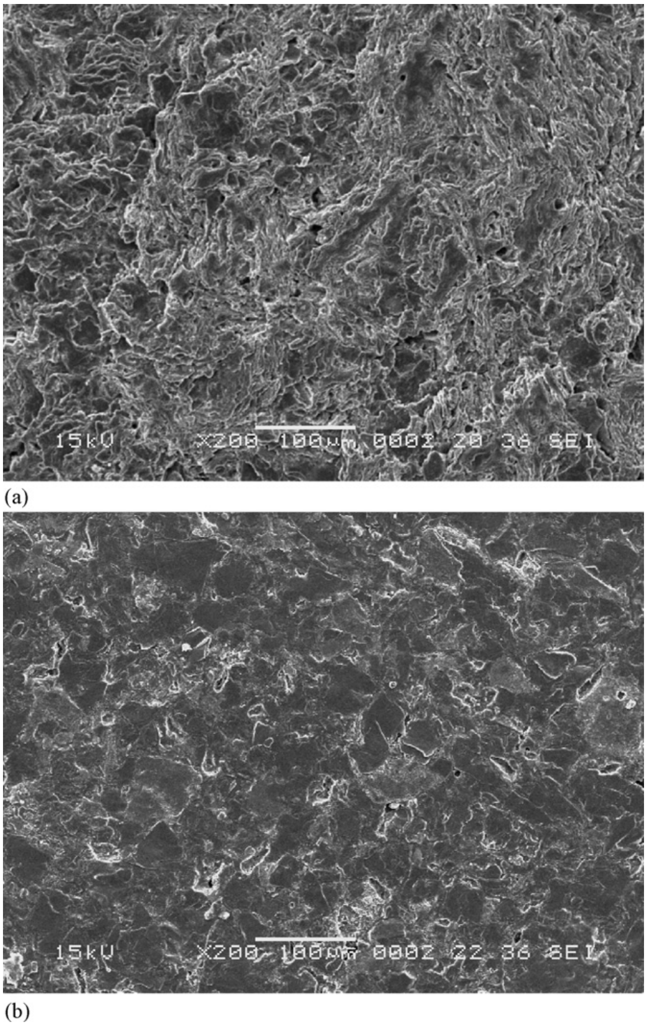

Fig. 4. The SEM micrographs of (a) MASB0 and (b) MASB20 samples sintered at 1200 °C.

MASB5 specimens had a density of around 77% of theoretical density at 1000 °C and reached to 81% of the theoretical density at 1200 °C, while the relative densities of MASB10 specimens were 83% and 87% at 1000 °C and at 1200 °C, respectively. On the other hand, highest values were obtained in MASB20 specimens which had a relative density around 92% and 97% for 1100 and 1200 °C temperatures. In the present study, high-density values of around 2.62 g/cm^3 (around 97% of theoretical for MASB20 samples) were obtained at 1200 °C by incorporating B_2O_3 as the sintering aid in spite of starting with raw materials having larger particle sizes.

SEM micrographs of MASB0 and MASB20 samples sintered at 1200 °C are shown in Fig. 4. Large pores were easily seen in Fig. 4(a) for samples without B_2O_3 addition. As natural process, visible cracks also developed in steatite body. As shown in Fig. 4b, pore elimination and enhancement of densification for MASB20 samples were promoted by the viscous flow B_2O_3 addition, and hence a nearly dense ceramic was obtained. Easier densification of MASB specimens with B_2O_3 addition may be due to the flux effect of B_2O_3, which enhances the densification during sintering. Because of the low melting point of B_2O_3, the procedure of sintering MASB ceramics would be a reactive liquid-phase sintering process. With liquid formation, there was a densification, even at low sintering temperatures such as 1000 °C, due to capillary force exerted by the liquid on the particles, while actual sintering temperature was around 1400 °C for steatite ceramics [9–11].

4. Conclusions

By using B_2O_3 additions, a dense steatite ceramic can be obtained at a lower sintering temperature than that of industrial practice. This will have a potential for manufacturing of steatite ceramics using B_2O_3 addition from talc, clay, and barium carbonate with coarse particle sizes. There is only protoenstatite crystalline phase in 1.5% B_2O_3-added steatite sintered at 1200 °C. The addition of B_2O_3 accelerates the crystallization of protoenstatite phase. The temperatures needed to densify MASB steatite ceramics decrease with increase in the amounts of B_2O_3 addition.

Acknowledgements

The author gratefully acknowledges Dr. Z. Aslanoğlu and Dr. Y. Karakaş of Erdemir Group, Turkey, for their valuable discussions in this study.

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