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The microstructure, coefficient of thermal expansion and flexural strength of cordierite ceramics prepared from alumina with different particle sizes

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ABSTRACT

Cordierite ceramics were produced from alumina with 5 and 0.65 μm particle sizes or AlOOH and talc, clays and feldspar, to determine the influence of the alumina particle size on the microstructure, coefficient of thermal expansion (CTE) and flexural strength (FS) of the ceramics. After sintering at 1300 °C the ceramics made from 5- μm -sized alumina consisted of cordierite, glass, quartz, mullite and alumina, and had the highest density, FS and CTE. The alumina grains act as inclusions, from which the trajectories of the cracks were deflected or terminated, which increases the FS and CTE. The ceramics from sub-micrometre-sized alumina or AlOOH contained a negligible amount and no alumina, respectively, together with other phases. This is reflected in the low CTE and FS. The cordierite ceramic with the lowest CTE of $\sim 2.0 \times 10^{-6} \text{ K}^{-1}$ and a high FS of 100 MPa was prepared from the 0.65- μm -sized alumina particles.

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1. Introduction

The major phase of the cordierite ceramics is the mineral cordierite with the nominal composition $2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$. It crystallises in the hexagonal crystal structure. It is characterised by large anisotropic coefficient of thermal expansion (CTE) of $2.5 \times 10^{-6} \text{ K}^{-1}$ along a and b axes and $-0.9 \times 10^{-6} \text{ K}^{-1}$ along c-axis, which generates thermal stresses during heating or cooling [1,2]. The reported values of CTE for the cordierite ceramic range from about $1\text{--}5.7 \times 10^{-6} \text{ K}^{-1}$ in the temperature range from room temperature to 600 °C and depend largely on the phase composition and amount of phases in the cordierite ceramic [3–5]. Due to the low CTE and therefore superior thermal shock resistance, a high flexural strength of about 50 MPa [3] as well as chemical and thermal stability at high temperatures the cordierite ceramic has long been utilised as a refractory material, a catalyst carrier in automobile exhaust systems and a filter for molten metals. In addition, it is widely used in electrotechnics for various applications such as a holder for electrical heaters. Cordierite has a dielectric constant of 6 and a CTE value that is significantly lower than that of alumina. Alumina has a dielectric constant of 9 and a CTE of $8 \times 10^{-6} \text{ K}^{-1}$.

Thus the cordierite ceramic has the potential to replace alumina substrates in microelectronic packaging [6,7], as well as to substitute the common alumina ceramic filler with cordierite particles in low-temperature co-fired ceramic tapes to decrease the dielectric constant of the fired tapes [8].

The processing of single-phase cordierite ceramics is demanding since it requires a temperature as high as 1430 °C [5]. In order to process cordierite ceramics at lower temperatures and with a large amount of cordierite phase, various methods were used such as coprecipitation [5,9,10], the Pechini method [11], sol-gel [12–14] or by re-crystallisation of the molten glasses with a nominal cordierite composition [15,16]. However, these methods require either rather expensive precursors or, in the case of glass ceramics, high melting temperatures of around 1600 °C.

The most widely used method for the large-scale, economic, processing of cordierite ceramics is solid-state synthesis. Cordierite ceramics were synthesised from pure oxides or nitrates [8,17] or from natural raw materials such as talc and clays [18–20]. After sintering, typically in the temperature range between 1300 and 1400 °C, the ceramic contains cordierite, but also various secondary phases, such as a glass phase, alumina, spinel, mullite or silica. The glass phase promotes the densification during firing; however, the other secondary phases are not desirable as they increase the CTE of the cordierite ceramics [5,19]. In attempting to improve the thermal, mechanical and electrical properties of the cordierite ceramic,

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Table 1
Chemical compositions of clay and feldspar (wt.%).

Oxide	Clay	Feldspar
SiO ₂	57.45	66.9
Al ₂ O ₃	32.5	17.4
Fe ₂ O ₃	1.5	0.04
MgO	0.3	0.01
CaO	0.15	0.02
Na ₂ O	0.1	0.7
K ₂ O	1.9	14.2
TiO ₂	1.6	0.06
BaO	–	0.32
loss on ignition	4.5	0.35

Table 2
Denotation of samples.

Source of alumina/particle size	Supplier	Powder	Ceramic
Al ₂ O ₃ /5 μm	Nabaltec	AO5-P	AO5
Al ₂ O ₃ /0.65 μm	MTI	AO0.65-P	AO0.65
AlOOH/1.8 μm	Nabaltec	AOH-P	AOH

the effort has focussed on processing a dense cordierite ceramic and eliminating the undesired secondary phases. The influence of various compounds such as TiO₂ [21], CeO₂ [22], B₂O₃ [10], and Na₂B₄O₇ [5] on the processing and properties of the cordierite ceramic were studied. Even though the use of these compounds is reflected in an improved density and/or the mechanical properties of the ceramic, the CTE of the resulting cordierite ceramic increased. It was shown that the intensive milling of raw materials, e.g., talc, kaolin and boehmite, promotes the formation of cordierite and resulted in a dense ceramic with a large amount of cordierite phase after sintering at 1300 °C. The CTE of the resulting ceramic was not reported [20].

In this paper we report on the synthesis and properties of a cordierite ceramic prepared from talc, kaolin, clay, feldspar and alumina. The talc, kaolin, clay and feldspar are natural, low-cost, raw materials, while the alumina is a synthetic source in the form of Al₂O₃ (corundum) and AlOOH (boehmite). The aim of this paper is to examine the influence of alumina with different particle sizes and the influence of different sources of alumina on the characteristics of cordierite ceramics. The phase compositions and the microstructures of the ceramics were investigated and correlated with their thermal and mechanical properties.

2. Experimental

The natural raw materials, i.e., talc, kaolin, clay and feldspar, and the synthetic alumina were mixed together in a stoichiometry corresponding to the nominal composition of the cordierite 2MgO·2Al₂O₃·5SiO₂. The chemical compositions of the clay and feldspar are given in Table 1. The alumina was added as corundum Al₂O₃ with particle sizes of 5 μm (denoted AO5-P) and 0.65 μm (denoted AO0.65-P) or as AlOOH (denoted AOH-P). The addition of AOH-P was calculated for the required amount of alumina. Three sets of samples were prepared as listed in Table 2.

Suspensions were prepared by dispersing 60 wt.% of AO5-P, 60 wt.% of AOH-P, and 40 wt.% of AO0.65-P in water with a deflocculant based on polyacrylic acid. Afterwards, the raw materials were wet milled for 16 h in a planetary mill with alumina balls and then dried in plaster moulds at room temperature to a water content of 24 wt.%, suitable for extrusion. Test samples in the form of circular bars were prepared on a laboratory vacuum-extrusion machine (Netzsch, V5). The samples dried at 105 °C for 4 h are denoted throughout the text as “as-prepared”. One set of samples, i.e., AO5, AO0.65 and AOH, were sintered in a laboratory electric furnace (Naber N20/14) at 1300 °C for 2 h. The heating rate was 3 K/min.

A second set of samples, i.e., AO0.65 and AOH, were isostatically pressed with a pressure of 300 MPa and sintered under the conditions already described. These sintered samples are denoted as AO0.65 I and AOH I.

The particle size and the particle-size distribution of the powders were determined with a static light-scattering particle size analyser (Microtrac S3500, Montgomeryville, PA, USA). The results were derived from the volume particle size distribution (d_v).

Thermogravimetric (TG) and differential thermal analyses (DTA) of the AOH-P were performed simultaneously using a Netzsch STA 429 analyser (Hann, Germany). The analysis was performed in PtRh crucibles. The samples were heated from 25 °C to 1100 °C with a heating rate of 5 K/min in a flow of synthetic air.

Bulk densities of the ceramics were measured with Archimedes' method using deionized water as the immersion medium. Three sets of samples were used for the bulk-density determination.

The linear thermal expansion and the corresponding curves of the CTE were measured on a ceramic with a diameter of 6 mm and a length of 50 mm using a dilatometer (Netzsch DIL 402EP, heating rate 5 K/min) in the temperature range 30–980 °C.

The flexural strength was determined on a ceramic with a diameter of 10 mm and a length of 120 mm at room temperature using a three-point bending strength tester (Netzsch 401/3) with a 100-mm span width. The test was run at increased load from 0 to 400 N with a step of 4 N/s until the sample is fractured. The flexural strength of the sample was calculated using the relationship:

$$\sigma = \frac{F_{max} * L}{\pi R^3}$$

where σ is flexural strength, F_{max} load at fracture, L span width, R radius of the sample. Seven samples of each composition were measured. The maximum and minimum values were eliminated and, thus, the flexural strength was calculated as an average of five measured values.

X-ray powder diffractions (XRD) were recorded with a diffractometer (PANalytical, X'Pert PRO MPD, Almelo, The Netherlands) at room temperature using Cu–K α radiation. The data were collected in the 2 θ range from 15 to 36°, in steps of 0.017°, with an integration time of 200 s. The crystalline phases were identified using the PDF-2 database [23]. Microstructures were observed and analysed with scanning electron microscopes (SEM, JSM-5800 and JSM 7600F, both Jeol, Tokyo, Japan) equipped with an energy-dispersive X-ray spectrometer (EDXS). Samples in a cross-sectional orientation were prepared with grinding and polishing using standard metallographic technique.

The indentation tests were carried out on a Mitutoya MV8 H2 testing machine (Mitutoya America Corporation, Aurora, Illinois, USA). A diamond pyramid indenter with an angle of 136° between opposite faces (Vickers indenter) was pressed into the polished surfaces of the samples under a load of 10 N. After the tests, the indents were observed with the SEM in order to determine the crack propagation in the investigated sample.

3. Results and discussion

SEM images and the particle size distributions (PSD) of the AO5-P and AO0.65-P powders are shown in Fig. 1a and b, respectively. From the PSD it is evident that the AO5-P powder contains particles ranging from about 0.5–25 μm, which is in agreement with the SEM image (Fig. 1a). The median particles size is 3.76 μm. The PSD of the AO0.65-P powder is narrow with the median particle size of 0.65 μm (see inset in Fig. 1b). The particles with a relatively uniform size from a few hundreds of nm to about 2 μm are also evident in the SEM image of the AO0.65-P powder (Fig. 1b). Both powders crystallise in a rhombohedral structure (corundum, PDF 75-1865).

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