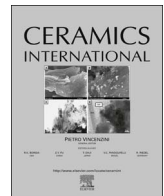




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Thermo-mechanical and high-temperature dielectric properties of cordierite-mullite-alumina ceramics

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ABSTRACT

Heterogeneous ceramics made of cordierite (55–56 wt%), mullite (22–33 wt%) and alumina (23–11 wt%) were prepared by sintering non-standard raw materials containing corundum, talc, α -quartz, K-feldspar, kaolinite and mullite with small amounts of calcite, cristobalite and glass phases. The green specimens prepared by PVA assisted dry-pressing were sintered within the temperature range of 950–1500 °C for different dwelling times (2–8 h). The effects of sintering schedule on crystalline phase assemblage and thermomechanical properties were investigated. The sintered ceramics exhibited low coefficients of thermal expansion (CTE) ($3.2\text{--}4.2 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$), high flexural strength (90–120 MPa and high Young modulus (100 GPa). The specimens sintered at 1250 °C exhibited the best thermal shock resistance ($\Delta T \sim 350 \text{ }^\circ\text{C}$). The thermal expansion coefficients and thermal shock resistance were studied using Schapery model, the modelling results implying the occurrence of non-negligible mechanical interactions between the phases in bulk. The dielectric properties characterized from room to high temperature (RT–HT, up to 600 °C) revealed: (i) noticeable effects of sintering schedule on dielectric constant (5–10) and dielectric loss factor ($\sim 0.02\text{--}0.04$); (ii) stable dielectric properties until the failure of the electrode material. The thermomechanical properties coupled with desirable dielectric properties make the materials suitable for high density integrated circuitry or high temperature low-dielectric materials engineering.

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1. Introduction

Cordierite ceramics are characterized by low coefficients of thermal expansion ($1.5 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$), high thermal shock resistance ($\Delta T = 325 \text{ }^\circ\text{C}$) [1], relatively high mechanical flexural strength ($> 100 \text{ MPa}$) [2] and high Young modulus ($> 100 \text{ GPa}$) [3]. Such thermo-mechanical properties make cordierite ceramics attractive for kiln furniture, catalyst supports [4,5], refractory materials in metal foundry for filtering during casting, and other industrial applications [6]. The low values of dielectric constant and loss factor, and the enhancement of fracture toughness, especially when mullite needles are formed, make cordierite ceramics ideal materials for electronic packaging [7–11].

Monolithic cordierite has a molar ratio of $2\text{MgO}:2\text{Al}_2\text{O}_3:5\text{SiO}_2$ with chemical formula $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ [12]. This corresponds to a composition of 13.7 wt% MgO, 34.9 wt% Al_2O_3 and 51.4 wt% SiO_2 . In General, cordierite has narrow crystallization temperature, above (1250 °C) [13,14] and below 1000 °C μ -cordierite formed first and then α -cordierite formed above 1200 °C [9]. Due to high

costs of chemical grade precursors non-stoichiometric cordierite-based ceramics are often synthesized from natural raw materials such as clays and oxide powders [15,16]. The consequent formation of corundum and mullite upon sintering is desirable to further improve the mechanical properties [17–20]. Cordierite-based materials exhibit low values of dielectric constant and loss tangent that confer them a great potential for microelectronic applications, contributing to the miniaturization trend in terms of integrated circuits (IC). They enable the capacitance and the charge collected at the capacitor to be dramatically increased. Cordierite-mullite or cordierite glass ceramics are promising packaging materials with tunable dielectric constant at room temperature, $\epsilon_r = 5\text{--}5.2$ at 1 MHz and better mechanical properties [7–10,21]. The thermal expansion coefficients of cordierite-based glass-ceramics could be fine-tuned to that of silicon ($3.6\text{--}4.8 \times 10^{-6} \text{ }^\circ\text{C}^{-1}$) currently in IC, by doping with 1.5–3.0 wt% ZnO [22], thus minimizing the thermal stresses induced by the rise in temperature ($\sim 1000 \text{ K}$) due to air friction [22,23].

Cordierite ceramics are also commonly used in the aerospace industry as radomes for missiles or vehicles travelling at high speed ($\text{Mach} > 2$) to protect their radar antennas from the environment [24,25]. In order to keep the quality of communication and guidance a good radome material should meet following

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standards: easy to fabricate, low dielectric permittivity and low loss factor at RT and elevated temperatures, high strength and high modulus to prevent the thin-walled radome from buckling. In this context, cordierite has been regarded with alacrity because it suitably combines a set of interesting properties: (i) low k value; (ii) good mechanical strength, and thermal and chemical stability [23,24]; (iii) low fabrication costs, especially when starting from natural raw-materials.

It is noteworthy that despite the numerous literature reports on dielectric properties of ceramics, high temperature data about dielectric constant and loss factor are only available so far for Si_3N_4 -based and glass ceramics commercial products [24]. Also, only a few works dealing with low-cost fabrication of cordierite-mullite ceramics investigated the relationship between processing and dielectric properties, and none of them evaluated the dielectric properties change as temperature increases [7–10,22]. This has been attempted in a single report about stoichiometric cordierite ceramics prepared by the polyacrylamide gel method [9]. Therefore, it is timely to provide an insight on elevated temperature dielectric properties of cordierite ceramics prepared from natural raw materials to assess their suitability for advanced engineering applications. Sintering from glass frit prepared at high temperature, sometimes in the presence of additives in order to reduce sintering temperature, is the most common route to obtain cordierite ceramics. Complete densification within the 900–950 °C temperature range was reported for a glass frit (13 wt% MgO, 27 wt% Al_2O_3 and 60 wt% SiO_2) powder having an average particle sizes of $\sim 2 \mu\text{m}$ by adding P_2O_5 and B_2O_3 ($\text{B}_2\text{O}_3 + \text{P}_2\text{O}_5 < 5 \text{ wt}\%$) [26]. Chen [22] found that batch melting temperature, glass transition temperature, and crystallization temperature of stoichiometric cordierite prepared from high-purity oxides all decreased with increasing added amounts of ZnO (1.5–3.0 wt%). Hu et al. [27] also disclosed improved sintering ability by using glass compositions with excess MgO and less Al_2O_3 or by adding sintering aids such as P_2O_5 and B_2O_3 .

Non-standard low cost natural raw materials that lead to heterogeneous cordierite-containing ceramics are used in the present work. The aim is to study the sintering behaviour, characterize the mechanical and electrical properties, and determine the optimal processing variables. The thermo-mechanical and dielectric properties at high temperature of the obtained materials are also characterized for the first time.

2. Experimental procedure

2.1. Materials and preparation of testing specimens

A commercial spray dried powder of the precursor cordierite-mullite-alumina ceramics was supplied by Rauschert (Rauschert Portuguesa SA, São Domingos de Rana, Portugal). The composition of the starting non-stoichiometric powder has been analysed and reported earlier [17]. A PVA (Mw 146,000–186,000, 99% hydrolysed, Sigma Aldrich, Germany) solution (2 wt%) was sprayed onto 200 g of granulated powder in a rotary mixer in the required volume in order to obtain an added amount of 0.85 wt% of PVA relative to the mass of solids. Since the hydration level of the powder is a critical factor in dry pressing, the wet mixture was partially dehydrated. The optimal hydration degree was determined in trial-and-error dry pressing experiments to obtain green parallelepiped bar specimens of $50 \times 5 \times 5 \text{ mm}$ (length - height - width) for flexural strength measurements. Very fragile specimens were obtained for too low water contents, while an excessively wetted powder resulted in specimens sticking to the inner surface of the mould. The optimal water content was found to be 2.5 wt%. Therefore, the weight loss of the powder wetted with the PVA

solution was carefully monitored during drying in an oven at 80 °C. It was found that a drying period of 24 h under these conditions allowed to achieve the final targeted humidity of 2.5 wt%. The powder was passed through a 500 μm sieve to obtain a uniform particle size distribution to consolidate the green bar specimens. Sintering was carried out in air within the temperature range from 950–1500 °C, using a heating rate of $5 \text{ }^\circ\text{C min}^{-1}$, dwelling times varying from 2–8 h, followed by natural cooling to RT inside the furnace.

2.2. Characterizations techniques

The crystalline phase analysis of sintered samples was performed using a XRD (Philip PANalytical X'pert MPD, Netherlands) from 5–80° with 0.02° step. The flexural strength was characterized by three-point bending (Shimatzu Universal Machine EZ Graph AG25 TA, Japan) using a strain rate of 0.5 mm min^{-1} . Young's modulus was measured by Impulse Excitation method according to ASTM standard E1876 [28]. Thermal expansion coefficients were measured with a dilatometer (Bahr, DIL801L, Germany).

Since the sintered products have complex compositions, the true density values of powdered samples were measured by Pcnometry method (Quantachrome, USA). Thermal shock resistance was determined by measuring the retained flexural strength of ceramic samples heated to 900 °C for 2 h followed by quenching in water at room temperature according to ASTM standard C1525–04 [29]. The criteria in [1] were used to determine the critical temperature at which the flexural strength decreases by 40%. The thermal shock resistance (TSR) modelled by thermal induced stresses was calculated according Hasselman et al. [30]:

$$\sigma_{thermal} = \frac{\psi E \alpha \Delta T}{1 - \nu} \quad (1)$$

where ΔT is the temperature difference between the temperature to which the sample was heated and the temperature of the quenching medium, α is linear coefficient of thermal expansion, ν is Poisson ratio, E is the young's modulus of the bulk and ψ is the stress reduction factor. The Poisson ratio values already reported [17] (0.270, for specimens sintered at 1250 °C for 2 h, and 0.267 for specimens sintered at 1350 °C for 2 h), experimentally determined by the Impulse Excitation technique were used in this study. For very rapid heat transfer, meaning quickly moving the specimen from the furnace to quenching water, ψ is defined as 1 [1]. From Eq. (1), the thermal shock resistance is worked out by equating thermal stress $\sigma_{thermal}$ to the flexural strength σ_f of specimen (ignoring the residual stress due to heterogeneous properties of the ceramics), hence:

$$\Delta T_c = \frac{\sigma_f(1 - \nu)}{E\alpha} \quad (2)$$

Eq. (2) enables estimating the thermal shock resistance from the linear coefficient of thermal expansion, Young's modulus, flexural strength, and Poisson ratio. The measurements were performed for 6 specimens from each sintering schedule.

The dielectric properties were measured using LCR (Inductance-Capacity-Resistance) metre from 10^3 Hz to 10^6 Hz . The specimen holder includes a ceramics tube (alumina) with thermal couple inserted into the tube as schematically shown in Fig. 1. Au electrodes were pre-coated on both surfaces by arc-deposition in vacuum ($< 10^{-3} \text{ Pa}$). The dielectric constant was calculated from the equation for parallel plate of circular cross section sample as:

$$\epsilon_r = \frac{Cd}{\epsilon_0 \pi r^2} \quad (3)$$

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