



Synthesis and characterization of single phase and low temperature co-fired cordierite glass-ceramics from perlite



Yongsheng Yu^{a,b}, Xiaofei Hao^a, Luxia Song^a, Zhen Li^{a,*}, Li Song^a

^a Faculty of Materials Science and Chemistry, China University of Geosciences, Wuhan 430074, PR China

^b College of Chemistry and Chemical Engineering, Xinyang Normal University, Xinyang 464000, PR China

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ABSTRACT

Single α -cordierite glass-ceramics for low temperature co-fired ceramics (LTCCs) substrates were prepared successfully from perlite as the basic raw material. The glass-ceramics could be highly densified above 850 °C and the α -cordierite being the only crystalline phase precipitated from the glass power sintered within 900–950 °C. With increasing sintering temperature or sintering holding time, μ -cordierite was effectively transformed into α -cordierite, and completely transformed into α -cordierite at 900 °C for 6 h. The synthesized single-phase α -cordierite glass-ceramics possessed low dielectric constants (5.6–6.1), low dielectric losses (0.0019–0.02), appropriate flexural strength (112–117 MPa) and suitable coefficients of thermal expansion ($2.56\text{--}2.91 \times 10^{-6} \text{ K}^{-1}$), which qualify them as a potential material for LTCC substrates application.

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1. Introduction

Increasing development in electronics industries, wireless communication electronic devices need to have light weight, miniaturization and multifunction. Fortunately, LTCCs technology with α -cordierite and α -cordierite-based glass-ceramics materials have been demonstrated to satisfy the rigorous requirements of the electronic devices mainly due to their beneficial characteristics, i.e. low dielectric constant, low dielectric loss, low CTE (close to Si or GaAs), high strength, high chemical durability, modest processing costs, and ability to be co-fired with electrodes of high conductive such as Cu (1083 °C), Ag (961 °C), or Au (1061 °C) below 950 °C [1–4].

Generally, the properties of α -cordierite-based glass-ceramics depend on the microstructures and the main crystalline phases determined by the addition of sintering aids as well as compositions of the parent glass [5]. Furthermore, when chemical composition of the parent glass comes to the stoichiometric ratio of cordierite, the completely densified α -cordierite glass-ceramics are difficult to be obtained at sintering temperature below 950 °C. This is due to high viscosity, narrow sintering range and the μ -cordierite formation of the cordierite-based glasses [6]. High densification of glass-ceramics contributes to obtain low dielectric loss and high mechanical strength. Nevertheless, thermal, mechanical and dielectric properties of the glass-ceramics were deteriorated by the formation of μ -cordierite that usually presented during

the synthesizing α -cordierite. Thus, in order to fabricate dense α -cordierite glass-ceramics at low temperature, it is very important to design glass composition, control the crystallization of the glass and add sintering aids reducing glass viscosity. Several studies have been successfully synthesized α -cordierite glass-ceramics at or below 950 °C using glass crystallization method [7–10]. Significantly, further researches were carried out to synthesized α -cordierite glass-ceramics using natural minerals as the starting raw materials. Currently, the natural minerals such as potassium feldspar [11–13], kaolin and talc [14–18] have been successfully used to fabricated fully dense α -cordierite glass-ceramics below 950 °C. It is considered an economical synthesizing technique. The reason is that the natural minerals are already containing sintering aids such as Na₂O, K₂O, CaO and P₂O₅, which can improve sintering ability of glass-ceramics and lower the sintering temperature of α -cordierite phase. This glass-ceramics prepared from cheap natural minerals were demonstrated to be promising LTCC substrates with low dielectric constant and loss, high flexural strength and matched CTE with Si, and so no sintering aids were required in the formulation.

In recent years, our research group has been successfully fabricated α -cordierite glass-ceramics for LTCC substrates using potassium feldspar [11–13]. Currently, the research of fabricating glass-ceramics for LTCC substrates using cheap perlite mineral has been barely reported. The aim of the present work is to investigate synthesizing highly densified α -cordierite glass-ceramics sintered at 850–950 °C using perlite as the starting raw material, and the sintering and crystallization behavior of glass-ceramics. The obtain single-phase α -cordierite of glass-

* Corresponding author.

E-mail address: zhenli@cug.edu.cn (Z. Li).

ceramics showed low dielectric constant and loss, high flexural strength and matchable coefficients of thermal expansion.

2. Materials and experimental procedure

The starting raw material for the preparation of cordierite glass-ceramics is perlite (Xinyang Shangtianti, Henan, China) and its chemical compositions are given in Table 1. Silica, alumina and magnesia (which purchased from Sinopharm Chemical Reagent Co., Ltd., AR) are added to compensate the formulation of the composition. The impurities of the alkali metal oxides as Na₂O and K₂O introduced by the perlite are network modifiers in the glasses. The detailed chemical composition of the glass is shown in Table 2. The mass content of perlite in the parent glass is 42 wt.%. The uniform mixture of raw ingredients according to Table 2 was melted in corundum crucibles in electrical furnace at 1550 °C for 5 h with a heating rate of 5 °C/min. Glasses in frit form were produced by quenching of the melts in cool distilled water, and pulverized to obtain glass powders by the wet ball-milling technique. To fabricate the bulk samples, the obtained glass powders with average particle sizes in the range of 1–3 μm were pelletized with 5% polyvinyl alcohol (PVA), and then pressed into disks with a diameter in 40 mm at a uniaxial pressure of 15 MPa by Hydrotex hand press. The samples were sintered within 850–950 °C for 6 h at the heating rate of 10 °C/min. Subsequently, the sintered samples were cooled down to room temperature in the furnace.

To research the thermal effect of the glasses, differential scanning calorimetry (DSC, SDT-Q600, TA) was used to record the thermogram of the obtain glass powder in the temperature range of 25–1200 °C with 10 °C/min heating rate. Alumina was used as the reference material. Apparent density of the glass-ceramic samples was calculated by Archimedes method using distilled water as medium. Absolute density of the glass-ceramics was measured by gas displacement principle using high purity nitrogen. The crystalline phases of sintered samples were determined by the X-ray diffraction (XRD, D8-FOCUS) with Cu Kα radiation, in the 5° ≤ 2θ ≤ 70°. The microstructure of sintered samples was characterized on fresh fracture surfaces by field emission scanning electron microscopy (FESEM, Hitachi S4800). Average linear thermal expansion coefficient was measured using a dilatometer (DIL 402C, NETZSCH) in air at a heating rate of 5 °C/min from room temperature to 600 °C. The dielectric properties of the glass-ceramics were measured using a precision impedance analyzer (4294 A, Agilent) from 40 Hz to 10 MHz at room temperature. The flexural strength of specimens (30 × 5 × 2 mm) was tested using three-point bending strength method at the cross-head speed of 4 N/s.

3. Results and discussion

3.1. DSC analysis

Crystallization behavior of amorphous phase is studied using DSC for the sample melted at 1550 °C for 5 h. Fig. 1 shows DSC result of the glass sample under a non-isothermal heat treatment at a constant heating rate of 10 °C/min. From Fig. 1, the glass sample exhibits two exothermic peaks. The glass transition temperature (T_g) implies the sintering phase, and its onset temperature is around 784 °C. The peak temperatures (T_p) of the two exothermic peaks are about 948 °C and 1055 °C respectively. The exothermic peak indicates the crystallization of a certain phase. The first exothermic peak can be assigned to the crystallization of μ-cordierite, and the second exothermic peak represents the crystallization of α-cordierite. In the sintering phase, prolonging holding time

results in crystallization and complete growth of the crystalline phase. These can be deduced from the results of XRD and SEM investigation.

3.2. XRD analysis

Fig. 2 shows the X-ray diffraction patterns of the glass-ceramics sintered at different temperatures. From Fig. 2, it is found that typical μ-cordierite patterns with considerable strong peak are observed in the sintered sample at 850 °C. Increasing the sintering temperature to 875 °C, the predominant crystalline phase in the glass-ceramics is still μ-cordierite, and only a few α-cordierite crystals produce simultaneously. With the increase of sintering temperature to 900 °C, μ-cordierite crystal phase in the glass-ceramics disappears, and only single crystal phase of α-cordierite exists in the glass-ceramics. Moreover, when sintering temperature increases to 925 °C and 950 °C, the crystallinity of the α-cordierite in the samples is slightly enhanced, as demonstrating the intensity increase of crystalline peaks in the XRD patterns.

For LTCC substrate, the α-cordierite (Indialite) is a promising material, and is the predominated crystalline phase deposited from the glasses. However, μ-cordierite is a semi-stable form which commonly crystallizes from glass samples sintered within 830–900 °C [7–9,19, 20]. Increasing the sintering temperature, addition of sintering aids or holding isothermally at a longer time will result in the transformation of the μ to the α crystalline phase [20]. In this composition, μ-cordierite is obviously precipitated from the glasses at low temperature. Fig. 3a shows XRD patterns of the glass-ceramics sintered different holding times at 875 °C and 900 °C. In Fig. 3a, partial enlarged details are at right side. Fig. 3b shows intensity of μ- and α-cordierite peaks count at (101) and (110) plane respectively as a function of sintering holding time at 875 °C and 900 °C. From Fig. 3a and b, with prolonging sintering holding time, the intensity of μ-cordierite characteristic peak (2θ = 25.90°) count at (101) plane firstly increases and then decreases, and finally disappears at 900 °C for 360 min. While the intensity of α-cordierite characteristic peak (2θ = 10.44°) count at (110) plane begins at 875 °C for 6 h and 900 °C for 180 min respectively, and then increases. These demonstrate that with increasing sintering temperature or sintering holding time, μ-cordierite is effectively transformed into α-cordierite, and completely transforms into α-cordierite at 900 °C for 6 h.

3.3. Microstructure analysis

Fig. 4 shows the micrographs of natural fractured surface of the glass-ceramics sintered at different temperatures. It can be seen that the glass-ceramics sintered at 850 °C have quite porous. With increasing the sintering temperature, the porosity of the glass-ceramics decreases evidently, and reaches minimum at 900 °C. Then sintering temperature rises to 925 °C and 950 °C respectively, the porosity of the glass-ceramics slightly increases compared to that of the glass-ceramics sintered temperature at 900 °C. Moreover, the porosity of glass-ceramics sintered at low temperature is only open, and the transformation of open porosity into closed starts from 875 °C, while the porosity is practically only closed in the range 900–950 °C. The non completed sintering results in the residual open porosity, while the crystallization of cordierite leads to the formation of closed porosity, named induced crystallization porosity [21,22]. These demonstrate that the parent glasses have excellent sinterability, which suggests highly densified glass-ceramics can be successfully manufactured. During the sinter-crystallization process of glass-ceramics, the samples get an efficient densification resulted from the viscous flow of the glass above the glass transition temperature (T_g) which enhances sintering. When crystallization occurs, due to the crystalline phase formation substantially increased the system's viscosity, sintering will be inhibited by glass viscous flow. As a result, induced crystallization porosity is reserved in the glass-ceramics. This result corresponds to the relative density of samples as shown in Fig. 5.

Table 1
Chemical compositions of perlite in wt.%.

Composition	SiO ₂	Al ₂ O ₃	K ₂ O	Na ₂ O	Fe ₃ O ₄	MgO	CaO	TiO ₂	Others
Perlite	70.65	12.84	4.28	2.56	0.75	0.44	0.97	0.08	7.43

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