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The phase transformation and thermal expansion properties of cordierite ceramics prepared using drift sands to replace pure quartz

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Abstract

Cordierite ceramics were prepared using drift sands as the raw material instead of traditional pure quartz through solid-state sintering within an enlarged temperature range between 1100 and 1300 °C. The effects of adding $Na_2B_4O_7$ on the phase transformation and thermal expansion coefficients were also investigated. The experimental results show that compared with pure quartz, drift sands have a lower softening temperature. Thus the sample prepared from drift sands shows a lower temperature for cordierite phase transformation, which was further reduced by adding 1.5 wt% $Na_2B_4O_7$. Moreover, with an increase of $Na_2B_4O_7$, the sintering temperature range was enlarged, and the amounts of the interphases of quartz and spinel as well as the thermal expansion coefficient for the obtained cordierite ceramics were decreased.

Keywords: C. Thermal expansion; D. Cordierite; Drift sands; Phase transformation

1. Introduction

Cordierite ceramic ($Mg_2Al_4Si_5O_{18}$) is widely used in the preparation of kiln furniture, catalytic carriers, high-temperature filters and integrated circuits because of its excellent thermal shock resistance, low dielectric constant and low thermal expansion coefficient [1–5]. However, high purity cordierite ceramic is difficult to prepare due to its narrow sintering temperature range as well as an incomplete transformation towards the cordierite phase with the sintering temperature below 1430 °C [6]. The interphases accompanied by quartz (SiO_2) and spinel ($MgAl_2O_4$) increase the thermal expansion coefficient of the resulting ceramic [7]. Therefore, additives are used for the preparation of cordierite ceramics to eliminate the interphases and enhance the density by liquid phase sintering. It was found that the addition of CaO promoted the sintering process and increased the thermal expansion coefficient to (4.0–4.25) $\times 10^{-6}$/°C for the cordierite glass-ceramic, in which μ-cordierite and anorthite interphases were also generated [8]. The addition of TiO_2 powder and high-temperature sintering at 1300 °C led to a higher thermal expansion coefficient of 3.61×10^{-6}/°C [9]. The addition of CeO_2 effectively eliminated the interphases of quartz and spinel, and resulted in a dense cordierite ceramic with a lower thermal expansion coefficient of 2.14×10^{-6}/°C when sintering at 1400 °C. However, the addition of K_2O did not eliminate the interphases, and greatly increased the thermal expansion coefficient, even though liquid phase sintering was largely achieved [10]. Moreover, cordierite ceramic with a major cordierite phase can be produced using kaolin, ball-clay, diatomite, talc and an Al-rich sludge when sintered at 1350 °C, and the thermal expansion coefficient reached 1.92×10^{-6}/°C [11]. Spinel and cristobalite were retained in the cordierite ceramic prepared from talc, fly ash, fused silica and alumina when sintered at 1300 °C for 3 h and only the cordierite phase was produced at 1350 °C for 3 h. This indicated that the importance of sintering temperature for improving the crystallization of cordierite [12]. By sintering the pressed specimens containing magnesium compounds and kaolinite at 1350 °C,

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Table 1
The main chemical constituents of the drift sands, wt%.

Compositions	SiO_2	Al_2O_3	CaO	Fe_2O_3	Na_2O	K_2O	Others
Contents	78.91	9.50	2.33	2.26	2.00	2.08	2.92

a dense cordierite ceramic with a thermal expansion coefficient of 2.2×10^{-6}/°C was obtained [13].

So far, large quantities of minerals and oxide powders have been used as raw materials to prepare ceramics, such as clay, kaolinite, talc, andalusite, stevensite, diatomite, steatite, gibbsite, sepiolite and forsterite [14–17], which lead to resources consumption and contribute to environmental disruption. Mineral resources are gradually dwindling because of the increasing demand for large quantity of ceramic products. To conserve resources and protect the environment, it is necessary to find other raw materials to replace these minerals. As is well known, 33% of the surface area of the earth is covered by desert and 20% of the desert is covered by drift sands [18]. In most cases, the drift sands contain substantial mineral compounds, which are generally composed of silica and a small amount of calcia, alumina and magnesium oxides, the parentage of which depends on the local historical and geographic conditions [19,20]. Therefore, it is of great importance to find a way to utilize them in ceramic manufacturing. Unfortunately, little attention has been paid to this issue until now [21,22]. This work aims to prepare cordierite ceramics using drift sands as a raw material instead of pure by a traditional solid-state sintering method, and then to investigate the phase transformation and the thermal expansion coefficients of cordierite ceramics in an attempt to determine an effective technique for utilizing this natural desert resource.

2. Experimental

Drift sands were collected from the Inner Mongolian Desert in China, with the chemical composition shown in Table 1. Commercial Al_2O_3 and MgO powders were mixed with the drift sands to produce the cordierite ceramic with a stoichiometric composition (13.78 wt% of MgO, 34.86 wt% of Al_2O_3 and 51.36 wt% of SiO_2). Different amounts of $Na_2B_4O_7$ (0, 0.5, 1.0 and 1.5 wt%) were added to form four sample groups (DRS-X). In addition, a sample (ARC) with the stoichiometric cordierite composition consisting of analytical reagents of SiO_2, Al_2O_3 and MgO was used as a reference. The compositions of the samples are listed in Table 2. The mixed powders containing 50 wt% of water were milled in a planetary ball mill for 3 h and pressed into green disks ($\phi 50 \times 10$ mm) at a pressure of 30 MPa. The green bodies were sintered at 1100–1430 °C and held at these temperatures for 2–8 h.

The phase composition was analyzed using an X-ray diffractometer (XRD, RIGAKU, D/MAX-2500/PC) with a Cu Kα radiation (40 kV, 100 mA) at a scanning speed of 3°/min. The contents of the cordierite, glass and interphases in the ceramics were calculated using MDI JADE 6.5 via a whole pattern fitting method [23]; the graph processing was performed by the Origin data processing software. The phase transformation temperature was analyzed using a differential scanning calorimeter (DSC, NETZSCH STA 409PC Luxx) with a reference sample of α-Al_2O_3 at a heating rate of 20 K/min. The morphology of the ceramics was observed using a scanning electron microscope (SEM, HITACHI, S-3400N). The composition was analyzed using an energy dispersive spectrometer (EDS, HORIBA, EMAX) and the print-out was from Microsoft Word. The thermal expansion coefficient of the ceramics was detected using a thermal dilatometer (NETZSCH/DIL 402PC) within a temperature range of 30–800 °C with a heating rate of 5 K/min.

Table 2
The compositions of the samples, wt%.

Samples	Drift sands	SiO_2	Al_2O_3	MgO	$Na_2B_4O_7$
DRS-1	60.84	/	26.80	12.36	0.0
DRS-2					0.5
DRS-3					1.0
DRS-4					1.5
ARC	/	51.36	34.86	13.78	0.0

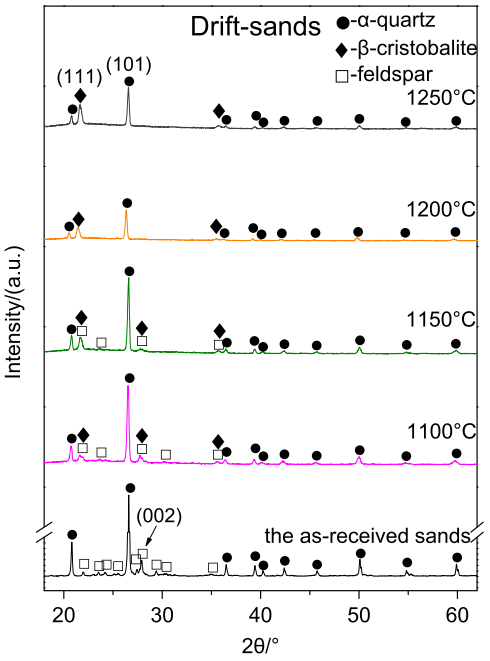

Fig. 1. XRD patterns of drift sands sintered at different temperatures for 2 h.

3. Results and discussion

3.1. Phase transformations

3.1.1. The phase transformation of the drift sands

Fig. 1 and Table 3 show the XRD patterns and the phase contents of the drift sands powder sintered at different temperatures for 2 h, respectively. The as-received sands consist of α-quartz (JCPDS: 85-0797) and feldspar (JCPDS: 41-1481). When sintered at 1100 °C, the liquid phase appeared