

Reactive-Sintered Low-Thermal-Expansion Cordierite Ceramic with High Stiffness

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Abstract

Low-coefficient of thermal expansion (CTE)-cordierite ceramic is one of the key materials for integrated circuit substrates, lithography machines and space telescope mirrors. Cordierite ceramic with high stiffness was produced by means of reactive sintering combined with hot isostatic pressing (HIP) treatment. This paper investigated thoroughly the phase evolution and densification processes. The cordierite phase formed at 1 160 °C, and only the cordierite phase and a small amount of residual spinel phase were present in the ceramic above 1 300 °C. Meanwhile, the shrinkage of the sintered body was clearly observed from 1 130 °C and the densification rate reached a maximum at 1 190 °C. After pre-sintering at 1 350 °C and followed by 1 310 °C HIP, fully dense cordierite ceramic was obtained, and its specific stiffness and CTE reached 57.5 MPa·m³·kg⁻¹, 1.75×10⁻⁶/°C (25–800 °C), respectively, i.e. values better than those of commercial cordierite products.

Keywords: Cordierite ceramics, reactive sintering, low coefficient of thermal expansion, phase evolution

I. Introduction

Cordierite ceramics have been widely applied in catalyst carriers, microwave dielectric ceramics, lithography machines for semiconductors, electronic packaging and substrates for integrated circuits, because of their extremely low coefficient of thermal expansion (CTE), good chemical stability and low dielectric constant^{1–5}. However, owing to the limited natural resources, cordierite powder synthesis and ceramic production technology has attracted great attention internationally.

Generally, cordierite powders are synthesized with solid-state reaction, sol-gel, melt quenching and other methods, and then ceramics are prepared by means of pressureless, hot pressing or spark plasma sintering (SPS)^{6–12}. Ogiwara *et al.* fabricated cordierite powder by solid state reaction at 1 270 °C and sintered it at 1 430 °C to obtain 98 %-density cordierite ceramics with a CTE of 1.8×10⁻⁶/K¹³. However, the sintering temperature was only 30 °C lower than its melting temperature (1 460 °C), which posed great difficulties in precisely controlling the sintering temperature. Later, the sintering temperature was reduced to 1 050 °C with the use of Li₂O-Bi₂O₃ as a sintering additive, but the flexural strength decreased from 243 MPa to 120 MPa and fracture toughness decreased from 3.5 MPa·m^{1/2} to 2.7 MPa·m^{1/2}¹⁴. Therefore,

reducing sintering temperature while avoiding the degradation of mechanical properties is the key to practical applications of cordierite ceramics.

Due to the ability to simultaneously achieve phase formation and ceramic densification, reactive sintering has become one of the most promising methods for preparing cordierite ceramics. For reactive sintering, natural minerals (talc, kaolinite, boehmite, diatomite, feldspar, zeolite and pearlite^{15–18}), solid waste residue or oxide powder (alumina and quartz^{19–21}) are usually used as raw materials, resulting in low cordierite phase and high impurity content. Furthermore, the presence of the alkaline metal, iron and calcium oxides will induce lattice distortion, thereby uncontrollably affecting the anisotropic thermal expansion of cordierite along *a* and *c* axes^{22,23}. Therefore, the production of cordierite ceramics by reactive sintering with high-purity oxides has received widespread attention. Obradović *et al.* obtained cordierite-mullite ceramics under pressureless sintering at 1 300–1 400 °C. Even combined with the driving force of reactive sintering, Bi₂O₃ is still needed to achieve low-temperature densification²⁴. Nouari *et al.* utilized SPS to achieve low-temperature reactive sintering at 1 150–1 200 °C using pure nano-oxide powder, achieving an optimal hardness of 8.46 GPa²⁵.

Although most studies have reported successful preparation of cordierite ceramics using reactive sintering, the addition of sintering additives affects their performance and the use of SPS limits their practical application. In addition, the properties of reactive-sintered ceramics have not

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been characterized. With this in mind, the reactive sintering method using oxide powders with high purity and high reaction activity as the starting materials was employed, without using sintering additives in this study. The phase evolution of the ceramics with sintering temperature was systematically investigated. Cordierite ceramics with high density, high specific stiffness, and low CTE were successfully fabricated.

II. Experimental and Characterization

The starting materials were commercially available high-purity magnesium oxide (purity, 99.99 %, D50, 200 nm), α -alumina (purity, 99.99 %, D50, 100 nm), and fumed silica (purity, 99.99 %, D50, 20 nm) which consisted of finely dispersed amorphous silicon dioxide. These high-purity oxide powders were accurately weighed according to the stoichiometric ratio of cordierite ($2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$) and charged into a nylon pot. Deionized water served as the solvent and 3-mm zirconia balls were selected as the ball milling medium. After mixing in a roller mill for 24 hours, a homogeneous slurry was obtained and then dried at 110 °C for 6 hours. The dried mixture was subsequently sieved through #80 mesh and then calcinated at 800 °C for 2 hours to remove organics introduced in the mixing process. Next, the mixed powder was pressed into plates with dimensions of $\varnothing 20 \times 5$ mm and then subjected to cold isostatic pressing under 200 MPa. After that, the plates were pre-sintered at the temperature of 1 100–1 350 °C in a muffle furnace for 2 hours. The phase and microstructure changes during pre-sintering were analyzed. Finally, the plates were sintered by means of hot isostatic pressing at 1 310 °C for 1 h.

The crystalline phases of the plates were analyzed by means of X-ray diffraction (XRD, Bruker D8 Advance) using $\text{Cu K}\alpha$ radiation in the 2θ range from 5° to 80° with the scan rate of 2°/min. TG-DSC of the green bodies was

performed employing STA449F3 equipment at the heating rate of 5 K/min. The fractured and thermal-etched surfaces of ceramics were observed by means of scanning electron microscopy (SEM, Verios G4 UC, ThermoFisher, USA). The average grain size of ceramics was estimated by the linear intercept method. The true average grain diameter can be expressed by the average apparent grain size obtained in two dimensions from SEM micrographs multiplied by 1.2 to ensure good measurement reliability²⁶. The density of ceramics was measured according to Archimedes principle using water as the medium. Thermal expansion of the green bodies and the sintered ceramics was tested with an automatic Netzsch DIL402 SE dilatometer at a heating rate of 5 K/min. The flexural strength of samples was tested in an INSTRON-1195 machine at 0.5 mm/loading rate. The experimental samples were processed to the standard size of $3 \times 4 \times 36$ mm³, and all four sides were polished and chamfered at 45°. Their elasticity modulus was measured with the UMS Advanced Ultrasonic Material Characterization System (UMS-100). Hardness (HV) and fracture toughness (K_{IC}) were characterized by applying a 5-N load for 10 s with a Vickers Hardness Tester (TUKON-2100B). They were calculated using the equations $\text{HV} = 1.8544P/d^2$, $K_{IC} = P \times 9.81 \times 1000 (3.14c)^{-3/2} [\tan(68 \times 3.14/180)]^{-1}$, where P was the loading, d was the indentation lengths, and c was the average crack length. The specific stiffness was calculated by dividing the elastic modulus by the density.

III. Results and Discussion

(1) Properties of starting materials

Fig. 1 shows SEM images of three starting powders MgO , Al_2O_3 and SiO_2 (Fig. 1(a), (b), (c)) and the mixture of these (Fig. 1(d)) and EDS mapping of the mixture used to prepare cordierite ceramics. The particle size distribution

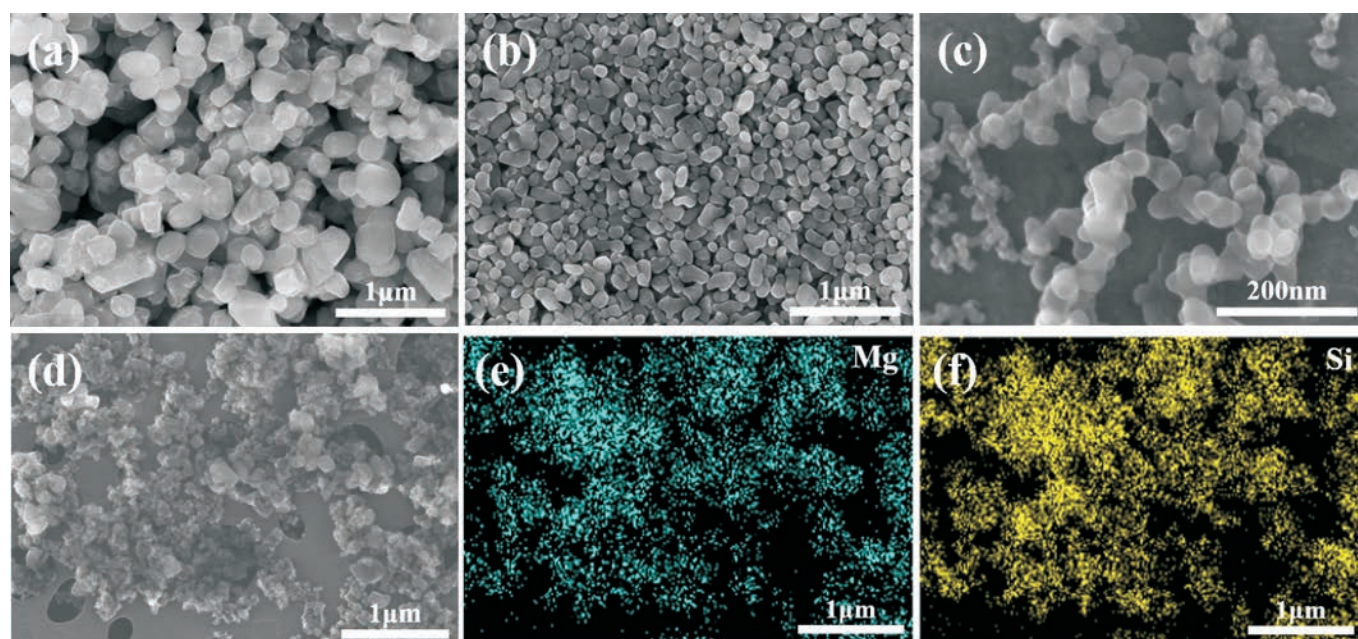


Fig. 1: SEM images of (a) MgO , (b) Al_2O_3 , (c) SiO_2 starting powders; (d) mixture of the three and EDS mapping for (e) Mg and (f) Si element of mixtures.

of the MgO powder and Al₂O₃ powder were 150–400 nm and 50–250 nm, in good agreement with the commercial data, respectively. The amorphous SiO₂ powder consisted of approximately spherical particles, and the particle size was in the range of 5–50 nm (Fig. 1(c)). Fig. 1(d), (e), (f) show the morphology of the mixture and that the distribution of Mg and Si almost coincided because the nano-scale SiO₂ reacted with MgO and H₂O to generate magnesium silicate hydrate at room temperature, which encapsulated the surface of MgO particles, prevented MgO hydration, and inhibited the formation of magnesium hydroxide. This is consistent with the results detailed in the literature²⁷. This can avoid an increase in the viscosity of the slurry, which is beneficial for the homogeneous mixing of raw materials.

(2) Phase and microstructure evolution during pre-sintering

Fig. 2 shows the XRD patterns of the samples pre-sintered from 1100 to 1350 °C respectively. After pre-sintering at 1100 °C, in addition to the corundum and enstatite (MgSiO₃) phases, tridymite and β-cristobalite (SiO₂) phase were observed, indicating that the reaction between MgO and SiO₂, and the phase transformation of SiO₂ had occurred.

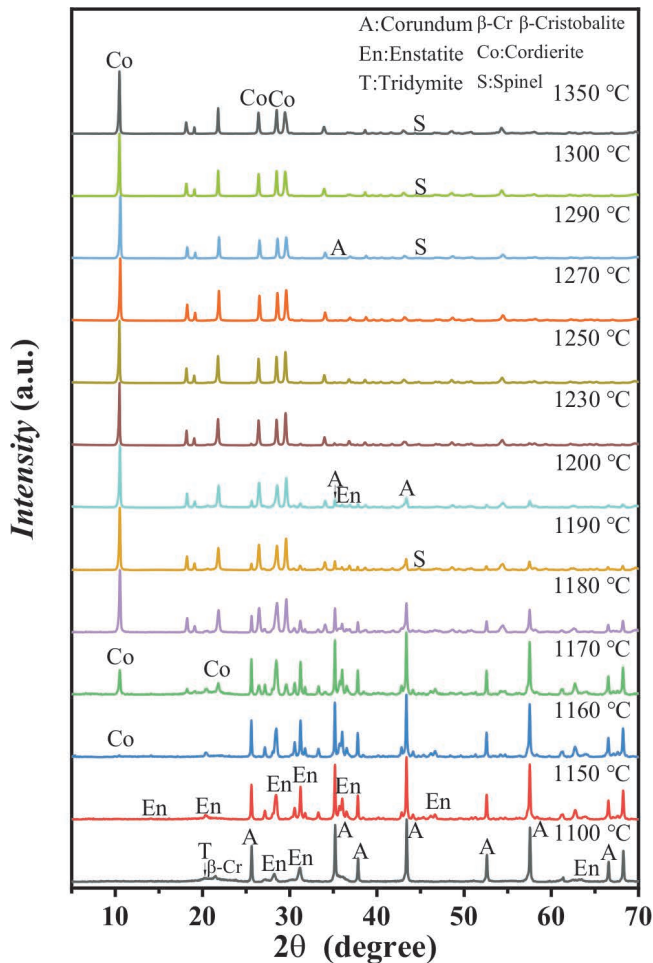
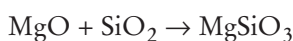
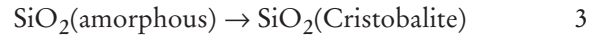
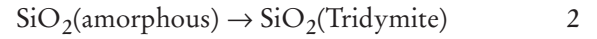


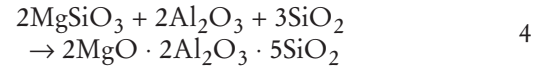
Fig. 2: XRD patterns of the samples pre-sintered at various temperatures for 2 h.



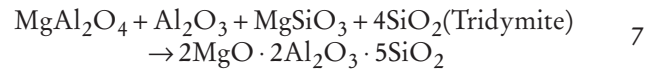
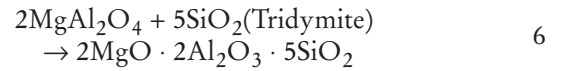
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As the temperature increased to 1150 °C, the reaction between different forms of SiO₂ and MgO continued, leading to an increase in the enstatite phase content. At 1160 °C, the peak for the cordierite phase appeared, due to the reaction among enstatite, alumina and silica. The reaction equation could be represented by the formula (4)²⁸:



At 1190 °C, the cordierite became the main phase and spinel (MgAl₂O₄) phase appeared. The synthesis of cordierite became more sufficient and the secondary phase decreased with the increase in temperature. For example, the tridymite phase disappeared at 1270 °C. Spinel could further react with other mesophases to synthesize cordierite. The reaction equations were as follows:



As the temperature further increased, there was no significant change in the peak intensities for cordierite and spinel. The main phase was cordierite with a very small amount of spinel.

Fig. 3 shows the TG-DSC curves of the green body in air. It was observed that a 2.7 wt% weight loss occurred before 800 °C, accompanied by an endothermic valley within a temperature range of 80 °C – 120 °C which can be attributed to the elimination of residual adsorbed water and structural water. As seen, the evident endothermic peak at 1188 °C corresponded to the crystalline transformation of silica and disappearance of a broad diffraction peak at around 2θ = 21.8°, which corresponded to the amorphous silica shown in Fig. 2. The exothermic peaks detected at 1195 °C and 1263 °C might correspond to the synthesis of cordierite, as represented by the formula (4) (6) (7).

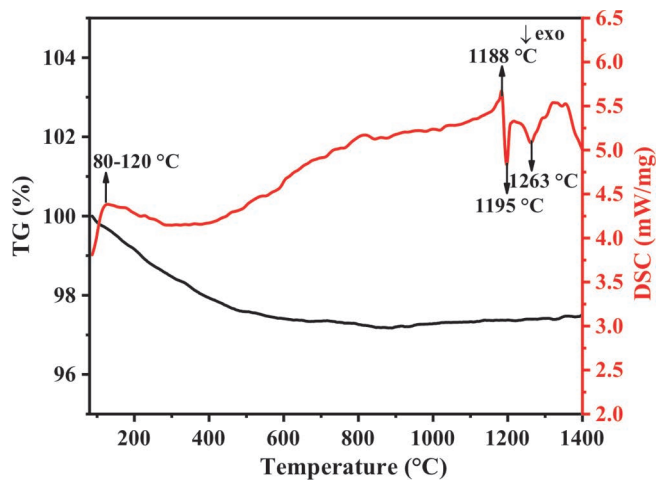


Fig. 3: TG-DSC curve for the compact of the mixture .

Based on the above experimental results, Table 1 summarizes the phases and possible reactions in the process of preparing cordierite ceramics with the reactive sintering method using high-purity oxide powders as raw materials.

Table 1: Phase evolution of the samples pre-sintered at various temperatures.

Sintering temperature	Phases evolution	Reactions
1 100 °C	Corundum, Enstatite, Tridymite, β -cristobalite	(1), (2), (3)
1 150 °C	Corundum, Enstatite (increase), Tridymite	(1)
1 160 °C	Corundum, Enstatite, Cordierite(appear), Tridymite	(4)
1 170 °C	Corundum, Enstatite, Cordierite (slight increase), Tridymite	(4)
1 180 °C	Cordierite (rapid increase), Corundum and Enstatite(decrease), Tridymite	(4)
1 190 °C	Cordierite, Corundum and Enstatite(decrease), Spinel(appear), Tridymite	(4), (5)
1 230 °C	Cordierite, Corundum and Enstatite and Tridymite (decrease), Spinel	(6), (7)
1 300 °C	Cordierite, Spinel	/

Fig. 4 shows the SEM images of the fracture surface of the samples pre-sintered at different temperatures. It could be seen that pre-sintered samples gradually became dense and pores became small and the amount of pores decreased as the temperature increased. In detail, at 1 100 °C, the SEM images revealed distinctive morphological features of the powder particles and the formation of sintering necks, indicating the beginning of sintering. At 1 150 °C, the initial particle shape disappeared, with numerous interconnected pores becoming apparent. This phenomenon can be attributed to the high activity of fumed silica particles, facilitating easy diffusion and mass transfer, thereby promoting densification even at low temperatures.

When the temperature increased to 1 160–1 200 °C, the SEM of samples showed that the number of pores decreased but several larger-sized pores existed locally. Subsequently, these pores disappeared as the temperature increased above 1 270 °C. Finally, a small amount of less than 1- μ m pores remained.

Fig. 5 (a) shows the density and apparent porosity of the bodies pre-sintered at different temperatures. The density of samples increased first and then decreased with the increase in temperature. We measured the bulk density of the green body as 1.67 g·cm⁻³ with the Archimedes method using kerosene as a medium. The theoretical den-

sity of cordierite is 2.52 g·cm⁻³. As the temperature increased to 1 160 °C, the open porosity reached 0. It means that sintering shrinkage and the volume expansion of the sintered body happen simultaneously. The density variety could be better comprehended by comparing the theoretical density of the materials listed in Table 2. In detail, as the temperature increased from 1 100 °C to 1 160 °C, a significant increase in density was observed, which aligned with the SEM images (Fig. 4). Based on the XRD (Fig. 2), the high-density phases enstatite (3.22 g·cm⁻³) and corundum (3.98 g·cm⁻³) are main phases, therefore the sample density increased. Above 1 170 °C, the secondary phase with the high-density enstatite and corundum decreased, largely accompanied with synthesis of cordierite, leading to the density of the sample decreasing. In this study, the open porosity of the sample reached 0 at a low temperature (1 160 °C), showing a rapid densification rate in the cordierite ceramics preparation by using nano-scale fumed silica as the raw material. Compared to the previous literature, our work reduced the pre-sintering temperature by almost 200 °C compared to that of the sample with a similar densification level¹³.

Table 2: The theoretical density of the materials.

Material	Lattice	Density/g·cm ⁻³
Enstatite (MgSiO ₃)	Orthorhombic	3.22
Periclase (MgO)	Cubic	3.58
Corundum (Al ₂ O ₃)	Hexagonal	3.98
Spinel (MgAl ₂ O ₄)	Cubic	3.58
Cordierite (Mg ₂ Al ₄ Si ₅ O ₁₈)	Hexagonal& Orthorhombic	2.52

Fig. 5(b) depicts the dimensional shrinkage and densification rate of the green body as the function of pre-sintering temperature. At around 1 130 °C, the green body began to shrink, and reached a maximum shrinkage of 23 % at 1 190 °C, and then began to expand. Two sharp peaks at 1 175 °C and 1 195 °C were observed from the densification rate curve. This implied that within a short temperature range of 40–50 °C, the green body underwent rapid and substantial shrinkage. The densification rate reached a maximum in this temperature range. According to the XRD pattern results (Fig. 2), a large amount of cordierite was synthesized at 1 180 °C. However, a volume expansion about 6 % would occur during the formation of cordierite²⁹. This meant that, before 1 190 °C, the effect of sintering shrinkage outweighed that of the reaction expansion. Beyond 1 190 °C, the sintering curve exhibited an upward trend, indicating that the effect of sintering shrinkage became less significant than that of the reaction expansion. Finally, at temperatures ranging from 1 290–1 350 °C, the sample attained a shrinkage of 21 %–22 %.

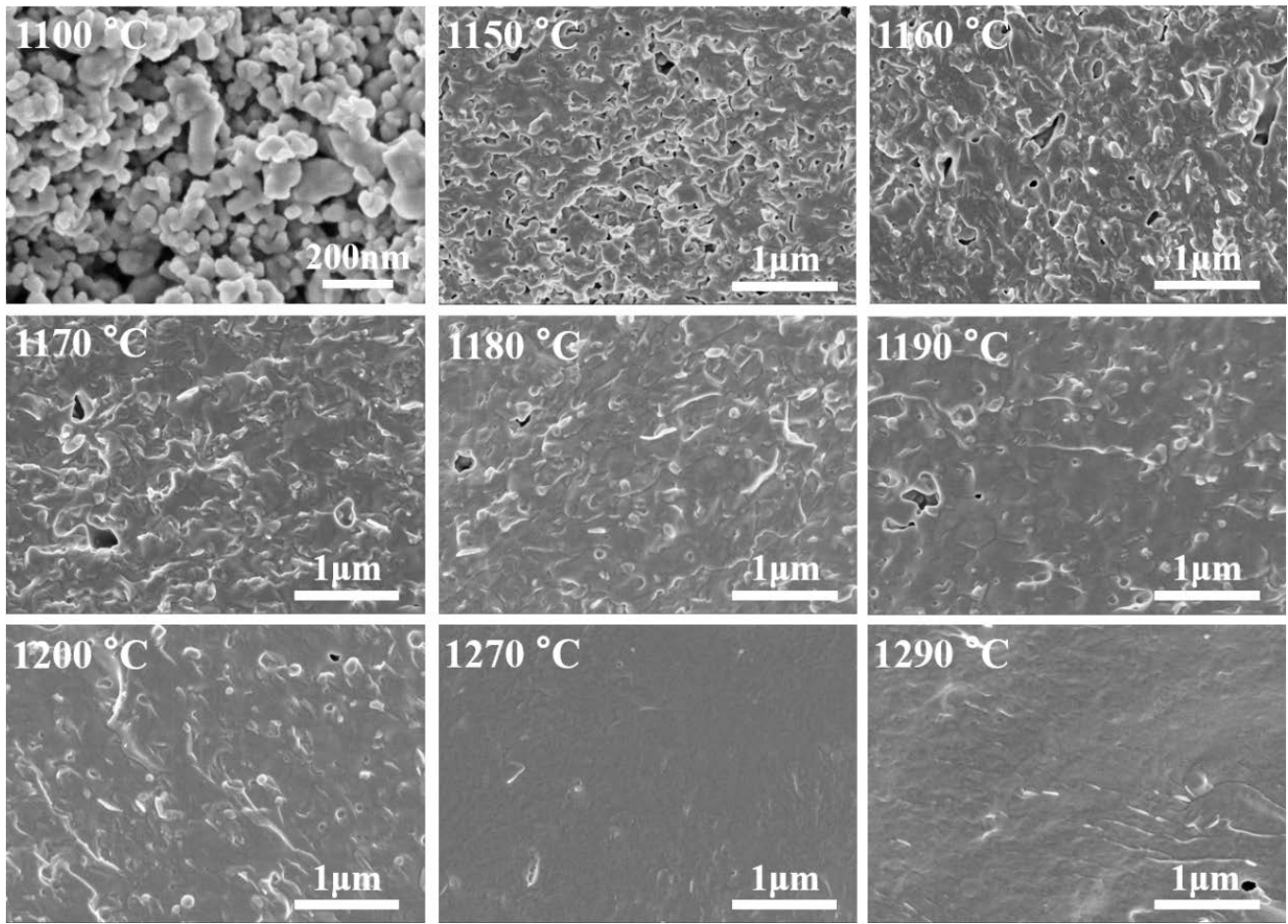


Fig. 4: SEM images of the fracture surface of the samples pre-sintered at 1100–1290 °C.

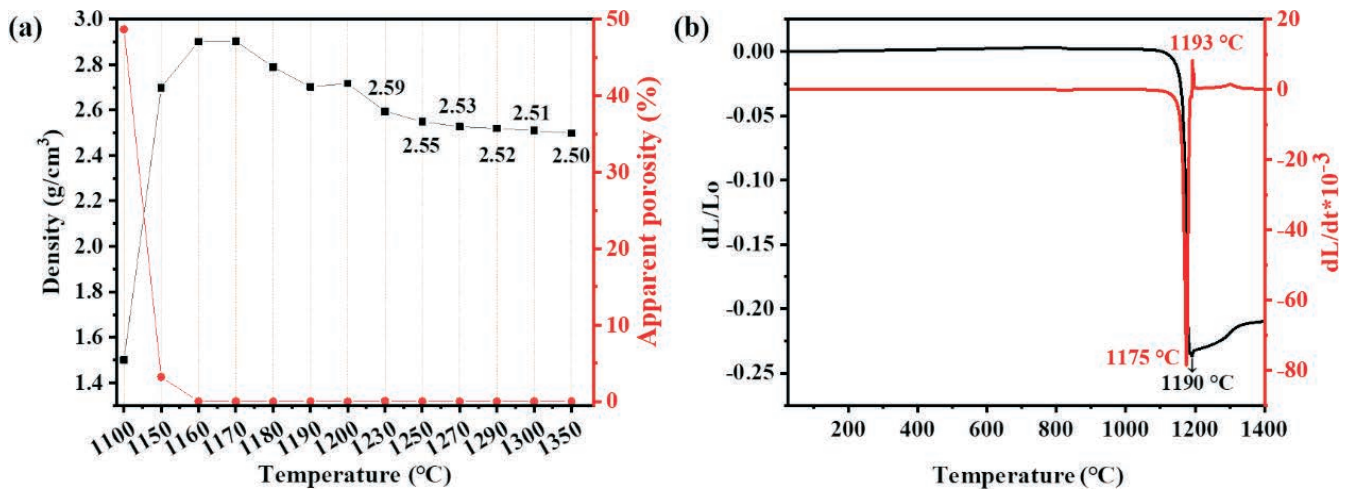


Fig. 5: (a) The density of pre-sintered bodies versus temperature, (b) thermal expansion shrinkage curve of the green body.

Fig. 6(a) shows the thermally etched surface microstructure of the samples pre-sintered at different temperatures. The grain grew with the increase in pre-sintering temperature. The cordierite grains developed completely and clearly by 1290 °C. Rectangular and hexagonal grains were found, corresponding to different directions of hexagonal cordierite or orthogonal cordierite³⁰. When the pre-sintering temperature was 1230 °C, there were a few unevenly distributed pores at the grain boundary. However, the density of this sample was highest of these

four samples as we can see from Fig. 5(a). Two main factors, including the pores and the phase composition, impacted the density. It is noticeable that the pores decreased as the pre-sintering temperature increased. The formation of cordierite from alumina, magnesium oxide, and silica resulted in a decrease in density. Above 1230 °C, the phase change had a greater effect on the density than that of pores, so the density decreased as the temperature increased. Fig. 6(b) shows the grain size distributions of the samples. As the pre-sintering temperature increased, the average grain size grew from 168 nm to 256 nm.

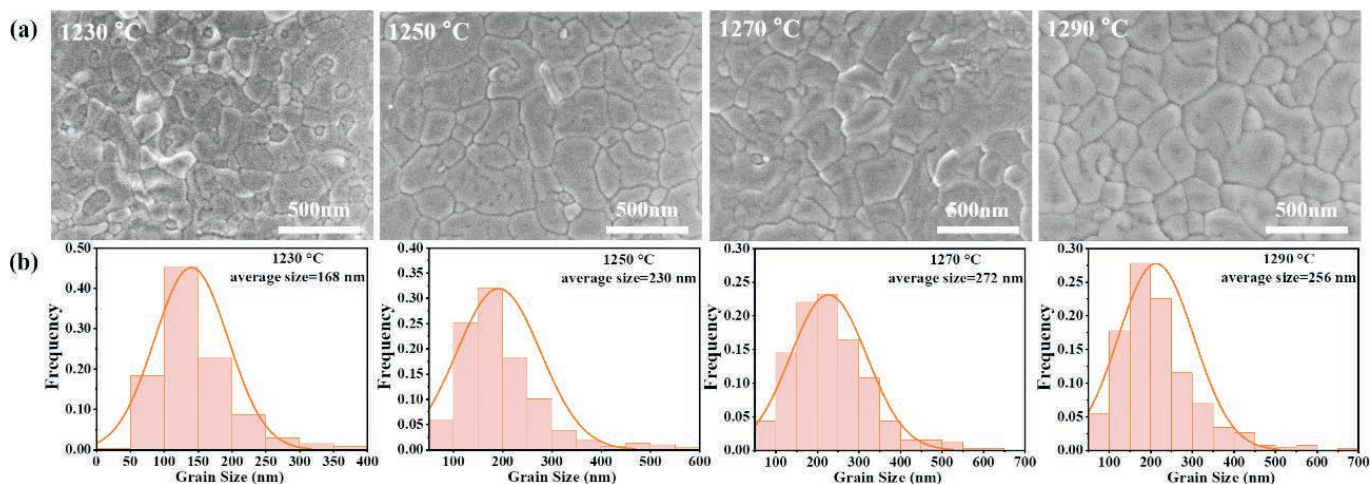


Fig. 6: (a) Microstructures of the thermally etched samples pre-sintered at different temperatures and (b) grain size distributions corresponding to (a).

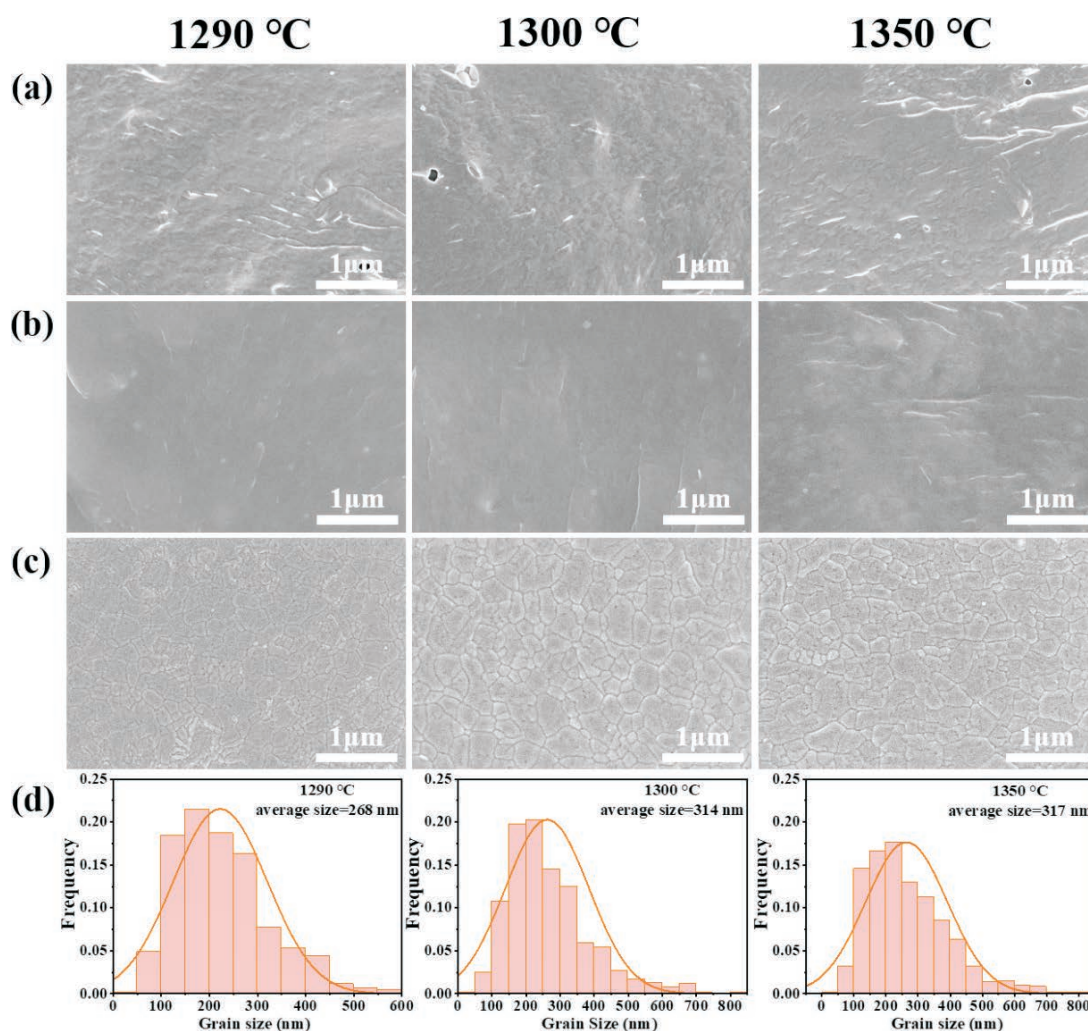


Fig. 7: SEM images of (a) the fracture surfaces of the samples pre-sintered at 1290 °C, 1300 °C, and 1350 °C, (b) fracture and (c) thermally etched surface of (a) further HIPed at 1310 °C, (d) grain size distributions corresponding to (b).

Fig. 7(a) shows the fracture surfaces of the samples pre-sintered at 1290 °C, 1300 °C, and 1350 °C, respectively. As the pre-sintering temperature increased, the size and number of the pores decreased. After subsequent HIP treatment at 1310 °C, there were no obvious pores on the

fracture surface (Fig. 7(b)) and thermally etched surface (Fig. 7(c)) of the HIPed samples, indicating that HIP treatment could effectively eliminate the intergranular pores located at the grain boundary. Moreover, it can be found in Fig. 7(d) that there was no significant grain growth as

the pre-sintering temperature increased from 1 300 °C to 1 350 °C. The microstructure of the ceramics was uniform, the average grain size of the ceramic was maintained about 317 nm. Table 3 shows the density and average grain size of ceramics HIPed at 1 310 °C. The density of samples is the same, which further reflected the similar high densification and phase composition of cordierite ceramics.

Table 3: Bulk density and average grain size of ceramics after pre-sintering at different temperatures and HIP sintering at 1 310 °C.

Pre-sintering temperature (°C)	Bulk density (g·cm ⁻³)	Average grain size (nm)
1 290	2.52	268
1 300	2.52	314
1 350	2.52	317

(3) Properties of cordierite ceramics

Fig. 8 illustrates the flexural strength and elasticity modulus of the HIPed samples pre-sintered at 1 290 °C, 1 300 °C and 1 350 °C. With the increase in the pre-sintering temperature from 1 290 °C to 1 350 °C, the flexural strength of the samples increased from 133 ± 3 MPa to 141 ± 6 MPa. The flexural strength of samples does not change as the pre-sintering temperature and the grain size increase. Because the fracture mode of ceramics is mainly trans-granular fracture as we see in Fig. 7(b). Furthermore, the densities of HIPed ceramics are the same (Table 3), which resulted in the similar flexural strength of the ceramics. However, the elastic modulus of the sample showed no significant change with temperature, which might be due to the same density of the samples after HIP treatment (Table 3). As we can see it from the XRD pattern (Fig. 2), the secondary phase peak intensity became lower with the increase of the pre-sintering temperature. There was no significant change in the content of this phase when the pre-sintering temperature reached 1 300 °C. Therefore, the elasticity modulus stabilized at 145 GPa. For this sample, the density was $2.52 \text{ g}\cdot\text{cm}^{-3}$, and the specific stiffness was calculated to be $57.5 \text{ MPa}\cdot\text{m}^3\cdot\text{kg}^{-1}$.

Fig. 9 shows Vickers hardness and fracture toughness of the HIPed samples pre-sintered at 1 290 °C, 1 300 °C and 1 350 °C. As seen, the hardness was approximately 9.0 GPa, and the fracture toughness of the sample was about $2.0 \pm 0.2 \text{ MPa}\cdot\text{m}^{1/2}$. The hardness and fracture toughness of the ceramics were influenced by porosity and phase composition. In this case, there were no distinct variations in porosity and phase composition among the HIPed samples pre-sintered at different temperatures, therefore the Vickers hardness and fracture toughness did not exhibit any significant change. The present Vickers hardness was higher than that of the sample prepared by means of SPS with the same phase composition and similar densification by Saheb³¹. This might be attributed to slight differences in phase content of samples³².

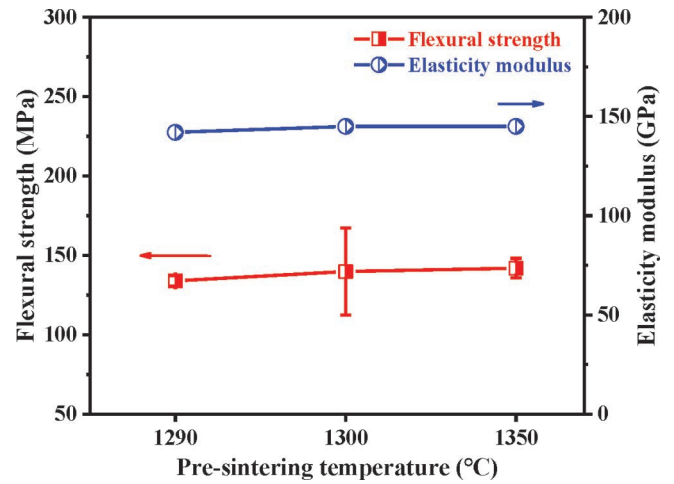


Fig. 8: Flexural strength and elasticity modulus of the 1 310 °C-HIPed samples pre-sintered at different temperatures.

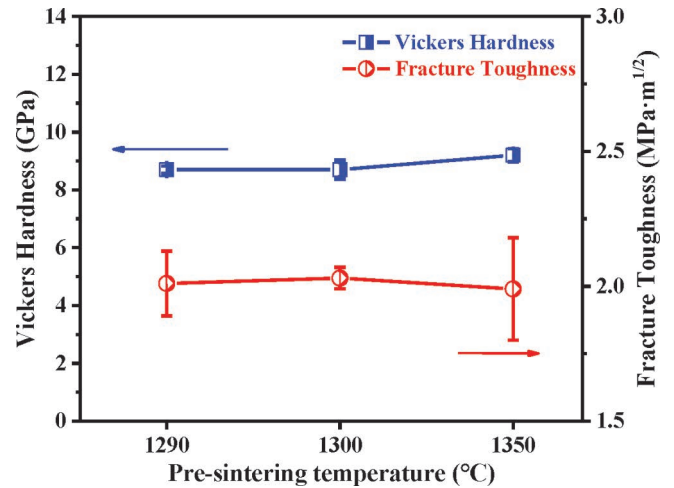


Fig. 9: Vickers hardness and fracture toughness of the 1 310 °C-HIPed samples pre-sintered at different temperatures.

The linear thermal expansion and the coefficient of thermal expansion (CTE) of the sample sintered at 1 350 °C for 2 h and then HIP-sintered at 1 310 °C are shown in Fig. 10. It can be seen that CTE initially decreased and then increased, reaching minimum value ($-0.15 \times 10^{-6}/^{\circ}\text{C}$) at around 78 °C, which reflected the negative thermal expansion characteristics of cordierite. The average value of CTE measured between 25 °C and 800 °C was calculated to be $1.75 \times 10^{-6}/^{\circ}\text{C}$, which is close to that reported in the literature³³.

Table 4 shows the sintering conditions and mechanical properties of cordierite ceramics from this work and literature. It is worth noting that the sintering temperature employed in this work was lower than the critical temperature of 1 430 °C utilized by a two-step solid state reaction for cordierite synthesis¹³. This achievement was credited to the utilization of oxide powders with high activity for both reaction and densification. At the same time, the value of hardness was the highest compared to other jobs and the value of specific stiffness was higher than that of commercial products. For example, the specific stiffness of $57.5 \text{ MPa}\cdot\text{m}^3\cdot\text{kg}^{-1}$ surpasses the value of CO-720, which

Table 4: Comparison of the parameters and properties of the ceramics from this work and others.

Parameters and properties	This work	Ogiwara <i>et al.</i> ¹³	Saheb <i>et al.</i> ³¹	Commercial products (CO-720)
Preparation method	Reactive sintering and HIP treatment	Two steps sol- id state reaction	SPS	
Sintering temperature (°C)	1 300–1 350	1 430	1 150	
CTE ($\times 10^{-6}/^{\circ}\text{C}$)	1.75 (25–800 °C)	1.8 (RT–800 °C)	/	2.1 (40–800 °C)
Flexural strength (MPa)	141	243	/	200
Specific stiffness ($\text{MPa}\cdot\text{m}^3\cdot\text{kg}^{-1}$)	57.5	/	/	56.0
Vickers hardness (GPa)	9.0	7.5	8.7	8.5
Fracture toughness ($\text{MPa}\cdot\text{m}^{1/2}$)	2.0	3.5	2.25	1 ~ 1.5

stands at $56.0 \text{ MPa}\cdot\text{m}^3\cdot\text{kg}^{-1}$. In a word, the cordierite ceramics fabricated by means of reactive sintering in this work exhibited low CTE and high specific stiffness, which should be well-suited for applications in integrated circuit substrates and space telescope mirrors.

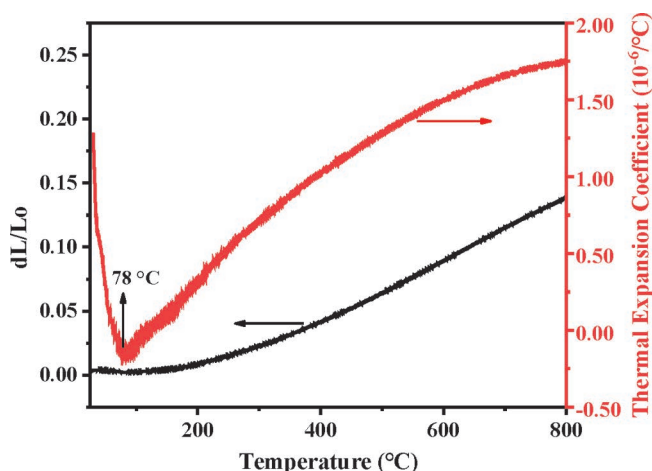


Fig. 10: Linear shrinkage and coefficient of thermal expansion of the sample pre-sintered at 1 350 °C and then HIPed at 1 310 °C.

In future, we will adjust the stoichiometric ratio of oxide powders to prepare cordierite ceramics with higher phase purity further to obtain the lower value of CTE.

IV. Conclusions

Cordierite ceramics were prepared by means of reactive sintering using nano oxide powders in this study. The phase, microstructure evolution and densification with pre-sintering temperature were systematically investigated. The cordierite ceramics with low thermal expansion and excellent mechanical properties were successfully synthesized under a lower temperature. The specific conclusions are as follows:

(1) The cordierite phase appeared at a pre-sintering temperature of 1 160 °C. Between 1 160–1 180 °C, the volume expansion caused by cordierite and the sintering densification occurred simultaneously. When the pre-sintering temperature exceeded 1 300 °C, the ceramic consisted of

predominantly cordierite phase along with minor spinel phase.

(2) The density of the ceramic after pre-sintered at 1 350 °C was measured to be $2.50 \text{ g}\cdot\text{cm}^{-3}$ (99 % RD) and increased to $2.52 \text{ g}\cdot\text{cm}^{-3}$ (100 % RD) after further HIP at 1 310 °C. The average CTE between 25 °C and 800 °C was $1.75 \times 10^{-6}/^{\circ}\text{C}$. In addition, the flexural strength was 141 MPa, the elastic modulus was 145 GPa and the specific stiffness was $57.5 \text{ MPa}\cdot\text{m}^3\cdot\text{kg}^{-1}$. The fracture toughness was $2.0 \text{ MPa}\cdot\text{m}^{1/2}$ and the Vickers hardness was 9.0 GPa. The present work provides a new approach to prepare cordierite ceramics by reactive sintering of nano-scale oxide powders which are active in both the reactive reaction and sintering densification.

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