Impact of Sintering Conditions for a Robust Cordierite Ceramic Reinforced with Nano-Silicon Nitride

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Abstract: Ceramic-composite materials based on cordierite/nano-silicon nitride (Si₃N₄) were prepared through the sol-gel route. The sintering process of the green cordierite alone at 1300 and 1400°C forms cordierite and mullite, respectively. Sintering of green cordierite alone with/without Si₃N₄ at 1350°C provided cordierite and cristobalite in an ambient condition, while in nitrogen condition, it allowed the crystallization of cordierite and Si₃N₄ phases. The microstructures of the ceramic composites showed irregular or clustered particles containing nano-sized grains of the major cordierite that developed in all the sintered samples at 1350°C. Upon these different sintering temperatures, the bulk densities ranged between 2.255 and 2.305 g/cc in the air and 2.254 and 2.355 g/cc in nitrogen conditions. Moreover, their apparent porosities were between 13 and 17 % in the air and 12.1 and 14 % in nitrogen conditions. The values of bending strength were between 50 and 325 N/mm². They were higher in the air than in the nitrogen condition.

Keywords: ceramic; cordierite; silicon nitride; sintering; magnesium aluminosilicate.

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1. Introduction

Cordierite ceramic features include promising electrical as well as thermo-mechanical ones. Therefore, enhancing the cordierite properties drew the attention of various researchers [1-4]. However, many routes like ceramic, glass-ceramic, composite, and wet chemical methods [5,6]. Ceramics can be applied in various applications [7-9]. Cordierite was prepared through glass-ceramic materials in the form of magnesium aluminum silicate phase (2Al₂O₃·5SiO₂·2MgO). It is considered an advanced ceramic material [10]. It can be used at a slightly high-temperature 1455°C. This is referred to as their phase stability; it has a wide range of applications at a suitable temperature from 25°C to 1350°C [11,12] when mentioned with respect to other ceramics [13-18]. Meanwhile, the poor mechanical properties act as a barrier against efficient functional applications. Recently, some research showed that incorporating Si₃N₄ into a glass-ceramic with magnesium aluminosilicate (MAS) matrix might fortify its mechanical properties [19,20]. In these composites, glass-ceramic MAS can be applied as potent sintering support with a perfect densifier behavior. Moreover, this material may be
considered a structural matrix [21-24]. Ceramics of silicon nitride are promising candidates in various engineering applications. Generally, it is difficult to sinter pure silicon nitride ceramics due to the low self-diffusivity of the covalent material. It does not allow densification by classical solid-state sintering techniques. Preparation of dense Si₃N₄ usually requires additives that react with the present SiO₂ on the Si₃N₄ surface to form a liquid phase. Moreover, these silicates facilitate densification. Many oxide additives have been successfully demonstrated in generating intensive Si₃N₄ substrates. The additives of silicon nitrides are featured as a glassy phase with grains. They decompose at elevated temperatures [25,26]. Hence, they turn to a crystalline material possessing a raised melting point. Supported barium aluminum silicate ceramics were adequately investigated. The reason is the ability to use their various applications with superior properties. Moreover, they are able to be easily produced at low costs. Hot pressing (HP) and pressure-less sintering (PLS) are among the familiar manufacturing procedures to obtain (BA S/Si₃N₄) composites. The resulting BA S/Si₃N₄ composite showed high fracture strength. The latter property for 30% BA S/Si₃N₄ composite was 5.4 MPa m¹/². Hence, these materials have the capability to keep a high strength at elevated temperatures reaching 1120 °C [27,28]. This indicates that the crystallized (BA S) matrix benefits significantly from its vigor at raised temperatures. Afterward, the composites based on BA S/Si₃N₄ ceramics are able to be applied at low and high-temperature implementations. Composite ceramic of whisker microstructure was prepared from cordierite glass-ceramic/silicon nitride and had higher toughness of 1.4 MPa m¹/² to 2.6 MPa m¹/² than the cordierite glass-ceramic, which is 1.4 MPa m¹/² [29]. The high strength of 1470 MPa for the composite material was prepared from cordierite (30%) through the solid-state and sol-gel method with either micro or nano Si₃N₄ powder sintered between 1350 to 1515°C [30].

Cordierite reinforced silicon nitride displays an integrated desirable property for high-temperature structural applications. These composites' high strength and moduli directly consequence the high strength and modulus of the nanocrystalline silicon nitride. It was deduced that cordierite could not be used for long-term applications prepared at more than 1000°C. High-temperature combustion cordierite is used as a structural material at 1000°C. This refers to its heat resistance in an oxidizing atmosphere and thermal shock resistance generated from its low thermal expansion.

This work aims to prepare and characterize cordierite minerals from alkoxide chemicals using a ternary system of SiO₂, Al₂O₃, and MgO via the Sol-Gel technique. Moreover, we are attempting to improve these systems' mechanical strength and microstructure upon introducing silicon nitride.

2. Materials and Methods

2.1. Materials.

Cordierite (Mg₂Al₃Si₅O₁₈) was prepared by gel method using pure aluminum tri-isopropoxide (Fluka, Germany), magnesium nitrate hexahydrate (BDH, UK), extra pure tetraethyl orthosilicate (TEOS) (Loba chemicals, India). Silicon nitride nano-powder (Elektroshmeltz Werk Kempten GmbH, Germany, having a mean particle size of 180 nm) was added in 10, 20, and wt. 30% (Table 1).
2.2. Preparing the composites.

According to the stoichiometric cordierite (Mg₂Al₄Si₅O₁₈), calculated amounts of Mg(NO₃)₂.6H₂O, aluminum alkoxide, and TEOS (formula) was carefully weighed. Firstly, the boehmite solution was prepared by hot water hydrolysis of aluminum alkoxide. Excess amount of water [1 (alkoxide):100 (H₂O)] was added to ensure complete hydrolysis. Continuous stirring for 2 hours was maintained at 80°C. HNO₃ was introduced to the solution in 4 ml. Afterward, a clear solution was obtained. The aqueous solution of Mg(NO₃)₂.6H₂O was appended. Excess hot water was used to hydrolyze TEOS at the same reaction temperature. HNO₃ was utilized as a catalyst to get silica sol. The latter was loaded to the Al-Mg clear sol with stirring for one more hour at the same temperature. This was followed by introducing Si₃N₄ in weight proportions of 10, 20, and 30 wt% prior to the final addition of nitric acid (gelation step). Hereafter, these mixtures are denoted as CSN10, CSN20, and CSN30, as shown in Table 1. Another 5 ml of nitric acid was added to the mixture under gentle stirring until complete gelation. The next equations can show the steps for the preparation process:

\[
\text{C}_2\text{H}_2\text{AlO}_3 + \text{Hot water (excess)} \rightarrow \text{AlO(OH)} + \text{H}_2\text{O} \quad (1)
\]
\[
\text{AlO(OH)} + \text{HNO}_3 \rightarrow \text{AlNO}_3 \quad (2)
\]
\[
\text{Mg(NO}_3\text{)}_2 + \text{HNO}_3 \rightarrow \text{Mg(OH)}_2 \quad (3)
\]
\[
\text{SiC}_8\text{H}_{20}\text{O}_4 + \text{HNO}_3 \rightarrow \text{Si(OH)}_4 \quad (4)
\]

\[
(2) + (3) + (4) \rightarrow 2\text{MgO-2Al}_2\text{O}_3-5\text{SiO}_2
\]

Table 1. The composition of the prepared cordierite/Si₃N₄ nanocomposites.

| Sample | Cordierite (wt. %) | Si₃N₄ (wt. %) |
|--------|-------------------|---------------|
| Blank  | 100               | 0.0           |
| CS10   | 90                | 10            |
| CS20   | 80                | 20            |
| CS30   | 70                | 30            |

The gels were calcined at 650°C to eliminate any organic residues. The dried samples were shaped into rectangular with 15 x 5 x 5 mm dimensions and uniaxially pressed at 100 MPa. The pre-molded samples were divided into two equal numbers and fired at 1350°C in air, while the other samples were fired at the same temperature under a flow of nitrogen gas with a soaking time of 2 h for all of the samples. However, the cordierite gel was tested for thermal behavior up to 1350°C.

2.3. Characterization.

Identification of crystalline materials after the sintering process was considered with X-ray diffraction (XRD) analysis. Moreover, scanning electron microscopy (SEM/EDX) was important for morphological clarifications. The applied X-ray diffraction analysis was monochromatic Cu Kα radiation XRD (D 500, Siemens, and Mannheim, Germany). The microstructure was photographed by scanning electron microscopy, SEM (Philips, XL 30 - Eindhoven, Netherlands) attached with X-ray microanalysis EDX.

Archimede's's water displacement process was employed to determine bulk density and porosity. It was carried out according to (ASTM C-20) for the fabricated sintered composite samples. Bending strength was measured by a 4-points bending test for a sample. All of the
tests were carried out by the universal testing machine Instron Model 1186, USA. The specimens were tested at a crossed head velocity of 0.5 mm/min. The values of bending strength were calculated according to the following equation:

$$\sigma_{4B} = \frac{3F (L1 - L2)}{2bh^2} = \frac{N}{mm^2}$$

where:
F: Force of compression. (N).
L1: Outer Span. (mm).
L2: Inner Span. (mm).
B: Width of the Sample. (mm).
H: Height of the Sample. (mm).

3. Results and Discussion

3.1. X-ray diffraction.

The sintering crystallization behavior of cordierite gel was tested as received dried gel (under air atmosphere) and at different temperatures, as indicated in Figure 1. The as-received dried CSN0 sample shows an amorphous pattern, whereas an increase of sintering temperature up to 1300°C for zero or half an hour showed only the crystallization of cordierite (Mg$_2$Al$_4$Si$_5$O$_{18}$, ICDD card 89-1488). Increasing the temperature up to 1400°C for zero-time mullite was developed alone [Al$_2$ (Al$_{2.8}$ Si$_{1.2}$) O$_{9.6}$, ICDD card 79-1275] with complete disappearance of cordierite as shown in Figure 1.

However, the developed crystalline phases after the sintering process at 1350°C for 2 hours in an air and nitrogen atmosphere were illustrated in Figure 2. In air condition, the X-ray diffraction patterns of the blank CSN0 sample showed typical diffraction peaks [3] at a 2 theta angle of 10.48, 18.038, 19.08, 21.68, and 26.48. Additional peaks arose at 28.48, 29.48, and 33.98 of cordierite and cristobalite (of the highest line at 2 theta 21.94, ICDD card 76-0935) as illustrated in Figure 2. In the case of nitrogen atmosphere, cordierite was a major phase, whereas incorporation of SiN shows its appearance through diffraction peaks at 2 theta at 23.41, 27.09, 33.73, 36.1, 41.4, and 52.2 (ICDD card 71-0623).

The X-ray diffraction results show that cordierite was developed as the main phase in both atmospheric conditions (i.e., air and nitrogen). Under air condition, Si$_3$N$_4$ decomposes, and silicon is oxidized into cristobalite. Meanwhile, under inert conditions (i.e., nitrogen), Si$_3$N$_4$ persists in decomposition and appears as a minor phase.

![Figure 1. XRD patterns of sintered cordierite up to 1400°C under air atmosphere.](https://biointerfaceresearch.com/)
Figure 2. XRD patterns of the sintered samples at 1350°C in air and nitrogen atmosphere.

Figure 3. SEM micrographs of sintered CSN0, CSN10, CSN20 and CSN30 at 1350°C in air.
3.2. Investigating the surface morphology.

The SEM micrographs of cordierite samples either sintered in the air or under a nitrogen flow are displayed in Figures 3 and 4. SEM micrographs show the comparison between the sintered samples at 1350°C in air and nitrogen. Irregular clusters of crystalline phases were spread in the samples. Moreover, generally little pores were still present through the sintered samples between the cluster granules [12] are yet present. This means the good densification and sintering process of all samples. The spread clusters containing nano-scale grains of the major cordierite are shown in Figures 3 and 4.

Moreover, the later nano-granular crystals were rounded in all the samples, whereas the high Si$_3$N$_4$-containing sample (CSN30) showed a spreading nano-rod shape, as displayed in Figure 4. How nano-grain were developed in the samples sintered in the air may be wondered. Therefore, we can imagine that the decomposition of nano-Si$_3$N$_4$(Si$_3$N$_4$+3O$_2$→3SiO$_2$+2N$_2$) can form sub-nano-sized cristobalite (SiO$_2$), which form as nucleate for the subsequent formation of cordierite. Furthermore, Kingery et al. [31] suggested forming the solution-precipitation mechanism of liquid-phase formation glassy matrix to attain sintering. Whereas the sintering process in nitrogen condition, the nanoparticles of Si$_3$N$_4$ can make more nucleate sinters for consequent crystallization of nano-scale particles of cordierite. However, we can imagine that the liquid phase is first formed by increasing the temperature, then redistributions and particle rearrangement. The EDX microanalysis in Table 2 shows (with exception) the presence of carbon with the components of cordierite structure in the whole investigated samples.

![SEM micrographs of sintered CSN0, CSN10, CSN20 and CSN30 at 1350°C in nitrogen.](https://biointerfaceresearch.com/)

The EDX microanalyses for area scans of both the cordierite and cordierite/S10 ceramic samples sintered at 1350°C for 1 hour are tabulated in Table 2. The microanalysis shows the possible formation of cordierite phases. All samples detect carbon elements. It may be due to
the TEOS starting materials. Meanwhile, the nitrogen element appeared due to the sintering condition. The whole sample showed the formation of a pure cordierite phase. The point analysis of cordierite/10SN sample spots to a sintered sample under air or nitrogen showing MgO deficiencies cordierite. The energy dispersive X-ray microanalysis cannot provide an accurate analysis. Its beam does not only give the chemical analysis grain in (mico or nano) size, but it gives the analysis for the outer surface area of the grain. Therefore, it is qualitative to determine the composition.

Table 2. EDX analysis of cordierite and CS\textsubscript{10} nanocomposite.

| Sample                  | C   | Mg  | Al  | Si  | O    |
|------------------------|-----|-----|-----|-----|------|
| Cordierite (air)       | 8.91| 7.27| 13.70|13.75|56.37 |
| Cordierite (N\textsubscript{2}) | 9.84| 8.65| 15.36|14.05|52.10 |
| Cordierite/CS\textsubscript{10} (air) | 11.58| 5.82| 10.73|12.92|58.95 |
| Cordierite/CS\textsubscript{10} (N\textsubscript{2}) | 7.35| 5.8 | 20.04|11.21|55.60 |
| Nominal cordierite     | -----| 8.31|18.45|24.01|49.23 |
| Mg\textsubscript{2}Al\textsubscript{5}O\textsubscript{18} | -----|

3.3. Apparent porosity and bulk density.

Figures 5 and 6 represent the apparent porosity as well as the bulk density of the cordierite silicon nitride nanocomposites samples. The apparent porosity values of the sintered samples in the air are higher than the sintered ones in nitrogen. This increment is due to the formation of pores accompanied by the oxidation process [3,5] for SiN\textsubscript{3} into SiO\textsubscript{2} and liberated NO\textsubscript{2}. However, it is clear that incorporating SiN\textsubscript{3} is associated with an increase in porosities and densities in both conditions (i.e., air and nitrogen). In general, the sintering in the air atmosphere provided a highly porous structure with low densities. However, it extended a small number of pores upon sintering in nitrogen conditions. As a result, they exhibit high densities with low porosity [4]. It was noticed that both densities and porosities showed an increasing trend as the content of Si\textsubscript{3}N\textsubscript{4} increased. The apparent porosity values of the samples sintered in air showed a proportional increase. This can be correlated to the different types and numbers of the produced porosities. Hence, it is accompanied by the oxidation of the silicon nitride phase forming SiO\textsubscript{2}. These results were confirmed by the scanning microscopy results. However, samples containing Si\textsubscript{3}N\textsubscript{4} showed a wavy shape. This may be due to the glassy phase with minimum values of porosities formed during sintering, as shown in SEM micrographs.

![Figure 5](https://biointerfacereasearch.com/)  
**Figure 5.** The apparent porosity of the cordierite silicon nitride nanocomposites sintered up to 1350°C.
3.4. Bending strength.

The composite samples' bending strength values are illustrated in Figure 7, either in the air (Figure 7a) or nitrogen condition (Figure 7b), which were between 50 and 325 N/mm². It has to be noticed that the lowest bending strength was 50 N/mm² for CSN30 in nitrogen conditions. The highest one was for CSN20 (325 N/mm²) in air condition. However, the bending strength values in the air were higher than those in the nitrogen condition.

4. Conclusions

Composite ceramics of cordierite (Mg₂Al₄Si₅O₁₈) with/without (Si₃N₄) were prepared through the sol-gel technique. Cordierite and mullite were developed when the green cordierite sample was sintered at 1300 and 1400°C, respectively. At the same time, the sintering for all of the green samples at 1350°C led to the crystallization of cordierite and cristobalite in normal conditions. Meanwhile, under nitrogen conditions, it provided cordierite in the case of green cordierite alone or with Si₃N₄ in the other ceramic samples. The composition for the specimens sintered at 1350°C showed irregular particles containing nano-sized grains of the major cordierite as observed by SEM. Consequently, clusters of nano-sized particles in the major cordierite were developed through all samples under nitrogen conditions. The bulk density of the sintered samples was between 2.255 and 2.305 g/cc in the air and 2.254 and 2.355 g/cc in...
nitrogen. The apparent porosities were between (13 and 17 %) in the air and (12.1 and 14 %) in nitrogen, respectively. The bending strength was between 50 and 325 N/mm². Generally, these values were higher in the air than those in the nitrogen atmosphere.

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**Conflicts of Interest**

The authors declare no conflict of interest.

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