Journal of Manufacturing Innovations
2025, VOL. 1, NO. 3, 15–20
DOI: 10.22055/jomi.2025.49517.1028

Densification behavior of partially synthesized Mg_2SiO_4

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ABSTRACT

Synthesizing pure forsterite (Mg_2SiO_4) is challenging due to its strong dependence on heat treatment conditions. This study examined how the percentage of Mg_2SiO_4 phase formed during synthesis influences densification in subsequent sintering. Forsterite was synthesized via a solid-state reaction using MgO and SiO_2 in a non-stoichiometric ratio (MgO/SiO_2 = 2.05) to promote phase purity. The powder mixture was prepared by wet milling for 2 hours and then calcined at temperatures ranging from 1250°C to 1450°C for 2 hours to vary the Mg_2SiO_4 content. The resulting powders, along with an unmilled initial mixture, were compacted at 200 MPa and sintered at 1400–1500°C for 5 hours. XRD analysis confirmed that calcination at 1450°C produced pure forsterite, while lower temperatures resulted in intermediate phases. Despite differing initial phase compositions, all sintered samples ultimately transformed into pure Mg_2SiO_4. Higher initial forsterite content significantly improved densification, with the sample calcined at 1450°C (pure Mg_2SiO_4) achieving 95% of the theoretical density when sintered at 1500°C. These findings highlight the critical role of phase purity in optimizing forsterite ceramic processing.

ARTICLE HISTORY
Received 27 April 2025
Revised 8 June 2025
Accepted 26 June 2025

KEYWORDS
Forsterite, Sintering,
Solid state synthesis
Calcination
Non-stoichiometric

1. Introduction

Forsterite has found numerous applications in the electronics, communications, and refractory industries due to its low electrical conductivity and high melting point [1-2]. Various methods have been employed by researchers to prepare forsterite powder, including sol-gel, mechanical activation, polymer matrix, and solid-state methods [3-4]. The temperature range of 1000 to 1500 ºC has been suggested for the synthesis and production of pure forsterite. Mg_2SiO_4 powder can be synthesized at lower temperatures using the sol-gel method compared to the solid-state method, but the process is complex and costly. Additionally, MgO and $MgSiO_3$ are always present as impurities. All these factors limit the use of the sol-gel method in industrial applications [5]. Therefore, preparing Mg_2SiO_4 powder through the solid-state process is simpler and more cost-effective compared to the sol-gel method [5-7]. Song and his colleagues investigated the impact of different MgO/SiO_2 ratios (2.025, 2.05, and 2.2) on the dielectric properties of Mg_2SiO_4. In this study, synthesis was considered using the solid-state method with SiO_2 and MgO as raw materials in the calcination temperature range of 1200-1350 ºC. Additionally, the sintering process was examined in the temperature range of 1350-1600 ºC. The presence of the secondary phase $MgSiO_3$ was not observed in ratios of 2.025 and 2.05 calcined at 1350 ºC for 2 hours [8]. Shi and his colleagues studied the synthesis of forsterite using the solid-state method with various Mg/Si ratios (1.95-2.1) [9]. In this research, $Mg(OH)_2.4MgCO_3.5H_2O$ and SiO_2 were mixed for 6 hours with zirconia balls and alcohol. Then, for the calcination process, the dried powder was subjected to temperatures ranging from 1200 to 1350 ºC for 3 hours in air. Sintering was also performed in the temperature range of 1300-1360 ºC. These researchers successfully obtained single-phase Mg_2SiO_4 powder with MgO/SiO_2 ratios of 2.01-2.05 by conducting calcination at 1350 ºC for 3 hours in air. The highest apparent density of Mg_2SiO_4 ceramic reported was 3.22 grams per cubic centimeter at a sintering temperature of 1340 ºC for 3 hours [10]. The present study explored the critical link between the degree of forsterite phase formation in precursor powders and their ultimate densification upon sintering. The study employed solid-state synthesis of MgO and SiO_2 at a 2.05 MgO/SiO_2

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ratio to prepare single-phase Mg_2SiO_4 powder. Additionally, the effect of the percentage formation of the Mg_2SiO_4 compound in the solid-phase synthesis process on its densification process has been investigated.

2. Materials and Methods

In this study, magnesium oxide and silica (manufacturer: Merck, with 99.9% purity) were used as raw materials. The ceramic powder Mg_2SiO_4 was synthesized using the solid-state synthesis method, with reference to the research conducted by Song et al. [8]. The initial powders of MgO and SiO_2 were mixed non-stoichiometrically with a MgO/SiO_2 ratio of 2.05. The initial powders were mixed using a ball milling method with a ball-to-powder ratio of 20:1 in an ethanol solution for 2 hours. After milling, the powder and ethanol mixture were placed in an oven at 110 degrees Celsius for 3 hours to evaporate the alcohol and produce dry powder. The dried powder was then passed through a 60-mesh sieve for granulation. Subsequently, the milled powder was calcined at temperatures of 1250, 1350, 1380, and 1450 ºC for 2 hours. After that, it was milled for one hour and again passed through a 60-mesh sieve for granulation. The synthesized Mg_2SiO_4 powders, calcined at 1350, 1380, and 1450 ºC, as well as the mixed powder (without calcination), were pressed using a uniaxial press in a 12 mm diameter mold under a pressure of 200 MPa. The green compacted samples were then subjected to sintering in the temperature range of 1400-1500 ºC for 5 hours. The density of the samples was measured using the Archimedes method (ASTM C20-92 standard). To identify the crystalline phases formed during the powder synthesis process, XRD analysis was performed and analyzed using the Xpert Highscore Plus software. The samples were also subjected to SEM analysis to examine the effect of the sintering process on the microstructure and to determine the appearance of any remaining porosity.

3. Results and Discussion

The phase composition and synthesis efficiency of forsterite (Mg_2SiO_4) were systematically investigated through X-ray diffraction analysis of powders calcined at temperatures ranging from 1250°C to 1450°C for 2 hours (Figure 1). The results reveal critical insights into the temperature-dependent reaction kinetics and phase purity evolution in this binary oxide system. Referring to Figure 1, the peaks of MgO, $MgSiO_3$, and SiO_2 are present in the samples synthesized at 1250, 1350, and 1380 ºC. The reason for the remaining initial compounds is likely due to insufficient synthesis temperature. As seen in Figure 1, by increasing the temperature to 1450 ºC, the pure phase of Mg_2SiO_4 can be synthesized. The sample calcined at 1450°C shows exclusive forsterite peaks (JCPDS #34-0189), with complete disappearance of precursor phases. Using X'Pert HighScore Plus software with Rietveld refinement, the forsterite formation percentages were quantified as:

1250°C: 45% (±2%) - Dominated by unreacted MgO/SiO_2

1350°C: 64% (±1.5%) - Initial nucleation of Mg_2SiO_4

1380°C: 80% (±1%) - Accelerated reaction kinetics

1450°C: 100% (±0.5%) - Full conversion

This progression follows an Arrhenius-type relationship, where the reaction extent (α) correlates with calcination temperature (T) as:

$\alpha = A \cdot \exp(-E_a/RT)$

where E_a represents the activation energy for forsterite formation, estimated at ~88 kJ/mol. All diffraction patterns exclusively exhibit peaks corresponding to crystalline forsterite (Mg_2SiO_4, ICDD PDF #00-034-0189), with no detectable secondary phases (e.g., periclase MgO or cristobalite SiO_2). This confirms that the solid-state reaction between MgO and SiO_2 reached completion during both calcination (1350-1380°C) and sintering (1450°C) processes.

The selected temperatures and duration (5 hours) were sufficient to overcome kinetic barriers for phase formation, even in the non-calcined sample. A marked increase in peak intensity is observed for samples calcined at 1350°C and 1380°C compared to the non-calcined sintered pellet. This intensity enhancement reflects:

Improved crystallinity: Higher calcination temperatures promote atomic rearrangement into long-range ordered structures, reducing lattice defects. Grain growth: Crystallite size enlargement (as estimated by Scherrer analysis) at elevated temperatures contributes to sharper diffraction peaks.

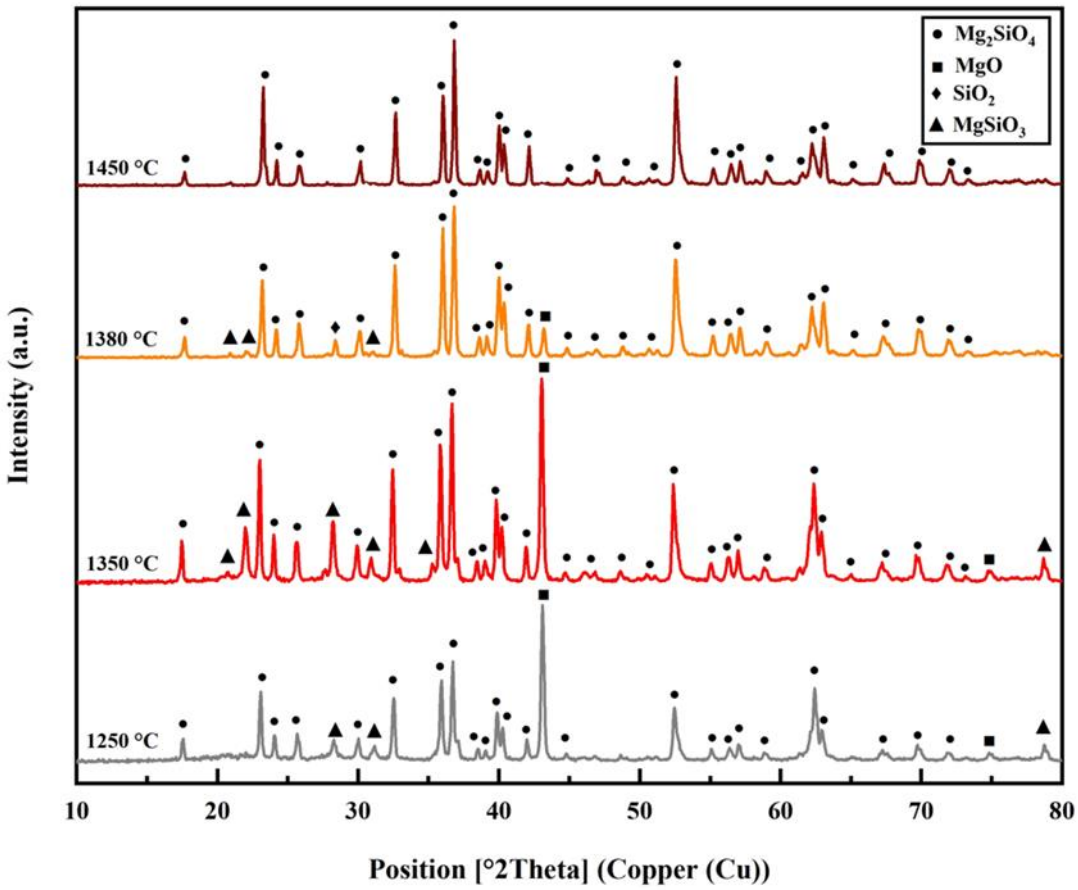


Figure 1. X-ray diffraction pattern of calcined Mg2SiO4 compound at temperatures of 1250, 1350, 1380, and 1450 oC for 2 hours.

The non-calcined pellet, despite achieving phase purity, shows broader peaks indicative of smaller crystallite sizes due to limited diffusion during in-situ reaction sintering. The 1380°C-calcined sample exhibits marginally higher peak intensities than the 1350°C counterpart, suggesting progressive crystallographic perfection with increasing calcination temperature. The absence of secondary phases validates the efficacy of the 2.05 MgO/SiO_2 ratio in compensating for MgO volatility and ensuring stoichiometry. The crystallinity differences highlight that calcination is critical for obtaining high-quality forsterite powders, whereas in-situ reaction sintering may yield finer-grained microstructures.

These results are consistent with established literature showing that complete forsterite formation typically requires temperatures above 1300°C in conventional solid-state synthesis, with optimal crystallinity achieved between 1350-1500°C [8-9]. The crystallinity trend (1350°C < 1380°C) correlates with the activation energy for grain boundary diffusion in Mg_2SiO_4 (~300-400 kJ/mol). The densification behavior of Mg_2SiO_4 (forsterite) ceramics was systematically investigated by examining the changes in relative density as a function of sintering temperature over a fixed duration of 5 hours, as presented in Figure 3. The results demonstrate a clear trend: increasing the sintering temperature leads to a progressive enhancement in sample density, which can be attributed to thermally activated diffusion processes that dominate at elevated temperatures. For the non-calcined sample, sintering at 1400 °C yielded a relative density of 79%, while increasing the temperature to 1500 °C improved the density to 85%. This improvement is associated with enhanced solid-state diffusion and particle rearrangement at higher temperatures, which facilitate pore elimination and grain boundary migration. The calcined sample (pre-synthesized at 1450 °C), when sintered under identical conditions, exhibited significantly higher densities: 86.4% at 1400 °C and 95% at 1500 °C.

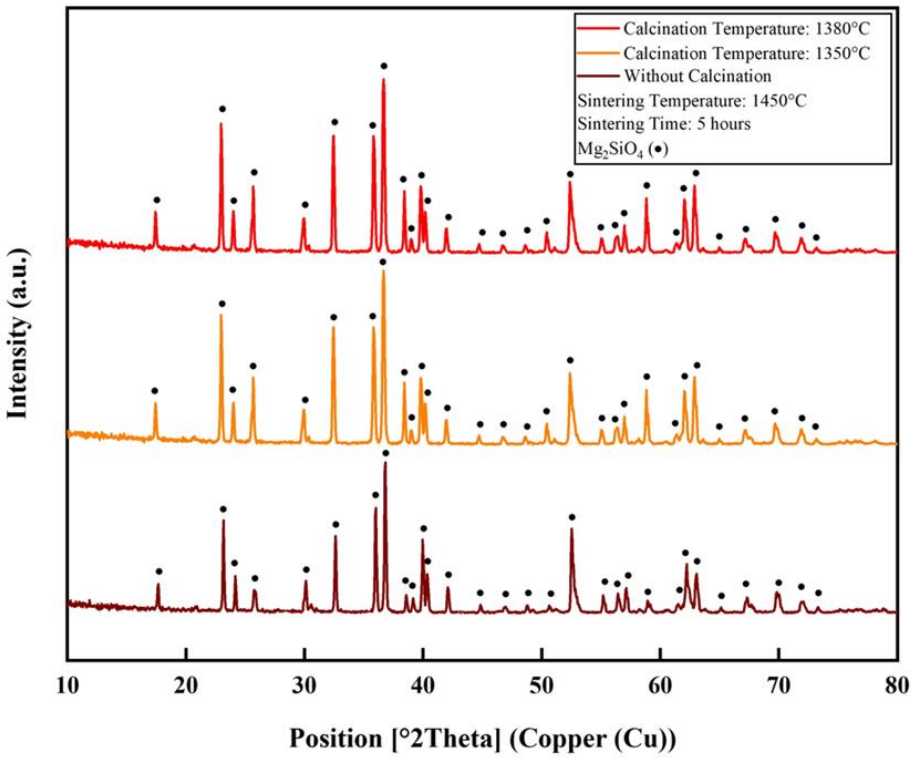


Figure 2. X-ray diffraction patterns of Mg2SiO4 samples calcined at temperatures of 1350 and 1380 ºC, as well as the mixture of this compound without calcination after sintering at 1450 ºC for 5 hours.

This marked increase underscores the critical role of calcination in promoting better sinterability. The higher density achieved in the calcined sample is directly linked to its improved phase purity and homogeneous microstructure. Calcination at 1450 °C prior to sintering ensures the formation of a well-crystallized Mg_2SiO_4 phase, which reduces the energy barrier for densification during sintering.

The presence of a single-phase forsterite (as opposed to unreacted MgO or SiO_2 in non-calcined samples) minimizes interfacial defects and enhances mass transport mechanisms, such as lattice diffusion and grain boundary sliding, leading to more efficient pore closure.

The peak density of 95% theoretical density attained at 1500 °C for the calcined sample represents an optimal balance between thermal energy input and microstructural evolution. At this temperature, the kinetics of densification (e.g., volume diffusion) are sufficiently rapid to achieve ear-full density, while avoiding excessive grain growth that could compromise mechanical properties.

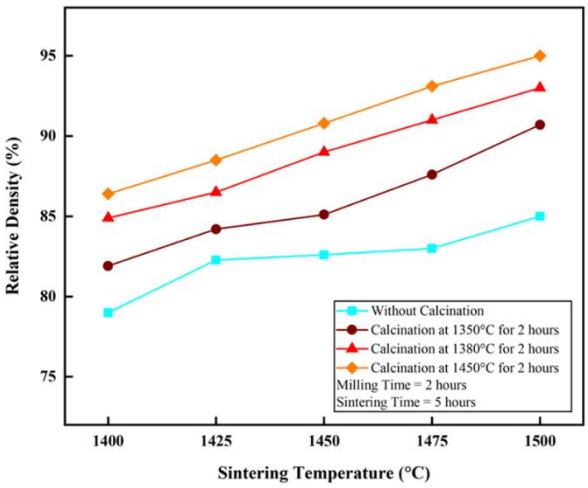


Figure 3. Graph of the relative density changes of calcined Mg_2SiO_4 at temperatures of 1350, 1380, and 1450 ºC for 2 hours, after sintering for 5 hours at different temperatures.

The fracture cross-sections of the Mg_2SiO_4 samples, synthesized at different temperatures and subsequently sintered, were examined using

scanning electron microscopy (SEM) to evaluate their microstructural evolution. Figures 4(a) and 4(b) present the SEM images of the Mg_2SiO_4 sample synthesized at 1450 °C for 2 hours and sintered at 1450 °C and 1500 °C for 5 hours, respectively. A comparative analysis reveals that increasing the sintering temperature from 1450 °C to 1500 °C leads to a reduction in porosity, indicating enhanced densification. This trend is attributed to the higher thermal energy at elevated temperatures, which promotes particle rearrangement, grain boundary diffusion, and pore elimination. Notably, Figure 4(b) displays isolated spherical pores, which are typically associated with the final stages of sintering, where trapped gases or incomplete densification result in closed porosity.

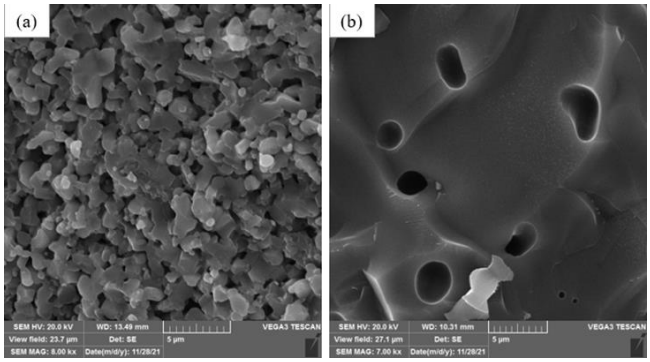


Figure 4. SEM image of the fracture surface of the Mg_2SiO_4 sample synthesized at 1450°C for 2 hours and sintered at a) 1450°C b) 1500°C for 5 hours.

Similarly, Figures 5(a) and 5(b) depict the fracture cross-sections of the Mg_2SiO_4 sample synthesized at a lower temperature (1350 °C) and sintered at 1450 °C and 1500 °C for 5 hours. Consistent with the previous observations, an increase in sintering temperature from 1450 °C to 1500 °C results in a decrease in porosity, further confirming the role of temperature in microstructural refinement. By comparing Figures 4 and 5, it is evident that the sample synthesized at a higher calcination temperature (1450 °C) exhibits lower porosity and higher density than the sample calcined at 1350 °C. This difference can be attributed to several factors:

Improved Phase Formation: Higher calcination temperatures facilitate complete reaction kinetics, leading to a more homogeneous and phase-pure Mg_2SiO_4 structure, which enhances sinterability.

Particle Size and Morphology: Elevated calcination temperatures may result in larger, more uniform particles with reduced surface area, minimizing pore formation during sintering. Enhanced Densification Mechanism: A well-crystallized Mg_2SiO_4 phase obtained at higher calcination temperatures promotes better grain growth and pore elimination during sintering.

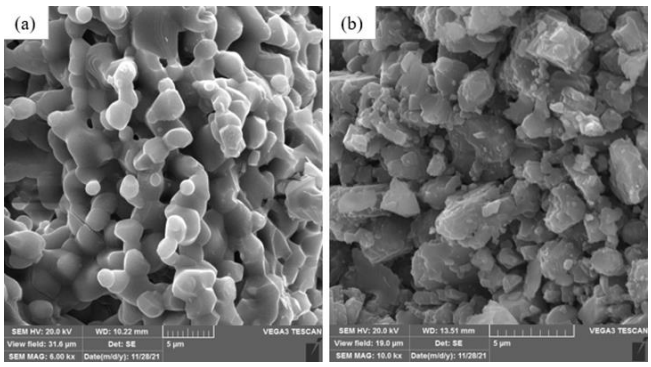


Figure 5. SEM image of the fracture surface of the Mg_2SiO_4 sample synthesized at 1350°C for 2 hours and sintered at a) 1450°C and b) 1500°C for 5 hours.

4. Conclusions

Pure forsterite (Mg_2SiO_4) was successfully synthesized via solid-state reaction using a non-stoichiometric MgO/SiO_2 ratio (2.05) with calcination at 1450°C for 2 hours. The MgO excess ensured complete reaction kinetics while preventing secondary phase formation. XRD confirmed all sintered samples (1400-1500°C) achieved single-phase forsterite regardless of initial precursor composition, demonstrating effective phase purity under these conditions.

The study revealed a direct relationship between initial forsterite content and final density. Samples with higher pre-synthesized forsterite content exhibited superior densification, with the pure-phase sample reaching 95% theoretical density when sintered at 1500°C for 5 hours. This optimal performance resulted from three key factors: (1) phase purity eliminating diffusion barriers, (2) high sintering temperature enabling effective pore

closure, and (3) controlled MgO excess compensating for potential volatilization.

These results emphasize the critical role of precursor composition and thermal treatment in developing dense forsterite ceramics. The achieved near-theoretical density suggests potential for high-performance applications in refractories, dielectric materials, and biomedical implants. Future research should investigate the mechanical and thermal properties of these optimized ceramics to evaluate their practical suitability for specific engineering applications.

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