The synthesis and properties of high-quality forsterite ceramics using desert drift sands to replace traditional raw materials

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High-quality forsterite ceramics were successfully prepared using desert drift sands to replace the traditional materials at lower temperatures, which not only saves mineral resources but also greatly reduces the cost of ceramic. Meanwhile, the effects of two different proportions of drift sands on the phase formation, physical and mechanical properties of ceramics were also investigated. The experimental results show that drift sands with impurity cations have the lower melting temperature compared to the pure quartz and easy to form liquid phase. As a result, the formation temperature (800°C) of forsterite phase falls by 300°C compared to the forsterite ceramic prepared using analytical reagent (1100°C). Furthermore, drift sands as raw materials can effectively improve the physical and mechanical properties of forsterite ceramics. Finally, the content of forsterite phase can reach up to 95.70 wt %, and the obtained forsterite ceramics show the best physical and mechanical properties when sintered at 1300°C for 2 h.

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1. Introduction

Forsterite (Mg2SiO4) is the most stable material in SiO2–MgO binary system.1) Besides traditional applications in refractory material, tunable laser and microwave telecommunication because of its high melting point (1890°C), good chemical stability, low dielectric constant and loss tangent,2) it also has potentials in biomaterials, such as the bone tissue regeneration and dental implants due to its high melting point (1890°C), good chemical stability, low dielectric constant and loss tangent,2) it also has potentials in biomaterials, such as the bone tissue regeneration and dental implants up to 1500°C for 2 h.9) A high-strength forsterite ceramic can be prepared using magnesium oxide (MgO, 97 wt %) and SiO2 (99.0 wt %) when sintered at 1200–1500°C, a maximum density of 91% and the highest mechanical properties were obtained respectively when sintered at 1500 and 1400°C.10) A forsterite was prepared utilizing 57.62 wt % serpentine and 42.38 wt % magnesium chloride water solution when sintered at 1300°C for 2 h, a small amount of SiO2, MgSiO3 and MgO still companied.11) The Ni, Co doped forsterite ceramic was synthesized via a sol–gel route and subsequent calcination at 1000°C for 3 h, the obtained ceramic contained a small amount of periclase (MgO), while the single-phase forsterite ceramic was obtained by adding NaCl mineralizer.12)

Traditionally, high-quality minerals such as magnesite, peridotite, serpentinite, talc and silica are used as raw materials to fabricate the forsterite ceramic and others that are applied in industry and daily life. The mineral resources are greatly reduced due to continuous consumption in fabrication of ceramics. In recent years, industrial wastes such as slags, fly ashes and coal gangues are used in the productions to reduce the consumption of minerals and to protect the earth environment from pollution. However, the data show that desert covers nearly 33% of the earth and most deserts are covered with drift sands.13) Drift sands consist of a large amount of silica and small amounts of calcia, magnesia, alumina, titania and ferric oxide, in which silica can be used in most of the ceramics14) and the impurity cations would play the role of sintering additives. If we can find a way to utilize the natural and abundant drift sands to replace the minerals in the preparation of ceramics, the mineral consumption, manufacturing cost and sintering temperature can be greatly reduced. Fortunately, the drift sands have been used to fabricate corindite and Si3N4O ceramics successfully.15–17)

However, a great attention has not been paid to the fabrication principles, fabrication techniques and evaluations of the ceramics. Therefore, the present work investigated the phase formation during the sintering process and the microstructure and properties of sintered products to seek for an effective route to fabricate high-quality forsterite ceramics (2MgO·SiO2) using desert drift sands instead of traditional raw materials.

2. Materials and methods

2.1 Materials

Drift sands were collected from Badain Jaran Desert, located in western Inner Mongolia of China. The chemical composition is given in Table 1. Moreover, industrial grade of magnesia (95.0%, Zehui Chemical Co., Ltd., China) and the AR grade of SiO2 (99.0%, XiLong chemical, China) and MgO (99.0%, XiLong chemical, China) were adopted.

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2.2 Methods
Two kinds of samples with different compositions were adopted: these are named Fo(A) with a stoichiometric composition of forsterite (42.70 wt% SiO₂, 57.30 wt% MgO) and Fo(B) with a quartz-rich composition (59.85 wt% SiO₂, 40.15 wt% MgO), in which drift sands and industrial magnesia were used instead of SiO₂ and MgO. The purpose of Fo(B) is to investigate the effect of excessive amounts of drift sands on forsterite ceramics, which will help to further decrease the cost of raw materials. In addition, a sample ARF with the stoichiometric composition of forsterite was prepared using the AR grade of SiO₂ and MgO as a reference in order to make some comparisons with Fo(A) and Fo(B).

2.3 Preparations
2.3.1 Powder preparation
The proportioned mixtures were wet-milled [mixtures:water:zirconia balls (wt %) = 1:1:3] in a planetary ball mill at a rotation rate of 200 RPM for 3 h. The average particle size 5.843 µm of the obtained powders was measured by a laser particle size distribution analyzer (BT-2003, Dandong Better Technology Co., Ltd., China) with water as their disperse medium, as shown in Fig. 1. Then, the powders were dried in a drying box at 120°C for 24 h.

2.3.2 Sample preparation
In order to improve the molding density, the water vapor produced by a water bath box was used for the powder humidification (moisture content was about 6–8 wt%), and then powders were pressed into cylindrical green disks (about φ50 × 10 mm) using a universal testing machine with a pressure of 30 MPa. Finally, the green disks were sintered with a heating rate of 10°C/min at 700–1300°C for 2 h in an electrical oven and cooled down to room temperature in air.

2.4 Test and analysis
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) and a scanning step of 0.02° at a speed of 3°/min. The contents of various phases in ceramics were calculated using MDI JADE via a whole pattern fitting method. The morphology of the ceramics was observed using a field-emission scanning electron microscope (FE-SEM, FEI, Quanta 650 FEI) and a benchtop scanning electron microscope (Benchtop SEM, JEOL, JCM-6000 NeoScope). The local composition was analyzed using an energy dispersive spectrometer (EDS, HORIBA, EMAX). The bulk density, apparent porosity and water absorption of the ceramics were investigated using the Archimedes drainage method according to national standard GB/T 3810.3-2006/[ISO 10545-3: 1995, MOD]. The Vickers hardness of ceramics were tested using a digital vickers hardness tester (TAIMING, HVS-30Z/LCD) with a test force of 24.518 N and for a duration time of 15 s according to national standard GB/T 16534-2009/[ISO 1044: 2008, MOD]. The three-point flexural strength of ceramics was determined using a microcomputer-controlled testing machine (Jinan Shijin, WDW-30, loading rate of 0.5 mm/min, 40 × 4 × 3 mm, span of 30 mm and 5 samples per group) based on the national standard GB/T 6569-2006/[ISO 1044: 2000, MOD].

3. Results and discussion
3.1 Phase formations of forsterite ceramics
The XRD patterns of samples sintered at different temperatures for 2 h are shown in Fig. 2. It can be seen that Fo(A), Fo(B) and Table 1. The chemical composition of drift sands, wt %

| Compositions | SiO₂ | Al₂O₃ | CaO | Fe₂O₃ | K₂O | Na₂O | Bal. |
|-------------|------|-------|-----|-------|-----|------|-----|
| Contents    | 78.91| 9.50  | 2.33| 2.26  | 2.08| 2.00 | 2.92|

![Fig. 1.](image1.png) The particle size distribution of the forsterite precursor powder after wet milling for 3 h.

![Fig. 2.](image2.png) XRD patterns of the samples sintered at different temperatures for 2 h: (a) ARF, (b) Fo(A) and (c) Fo(B). (unmarked peaks and #: forsterite, ▼: protoenstatite, +: spinel, ●: quartz, ■: periclase, ¥: albite).
ARF consist of different kinds of phases at the same temperature. Moreover, a large quantity of forsterite phases appear above 1200°C and the phase contents are given in Table 2. When sintered at 700°C, the Fo(A) sample consisted of quartz (JCPDS: 86-1630), periclase (JCPDS: 87-0651) and albite (JCPDS: 83-2215), while the Fo(B) sample was composed of quartz, protoenstatite (JCPDS: 76-1806) and albite. When sintered at 800°C, the forsterite phase (JCPDS: 85-1346) appeared in both Fo(A) and Fo(B) samples. The glass phase was resulted from the extra silica. In addition, the low-melting-point feldspar (Triclinic, framework structure) in drift sands forms large amount of liquid phase at 1100°C, which can also decrease the phase formation temperature by facilitating cations diffusion. It is known that the forsterite formation is a process that Mg\(^{2+}\) cations enter into the SiO\(_2\) lattices by di\(^{1+}\) cations in drift sands. Meanwhile, the forsterite phase occurred in the ARF sample at 1100°C, the forsterite phase formation temperature was much higher than that (800°C) of Fo(A) or Fo(B). When sintered at 1200°C, small quantities of spinel and quartz existed in Fo(A) samples, and some protoenstatite, quartz and glass existed in Fo(B) samples. The glass phase was resulted from the extra addition of drift sands and it can be identified by the enlarged base line view of Fig. 2(c) (up-convex base line). When sintered from 1250 to 1300°C, various phases in Fo(A) and Fo(B) had different changes. The forsterite phase in Fo(A) reached a maximum value of 95.70 wt\% at 1300°C, while a small amount of spinel and quartz remained. Meanwhile, the Fo(B) sample has a highest forsterite content of 80.31 wt\% at 1300°C, while a few protoenstatite, quartz and glass remained. This illustrates that the transition from quartz and protoenstatite to forsterite can be facilitated by increasing the sintering temperature. For the ARF, the forsterite and protoenstatite increased, and periclase and quartz decreased with an increase of temperature. These suggest that increase of temperature can improve the forsterite phase proportion in ceramics, while the over-burning phenomenon will occur in Fo(A) and Fo(B) ceramics to result in a decline in performance when sintered above 1300°C, which is the closely related to the more liquid phase caused by the low-melting-point of impurity cations in drift sands at high temperature. Moreover, for the ARF sample, the content of forsterite increased and the protoenstatite, periclase and quartz decreased with a continuous increase of temperature. Finally, the single-phase forsterite was formed at 1500°C. Overall, compared with ARF prepared using AR, drift sands instead of pure quartz can greatly reduce the formation temperature of forsterite phase, which makes forsterite ceramics form at low temperature.

### 3.2 Effects of drift sands on the phase formations of ceramics

Silica exhibits three crystal structures: quartz, tridymite and cristobalite. When sintering at high temperature, the volume of the silica will expand due to the phase transition. For the phase transition from α-quartz to β-cristobalite,\(^{19}\) silica in drift sands has a lower transition temperature (1200°C) than pure silica (1470°C). It is mainly due to the cations in drift sands like Na\(^+\), Ca\(^{2+}\), Al\(^{3+}\) leading to different crystal transformation of silica at a lower temperature.\(^{20,21}\) As a result, cations dissolve into the quartz structure of drift sands more easily than into that of pure silica. In addition, the low-melting-point feldspar (Triclinic, framework structure) in drift sands forms large amount of liquid phase at 1100°C, which can also decrease the phase formation temperature by facilitating cations diffusion. It is known that the forsterite formation is a process that Mg\(^{2+}\) cations enter into the SiO\(_2\) lattices by diffusion, so the diffusion ability determines the phase formation temperature of forsterite ceramic. By increasing the drift sands content, in a certain degree, liquid phase caused by impurity cations increases, which promotes the diffusion from Mg\(^{2+}\) to SiO\(_2\) lattice and reduces the phase formation temperature of forsterite.\(^{22,23}\) However, the content of forsterite could not be promoted continuously because a large amount of lower melting liquid phase can make part of forsterite (Mg\(_2\)SiO\(_4\)) transform into protoenstatite (Mg\(_2\)SiO\(_4\)) until the system reaches a thermodynamic equilibrium, as shown in Eq. (1).

\[
\text{Liq} + \text{Mg}_2\text{Si}_2\text{O}_5 \leftrightarrow \text{Mg}_5\text{Si}_3\text{O}_8
\]

(1)

Based on the above characteristics of drift sands, the phase formation temperature of forsterite prepared using drift sands is greatly reduced compared to the ARF sample due to Mg\(^{2+}\) cation entering into the SiO\(_2\) lattice in drift sands more easily than into that in pure silica.

### 3.3 Physical and mechanical properties of forsterite ceramics

#### 3.3.1 Surface morphology and phase distribution of forsterite ceramics

Surface morphology of the forsterite ceramics sintered at different temperatures is shown in Fig. 3. The surface morphologies of Fo(A), Fo(B) and ARF are very different. There is more porosity distribution in the Fo(A) ceramics and ARF ceramic, while there are a small number of evenly distributed closed pores in the Fo(B) ceramics. In addition, sintering temperature has less effect on the shape of the pores, just the size changes. It is generally known that the types and the number of pores usually affect the performance of the ceramic. Compared with the open pores in Fo(A) and ARF, the closed pores in Fo(B) ceramics are

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Table 2. Contents of phases in the samples sintered at different temperatures, wt %

| Sample | Condition | Forsterite | Protoenstatite | Periclase | Spinel | Quartz | Glass |
|--------|-----------|------------|---------------|----------|--------|--------|-------|
| Fo(A)  | 1200°C    | 92.80      | —             | —        | 3.20   | 4.00   | —     |
|        | 1250°C    | 93.65      | —             | —        | 3.25   | 3.10   | —     |
|        | 1300°C    | 95.70      | —             | —        | 3.30   | 1.00   | —     |
| Fo(B)  | 1200°C    | 57.44      | 24.05         | —        | —      | 2.61   | 15.90 |
|        | 1250°C    | 72.76      | 8.72          | —        | —      | 0.82   | 17.70 |
|        | 1300°C    | 80.31      | 3.02          | —        | —      | 0.67   | 16.00 |
| ARF    | 1200°C    | 43.38      | 1.80          | 24.26    | —      | 30.56  | —     |
|        | 1250°C    | 68.30      | 4.10          | 17.50    | —      | 10.10  | —     |
|        | 1300°C    | 80.50      | 16.40         | 2.20     | —      | 0.90   | —     |
|        | 1400°C    | 90.50      | 8.20          | 1.20     | —      | 0.10   | —     |
|        | 1500°C    | 100.00     | —             | —        | —      | —      | —     |
conducive to the performance of the forsterite ceramics, such as water absorption and mechanical properties.

Backscattered electron (BSE) images of Fo(A) and Fo(B) sintered at 1250°C for 2 h are shown in Fig. 4. It can be seen clearly from the BSE images that various phases are distributed in the ceramics. There are only dark regions (Point A) in Fo(A). Its element weight percentage is shown in point A. According to the results of XRD patterns (Fig. 2 and Table 2), the dark regions in Fo(A) are forsterite phases with small amounts of spinel and quartz. However, compared with Fo(A) and ARF, the Fo(B) has more complex phase distribution due to glass phase existing in it [Fig. 2(c) with the up-convex base line]. There are two types of regions in Fo(B): the dark regions B and the bright regions C, and the corresponding energy dispersive spectroscopy (EDS) analyses are shown in point B and point C, respectively. In addition, element weight percentages (Mg/Si/O) of forsterite (Mg$_2$SiO$_4$) and protoenstatite (MgSiO$_3$) with the stoichiometric composition were calculated, which were 34.55/19.96/45.49 and 24.21/27.98/47.81, respectively. It is observed that for the forsterite and protoenstatite, their corresponding element weight percentages (Mg/Si/O) are close to the values in energy spectrum analysis (Point B). According to the corresponding relationship of the element weight percentages, it can be confirmed that the dark regions B are the mixed phases of forsterite and protoenstatite, and the bright regions C are the glass phases. Furthermore, the area of the two regions in the BSE image indicates that the contents of forsterite and protoenstatite phases are much higher than those of glass phases in ceramics.
3.3.2 Bulk density, apparent porosity and water absorption of forsterite ceramics

The bulk density, apparent porosity and water absorption of forsterite ceramics are shown in Figs. 5(a)–5(c), respectively. The results showed that the bulk density of Fo(A) and ARF increased with the increase of sintering temperature, while Fo(B) exhibited an opposite trend. However, the ARF sample had the lowest bulk density in all samples, although it was sintered at 1500°C (Table 3). The apparent porosity and water absorption of all the samples decreased with the increase of sintering temperature, while ARF has the highest apparent porosity and water absorption. In general, the bulk density of Fo(A) is larger than those of Fo(B) and ARF, while the apparent porosity and water absorption of Fo(B) are superior to those of Fo(A) and ARF. This is closely related to the phase composition and sintering processing. In general, the ceramic bulk density is a sum of various phases in density. The Fo(A) samples are composed of forsterite (3.21–3.33 g/cm³), spinel (3.60–4.10 g/cm³) and quartz (2.59–2.65 g/cm³) phases, and the Fo(B) samples are composed of forsterite, protoenstatite (3.20–3.30 g/cm³), quartz and glass phases (with a loose structure) (Table 2). Based on the phase content of ceramics and the density of various phases, it can be concluded that the Fo(A) samples have higher density than Fo(B) and ARF samples, mainly due to Fo(A) samples consisting of the larger density sum of various phases caused by the dense phases such as spinel (3.60–4.10 g/cm³) compared to other samples.

On the other hand, unlike the Fo(A) and ARF ceramics, more drift sands are included in the Fo(B) ceramic, which contributes to the liquid phase sintering process and accelerates the densification of Fo(B) ceramic, so the apparent porosity and water absorption are greatly reduced. For the Fo(A) and ARF samples with few liquid phases, the connection and annexation process among the internal particles of the ceramic body would be more difficult in the sintering process.

3.3.3 Vickers hardness of forsterite ceramics

The Vickers hardness of forsterite ceramics is shown in Fig. 6. For the Fo(A) and ARF samples, Vickers hardness increased with an increase of sintering temperature, while for the Fo(B) samples, there was an opposite trend. The ARF sample had the lowest hardness (Fig. 6 and Table 4). In summary, the Fo(A) sample had the maximum hardness of 732.97 HV at 1300°C. Generally, the hardness of glass phase is much lower than that of other phases in ceramics. Unlike Fo(A) samples

Table 3. Physical properties of the ARF ceramics (single phase forsterite) sintered at 1500°C

| Frequency | 1  | 2  | 3  | 4  | 5  | Average |
|-----------|----|----|----|----|----|---------|
| Bulk density/g/cm³ | 2.28 | 2.30 | 2.31 | 2.29 | 2.32 | 2.30 |
| Apparent porosity/% | 27.21 | 27.98 | 28.10 | 26.32 | 26.74 | 27.27 |
| Water absorption/% | 11.79 | 11.23 | 12.03 | 10.98 | 13.17 | 11.84 |

Table 4. Vickers hardnesses of the ARF ceramics (single phase forsterite) sintered at 1500°C

| Frequency | 1  | 2  | 3  | 4  | 5  | Average |
|-----------|----|----|----|----|----|---------|
| Vickers hardness/HV | 227.41 | 230.32 | 227.30 | 231.53 | 226.44 | 228.60 |
containing large amounts of forsterite phases, there is more glass phase in Fo(B) samples, which results in the low hardness. For the ARF sample, the low hardness is caused by the presence of high open porosity and low density (Figs. 3 and 5).

3.3.4 Flexural strength of forsterite ceramics

The flexural strength of forsterite ceramics is shown in Fig. 7. The flexural strength of all the samples increased with an increase of sintering temperature. The Fo(B) sample had the maximum flexural strength of 110.76 MPa when sintered at 1300°C. For the ARF sample, its flexural strength was at a relatively low value of 20.18 MPa although the sintering temperature increased to 1500°C (Table 5). On the whole, the flexural strength of Fo(B) samples is superior to that of Fo(A) and ARF. The reason should be attributed to sintering behavior promoted by large amount of liquid phase. Especially, the Fo(B) ceramic has the highest flexural strength when the apparent porosity reaches a minimum of 0.73% at 1300°C [Fig. 5(b)]. Large amount of liquid phase in Fo(B) samples contributes to the formation of independent closed pores compared to the more open pores of Fo(A) and ARF samples, which reduces the apparent porosity (open porosity) of ceramics and improves their mechanical properties.

3.4 Effects of drift sands on the physical and mechanical properties of ceramics

The forsterite ceramics prepared using drift sands have high bulk density, low water absorption and good mechanical properties, which are closely related to the phase composition and microstructure. Compared with Fo(B) samples, the Fo(A) samples containing more dense phases have the higher bulk density and Vickers hardness, while the Fo(B) samples containing glass phase have the lower apparent porosity, water absorption and higher flexural strength. The ARF sample prepared using AR raw materials has lower density and poor performance. This is related to the phase composition of ceramics. Fo(A) contains more dense phase of forsterite (3.33 g/cm³, 92.8–95.7 wt %) and hard phase of spinel (4.10 g/cm³), which leads to the higher bulk density and Vickers hardness. The composition of Fo(B) samples contains more impurity cations introduced by drift sands. As a result, a large amount of liquid phase caused by impurity cations forms in ceramic, which fills in the particle spaces of ceramic to effectively decrease the apparent porosity and increase the flexural strength. Moreover, the migration and coalescence of pores are enhanced with an increase of liquid phase concentration, in favour of gases being excluded from the ceramic body. But for the ARF sample, almost no liquid phase appears in the sintering process, intergranular coalescence and nucleation might get really difficult even at high temperature, so its performance is difficult to be improved.

4. Conclusions

The forsterite ceramics were successfully prepared using desert drift sands to replace traditional materials via solid state reaction method. Compared with pure quartz, drift sands have lower melting temperature due to the effects of impurity cations. The impurity cations introduced by drift sands contribute to formation of liquid phase during the ceramic sintering process, which accelerates diffusion rate of Mg²⁺ in SiO₂ to form forsterite. As a result, the forsterite phase can be formed at a low temperature of 800°C, which is much lower than that of ARF sample (1100°C) prepared using analytical reagent.

In addition, the liquid phase caused by drift sands fills in the particle spaces to facilitate the migration and coalescence of pores, which effectively decreases the apparent porosity and improves the densification of ceramics. Therefore, the forsterite ceramics using drift sands as raw materials have higher physical and mechanical properties than ARF sample. Finally, when sintered at 1300°C, the Fo(A) forsterite ceramics with more dense phases have the highest forsterite phase content of 95.70 wt %, the maximum bulk density of 3.02 g/cm³ and the maximum Vickers hardness of 732.97 HV, while the Fo(B) forsterite ceramics with glass phase have the lowest apparent porosity of 0.73%, the lowest water absorption of 0.28% and the highest flexural strength of 110.76 MPa.

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