Effects of doping Al₂O₃/2SiO₂ on the structure and properties of magnesium matrix ceramic

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Abstract: To improve the sintering properties and the thermal shock resistance of magnesium matrix ceramic, this work used micron-sized MgO as the main raw material and nano-Al₂O₃ and SiO₂ as additives. Magnesium matrix ceramic was doped with different amounts of Al₂O₃ and SiO₂ (Al₂O₃ and SiO₂ at a molar ratio of 1:2) and sintered under standard atmospheric conditions. The effect of Al₂O₃/2SiO₂ doping amounts on sintering properties, thermal shock resistance, phase composition, and microstructure of composite ceramic materials were investigated. The results suggested that introducing Al₂O₃/2SiO₂ could improve the sintering properties of magnesium matrix ceramic. The grain boundary migration of its periclase phase could be hindered by the magnesium aluminate spinel and forsterite formed by solid reaction including Al₂O₃, SiO₂ addition, and the MgO itself. Furthermore, according to the mechanism of thermal expansion mismatching, with increasing Al₂O₃/2SiO₂ addition, thermal shock times increased significantly, the number of ceramic internal micro-cracks increased after the thermal shock and the material fracture surface energy was improved. It is found that the way of transgranular combining intergranular fractures can decrease the stress concentration at the crack tips and hinder crack propagation. The secondary phase particles deflected matrix internal micro-cracks and the crack propagation path length thus increased, residual stress field were subjected to interference and energy dissipation mechanisms improved. Meanwhile, thermal shock times for the material were enhanced and the degree of densification increased with increasing sintering temperature.

Keywords: ceramics, composite materials, sintering, thermal properties

1. Introduction

Magnesia material has high refractoriness, low thermal conductivity, high refractoriness under load, strong resistance to basic slag effects, etc.^[1,2]. The application of magnesia material is limited because of its large thermal expansion coefficient and poor thermal shock resistance^[3]. To improve the poor thermal shock spalling resistance, researchers, including Aksel *et al.*^[4] studied the thermal shock resistance of magnesia-spinel materials, it is believed that the thermal expansion mismatch between magnesia and spinel was significant. The thermal expansion mismatch could inhibit the accumulation of strain energy during thermal shock. Since spinel has a high melting point, high mechanical strength, relatively chemically inert and good thermal shock resistance.

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Ganesh *et al.*^[5-7] had introduced spinel into materials which played a critical role in improving the thermal shock resistance of magnesia-spinel^[8]; this has wide application prospects. Meanwhile, forsterite is a crystalline, refractory magnesium silicate with a low thermal expansion coefficient, low thermal conductivity, good chemical stability, and excellent insulation properties^[9]. In view of this, it can improve fracture toughness in the matrix ($K_{IC} = 2.4 \text{ MPa}\cdot\text{m}^{1/2}$)^[10], prevent internal crack growth and improve the sintering properties of the material.

Compared with traditional single-phase ceramics, ceramic composites that has become research focus on the study of high performance ceramics via improving the physico-mechanical properties, corrosion resistance, and fracture toughness^[11]. In accordance with the principles governing the toughness of ceramic composites, introducing Al_2O_3 into a magnesium matrix ceramic could form a magnesium aluminate spinel phase which generated thermal stress at the interface between the secondary phase particles and the surrounding matrix phase. Besides, due to the thermal expansion mismatch between periclase and spinel, there was a residual stress field which affected crack growth paths and improved the fracture toughness of the material^[12]. Meanwhile, introducing SiO_2 into the matrix could form a forsterite phase which enhanced the toughening effect, the thermal shock resistance, and other sintering properties. In the current work, introducing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ aims to improve the sintering properties and the thermal shock resistance of a magnesium matrix ceramic.

2. Experimental work

2.1 Raw materials

Commercial micro-magnesium oxide ($\text{MgO} \geq 98.5\%$, average grain size $3.31 \mu\text{m}$, and specific surface area $736.96 \text{ m}^2/\text{kg}$) was used as the main raw material, alumina (average grain size $64.19 \mu\text{m}$ and specific surface area $28.67 \text{ m}^2/\text{kg}$) and silica powder ($\text{SiO}_2 \geq 99.5\%$, average grain size 50 nm) were used as additives, and polyvinyl alcohol (at a concentration of $5 \text{ wt}\%$) were used as binders for the preparation of the magnesium matrix ceramic material.

2.2 Preparation

Al_2O_3 and SiO_2 were introduced into MgO with $n(\text{Al}_2\text{O}_3):n(\text{SiO}_2) = 1:2$ according to proportions of $0 \text{ wt}\%$, $15 \text{ wt}\%$, $30 \text{ wt}\%$, $45 \text{ wt}\%$, $60 \text{ wt}\%$, and $75 \text{ wt}\%$; the specific experimental formulation are shown in Table 1. The constituents were mixed by wet ball-milling for 3 h using ZrO_2 balls as the grinding medium, and then oven-dried at $60 \text{ }^\circ\text{C}$ for 8 h . The ceramic powders were pre-sintered at $600 \text{ }^\circ\text{C}$ for 2 h in a muffle furnace, then the pre-sintering powders were milled in an agate mortar for 30 min during which time polyvinyl alcohol was added to induce granulation. Finally the compound powders were pressed under a pressure of 10 MPa in a 769YP-24B tablet machine (the diameter of each specimen was 20 mm). The compacted ceramic green bodies were dried at $110 \text{ }^\circ\text{C}$, soaked for 24 h , and sintered at $1450 \text{ }^\circ\text{C}$ and $1550 \text{ }^\circ\text{C}$ for 2 h .

2.3 Characterisation

The porosity and bulk density of all specimens were measured according to Archimedes' principle. Using water cooling (from $110 \text{ }^\circ\text{C}$ to room temperature) the thermal shock resistance of the specimens was assessed. The phase compositions of specimens (after sintering) were analysed by X-ray diffractometer ($\text{Cu K}\alpha$ radiation, 40 kV , 100 mA , 0.02° , $4^\circ\cdot\text{min}^{-1}$, $0^\circ \leq 2\theta \leq 90^\circ$), at the

same time the **lattice constant** of the main crystal phase (periclase) was calculated by X ray diffractometry and supporting software (X 'pert High-score Plus). The crystal appearance and microstructure in the fracture surface of the specimens were examined on a Zeiss Σ IGMA field emission scanning electron microscope (Zeiss, Germany).

3 Results and discussion

3.1 Effects of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ on sintering property of magnesium matrix ceramic

XRD spectra of specimens sintered at 1450 °C and 1550 °C are shown in Fig. 1 which **displays** that the phase composition of Specimen 1 **is** periclase, the phase composition of Specimens 2 to 4 **contain** periclase, forsterite, and magnesium aluminate spinel. **In addition**, the diffraction intensity of the periclase phase decreased and the diffraction intensity of spinel and forsterite phases increased with increasing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition. It was suggested that Al_2O_3 and SiO_2 had gone into the matrix and combined with MgO . **Therefore**, magnesium aluminate spinel and forsterite were formed, respectively. Fig. 2 shows the $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$ ternary system phase diagram with the theoretical components of the experimental formula to **demonstrate** that the results agreed with those from XRD. The decreased periclase content **reduced** the diffraction intensity of the forsterite phase, but the diffraction intensity of the spinel phase increased further when the doping amount of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ further increased. At 75 wt% addition of $\text{Al}_2\text{O}_3/2\text{SiO}_2$, the phase composition of the specimen after sintering showed only the presence of magnesium aluminate spinel. In the phase diagram, **it is found that** this composition point **has** entered the primary crystal field and significant quantities of the spinel phase **are** generated at the same time. The diffraction intensity of spinel increased with increasing sintering temperature (comparing (a) with (b)).

The **lattice constant** and lattice volume of the sintering specimens according to 0 wt%, 15 wt%, 30 wt%, and 45 wt% of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition are as shown in Fig. 3. It was worth mentioning that the phase composition in sintered specimens of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ at contents of 60 wt% and 75 wt% had no periclase, so were not investigated. MgO **had** a face-centred cubic structure, O^{2-} **went** into the closest packing in face-centred cubic form, Mg^{2+} **occupied** octahedral sites, and the tetrahedral sites remain empty. According to the intensity of the ionic field (Z/r^2 , where Z **was** the valence and r the ionic radius) this **characterised** the extent of cation to anion attraction^[13,14], the value of Z/r^2 (10.48) of Al^{3+} **was** larger than that of Mg^{2+} (3.856), but lower than that of Si^{4+} (25.0). Hence, the integration of Si-O **turned** out to be most stable. In the $\text{MgO-Al}_2\text{O}_3\text{-SiO}_2$ ternary system, the inter-diffusion of Al^{3+} and Mg^{2+} **resulted** in spinel forming a solid solution^[15]. In the crystal structure of magnesium aluminate spinel MgAl_2O_4 , Al^{3+} **occupied** the octahedral position, Mg^{2+} **squeezed** into the tetrahedral sites and **remained** electrically neutral. The spinel solid solution **was** a cubic crystal system (Fd3m). The **interplanar spacing** d , indices of crystal face (hkl), and lattice constants a , b , and c can be calculated from Eqns (1) and (2).

$$\frac{1}{d_{hkl}^2} = \left(\frac{h}{a}\right)^2 + \left(\frac{k}{b}\right)^2 + \left(\frac{l}{c}\right)^2 \quad (1)$$

MgO has an isometric cubic crystal system ($a = b = c$), so Eqns (2) and (3) govern the MgO **lattice constant** and lattice volume.

$$a = b = c = d \times \sqrt{(h^2 + k^2 + l^2)} \quad (2)$$

$$V = a^3 \quad (3)$$

Fig. 3 shows that the influence of **lattice constant** and lattice volume, for doping with $\text{Al}_2\text{O}_3/2\text{SiO}_2$ on MgO: the **lattice constant** and lattice volume of MgO increased with increasing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition. **It was believed that** the increasing content of Al_2O_3 could promote the formation of structural **point** defects in the reactants **due to the lattice distortion** and increase the lattice energy. **It** could almost accelerate diffusion and enter into vacancies caused by the absence of Mg^{2+} and Al^{3+} ^[16]. An MgAl_2O_4 crystal nucleus **was** formed at grain boundaries or neighbouring lattice sites which increased the **lattice constant** and lattice volume^[17]. Indeed, at 30 wt% and 45 wt% $\text{Al}_2\text{O}_3/2\text{SiO}_2$ additions, the **lattice constant** and lattice volume of MgO were unchanged: this indicated that the solid solution effect of Al_2O_3 and MgO was limited^[18].

The influence of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ on the **linear dimension change** rate (LDCR) of specimens sintered at 1450 °C and 1550 °C is shown in Fig. 4, where it was **vital** to note that the surface of Specimen 6 appeared liquid, its edge melted and formed an even fracture, therefore its **linear dimension change rate** could not be measured accurately. The significant influence of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ on the **linear dimension change** rate of sintered specimens is seen in Fig. 4: the **linear dimension change rate** generally increased with increasing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition. These observations suggest that doping $\text{Al}_2\text{O}_3/2\text{SiO}_2$ can accelerate sintering of the ceramic matrix. **Besides, the linear dimension change rate** of specimens sintered at 1550 °C was higher than that at 1450 °C because the inter-crystalline structure became consolidated and the linear shrinkage rate increased with increasing sintering temperature; however, an inflection point at 45 wt% $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition **could fully evince that** the phase composition of sintered specimens included periclase, spinel, and forsterite at this time, the spinel content was increased and the formation of magnesium aluminate spinel occurred with a volumetric expansion of 5 to 7 %^[19]. This phenomenon could offset part of the volumetric shrinkage caused by sintering. The volume expansion of magnesium aluminate spinel played a main role therein, so the linear shrinkage rate was not obvious. On the other hand, the phase compositions of specimens under high temperature were spinel, forsterite, and a partial liquid phase at 60 wt% $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition. The key liquid phase promoted sintering; the liquid phase evaporated and the grains combined closely after slow cooling to room temperature. **Hence, it indicated that** the volume of sintered specimens changed **apparently**, and their linear shrinkage increased.

The effects of different **amounts** of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ on the bulk density and porosity of specimens sintered at 1450 °C and 1550 °C is shown in Fig. 5, the bulk density of sintered specimens increased and their porosity decreased with increasing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition at the two sintering temperatures used. **It is confirmed** that the value of Mg and Si electronegativity difference is greater than that of Mg and Al; generally the larger electronegativity difference **makes** it easier to form chemical compounds, and the smaller values **make** it easier to form a solid solution. The spinel content increased with increasing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition: Al_2O_3 entered lattice sites of MgO, its sintering properties **and its densification were increased**. **It is obvious that this information** agreed with that of **lattice constant** and lattice volume of the MgO. At 45 wt% $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition, there was an inflection point, the volume effect of spinel could largely offset the shrinkage from the sintering result in which the bulk density decreased. Then at 60 wt%

$\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition, the liquid phase content increased, the existence of this liquid phase enhanced the bulk density in some ways and reduced the porosity. Then, the bulk densities of specimens sintered at 1450 °C and 1550 °C were 3.450 g/cm³ and 3.389 g/cm³, respectively. Consequently, owing to the liquid phase was evaporated, the liquid phase content increased and the number of pores increased after slow cooling to room temperature: the porosity also increased with increasing sintering temperature. Therefore, the bulk density of specimens sintered at 1450 °C was higher than that of those sintered at 1550 °C.

3.2 Effects of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ on the thermal shock resistance of magnesium matrix ceramic

SEM images (magnified 2000 ×) in Fig. 6: (a), (b), and (c) indicate 0wt%, 30wt%, and 60wt% of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition to the specimens sintered at 1550 °C. The phase composition of Specimen 1 showed only periclase, the crystal structure was compact, and the grain boundary was clear. There were spinel and forsterite phases formed, as well as periclase. In addition, the periclase content, grain sizes, and porosity decreased with increasing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition. The analysis suggested that the formation of spinel and forsterite could hinder the movement of the periclase grain boundary and inhibit grain growth. Meanwhile, because of the inter-diffusion of Al^{3+} and Mg^{2+} , small spinel crystals may form in the enrichment zone at MgO-MgO grain boundaries. The pores within the periclase phase boundary were filled, and the degree of densification of the sintered specimens increased. With further increases in the amount of $\text{Al}_2\text{O}_3/2\text{SiO}_2$, in Specimen 5, quantities of an SiO_2 glass phase were precipitated in the boundaries and infilled the pores with a decreasing temperature because of the existing morphology of the SiO_2 glass phase at high temperatures, the bulk density increased. Fig. 6(d) shows the microstructure (magnified 3000 ×) of the white region highlighted in Fig. 6(c): the energy-disperse spectral analysis revealed that the phase composition of the sintered specimen included magnesium aluminium spinel and forsterite phases, the spinel content increased, and the grain growth was evident.

The thermal shock times of specimens sintered at 1450 °C and 1550 °C are shown in Fig. 7: it seems that the thermal shock time of the specimens increased with rising $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition. It is obvious that periclase phase has a high thermal expansion coefficient. With increasing temperature, its thermal expansion coefficient increased, elastic modulus decreased, and thermal shock resistance worsened. Therefore, at either 1450 °C, or 1550 °C, the thermal shock resistance of magnesium oxide ceramics was at its worst. In accordance with the principles governing the toughness of ceramic composites, MgO coupled with Al_2O_3 and SiO_2 formed spinel and forsterite respectively: the thermal shock resistance of the magnesium matrix ceramic was thereby enhanced. Spinel has a low thermal expansion coefficient and excellent thermal shock resistance. It can be seen from the XRD spectrum that, the periclase content decreased, the spinel content increased, and the thermal shock resistance of the magnesium matrix ceramic was improved upon increased $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition. On the other hand, specimen 4 exhibited low bulk density and large porosity large. It was demonstrated that the existence of pores could control thermal stress and reduce the concentration of stress at the crack front; the internal micro-cracking was deflected and the thermal shock resistance of sintered specimens was improved. When the sintering temperature was higher, the thermal shock resistance improved. Besides, as the specimen with 60 wt% $\text{Al}_2\text{O}_3/2\text{SiO}_2$, the higher the sintering temperature, the greater the amount of liquid phase generated. The cracks were more easily formed as defects at points of weakness, and as cracks

grew along the grain boundaries, the thermal shock resistance decreased.

The fracture SEM images of specimens sintered at 1550 °C by thermal-shocking at 1100 °C are shown in Fig. 8, where Figs 8(a), (b), and (c) are magnified 1000 × from samples at 0 wt%, 30 wt%, and 60 wt% Al₂O₃/2SiO₂; Figs 8(d), (e), and (f) are magnified 5000 × from each sample. As shown in Figs 8(a), (b), and (c), the number of cracks increased after thermal shock with the increase of Al₂O₃/2SiO₂ addition. Due to the difference between spinel and periclase, the thermal expansion mismatch mechanism in ceramic composites led to the formation of micro-cracks in specimens subjected to thermal shock. At the same time, the difference in phase composition and the composition of the specimens caused a difference in the thermal expansion mismatch. According to theory of thermal shock fracture, for short cracks, Eq. (4) applies:

$$R = \Delta T_c = \frac{\sigma_f(1-\nu)}{E\alpha} \quad (4)$$

Where R is the thermal shock parameter, ΔT_c is the critical thermal shock temperature difference for an unstable crack, σ_f is the fracture strength, ν is Poisson's ratio, E is the elastic modulus, and α is the coefficient of thermal expansion. Thus, the smaller the thermal expansion coefficient α , the larger the thermal shock parameter R , the better the thermal shock resistance of a material. The thermal expansion coefficient of spinel is smaller than that of periclase, so the thermal shock resistance of this ceramic could be improved by introducing the spinel, as secondary phase particles, within the periclase.

Compared with Fig. 8(a), cracks encountered the secondary phase particles and changed direction during propagation, the crack propagation path was thus extended, and the crack was narrow as seen in Fig. 8(b). The number of micro-cracks increased rapidly and most micro-cracks extended trans-granularly. On the one hand, trans-granular fractures needed a high activation energy to effectively reduce the stress concentration at the crack tip, so the trans-granular fracture process could effectively prevent cracks from further extension and formed many micro-cracks instead. On the other hand, according to the Hall-Petch fracture strength given by Eq. (5) and the fracture toughness (Eq. (6)):

$$\sigma = \sigma_0 + k_y d^{1/2} \quad (5)$$

$$K_{IC} = Y\sigma\sqrt{\pi a} \quad (6)$$

Where d is the average length of grain, σ_0 is the single crystal strength, K_{IC} is the fracture toughness, and a is half-crack length. Introducing nano-SiO₂, the average length of grain d decreased, the fracture strength σ increased, and the fracture toughness K_{IC} increased. At the same time, a partial SiO₂ glass phase was precipitated in the boundaries and infilled the pores at high temperatures, which could enhance the density of ceramic compounds and improve the thermal shock resistance of this magnesium matrix ceramic. Fig. 8(c) shows many microcracks which could inhibit the extension of the main crack and make a through-wall crack more unlikely to form: the structure of the ceramic was destroyed. Fig. 8(d) indicates that the cracks in the MgO ceramic, after thermal shock, mainly extended inter-granularly. The cracks were deep and through-wall cracks were formed; the energy dissipation mechanism was weakened. As shown in Fig. 8(e), Al₂O₃ and SiO₂ doping of a magnesium matrix ceramic made micro-cracks into trans-granular fractures and prevented their further expansion. Micro-cracks were formed in the internal main crack bifurcations. These could increase the effective surface energy along the main crack path

and disperse the stress concentration forming at the main crack tip. The crack propagation path was shortened and the thermal shock resistance of the magnesium matrix ceramic increased. Fig. 8(f) shows that, with increasing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition, the internal micro-cracks encountered spinel that the compressive stress of the second phase particle resulted in a region of crack deflection, and the increase in the length of the crack fronts and the fracture surface energy. Meanwhile, the stress intensity was decreased due to this disturbed thermal stress field, the crack propagation path became circuitous and further crack extension was hindered.

4 Conclusion

A magnesium matrix ceramic material was prepared by doping different amounts of Al_2O_3 and SiO_2 (Al_2O_3 and SiO_2 at a molar ratio of 1:2) on MgO by molding machine pressure and an atmospheric sintering process. The effect of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ doping on the sintering properties, thermal shock resistance, phase composition, and microstructure of the magnesium matrix ceramic were studied. Results suggest that magnesium aluminate spinel and forsterite are formed by doping Al_2O_3 and SiO_2 on MgO can hinder the grain boundary migration of the periclase phase, thereby inhibiting grain growth of periclase and improving the sintering properties of the magnesium matrix ceramic. In addition, a liquid phase, at high temperature, can promote sintering of the ceramic. With increasing $\text{Al}_2\text{O}_3/2\text{SiO}_2$ addition, the thermal shock time of each specimen increased, and the number of microcracks in the ceramic matrix increases after thermal shock. Micro-cracks were formed in the internal main crack. Intergranular fracture combines with trans-granular fracture are beneficial to enhance the fracture toughness. Furthermore, the existence of secondary phase particles cause the micro-cracks to deflect, the thermal shock resistance of the ceramic materials are thereby improved.

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FIGURE CAPTIONS

Fig. 1 Effect of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ doping on the phase composition of MgO specimens sintered at 1450°C (a) and 1550°C (b)

Fig. 2 $\text{MgO}-\text{Al}_2\text{O}_3-\text{SiO}_2$ ternary system phase diagram

Fig. 3 Effect of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ doping on the **lattice constant** and lattice volume of MgO specimens sintered at 1550 °C

Fig. 4 Effect of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ doping on the **linear dimension change rate** in MgO specimens sintered at 1450 °C

Fig. 5 Effect of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ doping on the bulk density and apparent porosity of MgO specimens sintered at 1450 °C (a) and 1550 °C (b)

Fig. 6 Fracture microstructure of specimens sintered at 1550 °C

Fig. 7 Effect of $\text{Al}_2\text{O}_3/2\text{SiO}_2$ doping on the thermal shock resistance of MgO specimens sintered at 1450 °C and 1550 °C

Fig. 8 Fracture microstructure of specimens sintered at 1550 °C after thermal shock

TABLE CAPTIONS

Table1 Formulation of experimental materials (wt%)

No.	MgO	Al ₂ O ₃	SiO ₂
1	100	0	0
2	85	6.9	8.1
3	70	13.8	16.2
4	55	20.7	24.3
5	40	27.6	32.4
6	25	34.5	40.5