

STRUCTURE AND MICROSTRUCTURE PROPERTIES OF A REFRACTORY CORDIERITE PREPARED FROM AMORPHOUS RICE HUSK SILICA RESULTING FROM PERICLASE INTRODUCTION

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ABSTRACT

A refractory cordierite ceramic prepared from amorphous rice husk silica is modified by the addition of 5 wt. % to 20 wt. % of periclase followed by a sintering treatment at a temperature of 1230 °C. The phases, the structural changes and the microstructure characteristics of the samples are investigated by x-ray diffraction (XRD) and scanning electron microscopy (SEM). The results obtained indicate the significant effect of the periclase addition on the phase decomposition of cordierite into spinel and forsterite. The addition of periclase is found to increase the presence of forsterite and decrease that of spinel. The formation of spinel and forsterite is connected with electrical resistivity and porosity decrease but density increase.

Keywords: cordierite, periclase, silica, rice husk, structure.

INTRODUCTION

At present, rice husk is a very attractive source of reactive silica as a raw material for the preparation of ceramics, since this agriculture residue has an abundantly high silica content. This kind of silica is shown to be a good material for the synthesis of many types of materials such as pure silicon, silica nitride [1] and silicon carbide [2]. In our previous investigations reactive silica has been obtained from rice husk by a simple acid leaching. It has been used for the production of several ceramics like borosilicate [3], carbosil [4], aluminosilicates [5], mullite [6, 7] and cordierite [8 - 10].

Cordierite ($2\text{MgO}\cdot 3\text{Al}_2\text{O}_3\cdot 5\text{SiO}_2$) is a material of a low thermal expansion and dielectric constant but a high thermal and mechanical stability. It is reported that the thermal expansion of cordierite is $2.2 \times 10^{-6}/^\circ\text{C}$ [11], while that of other ceramic materials refer to ca $3.3 \times 10^{-6}/^\circ\text{C}$ [8], $1-4 \times 10^{-6}/^\circ\text{C}$ [12], and $0.8-2 \times 10^{-6}/^\circ\text{C}$ [13]. Therefore, cordierite based materials are exten-

sively used in a broad range of applications, including catalysis, microelectronic and integrated circuit boards, membranes and refractories [14 - 17], electrical and thermal insulations [18, 19].

Several researchers study the feasibility of fabricating a dense cordierite ceramic by decreasing or increasing the content of MgO, Al_2O_3 and SiO_2 . It is necessary to consider in a number of applications where a change of the crystalline phase can affect the physical, thermal, and mechanical properties of cordierite. For example, Ye et. al. [19] synthesize cordierite by decreasing Al_2O_3 mole ratio from 2 to 1.4. They find a bulk density reaching a maximum value of 2.5 kg/m^3 , which is close to that of a dense cordierite ceramic. Meanwhile, an alumina addition to cordierite results in the formation of spinel, crystoballite and periclase phases [20]. Furthermore, the alumina addition increase from 10 wt. % to 20 wt. % leads to a significant increase of the density and the thermal conductivity of cordierite but electrical conductivity decrease.

The aim of this study is to identify the effect of MgO (periclase) addition on the structure and microstructure characteristics of cordierite prepared from amorphous rice husk silica. To gain insight on several basic characteristics, the structure and microstructure crystallisation of cordierite with periclase addition are studied by x-ray diffraction (XRD) and scanning electron microscopy (SEM).

EXPERIMENTAL

Sol silica was obtained by mixing 50 g of dried husk with 500 ml of 5 % KOH solution and boiling the mixture for 30 min in a beaker glass as reported in a previous study [7]. The preparation of cordierite was carried out by mixing MgO, Al₂O₃ and SiO₂ in a mass ratio of 2:2:5, correspondingly, as reported in a previous study [8]. The solid was ground using a mortar and sieved with a size of 200 meshes to obtain a powder. After that, periclase was added to the cordierite powder in a content referring to

0 wt. %, 5 wt. %, 10 wt. %, 15 wt. % and 20 wt. %. The powder was pressed in a metal die with the pressure of 2×10^4 N/m² to produce cylindrical pellets which were then sintered at a temperature of 1230°C with a heating rate of 3°C/min. The examination of the density and the porosity was done according to the Archimedes method [21]. The structure analysis was carried out using an automated Shimadzu XD-610 X-ray diffractometer, while the microstructural analysis was conducted with SEM Philips-XL. The electrical resistivity (ρ) was measured using the four-probe method.

RESULTS AND DISCUSSION

Fig. 1 shows the XRD patterns of the samples of a different periclase content sintered at a temperature of 1230°C. The phases identified on the ground of the PDF diffraction lines with the application of the search-match method [22] show clearly the presence of α -cordierite (PDF-13-0294) with a high peak intensity $2\theta = 10.5^\circ$,

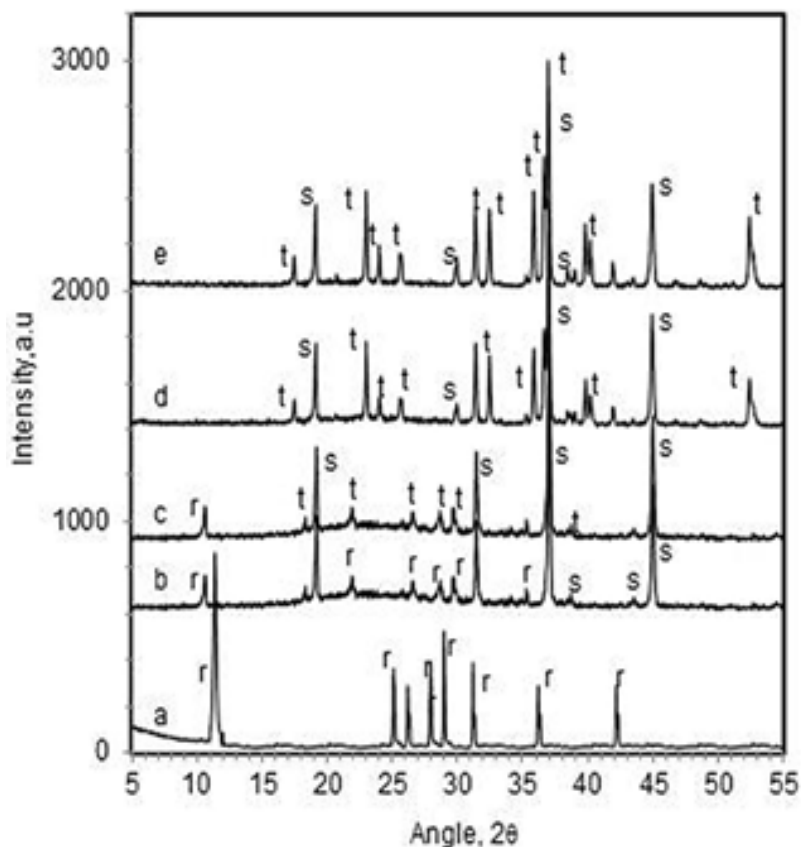


Fig. 1. X-ray diffraction patterns of samples sintered at 1230°C whose periclase content refers to: (a) 0 %, (b) 5 %, (c) 10 %, (d) 15 % and (e) 20 %; r = α -cordierite, s = spinel, t = forsterite.

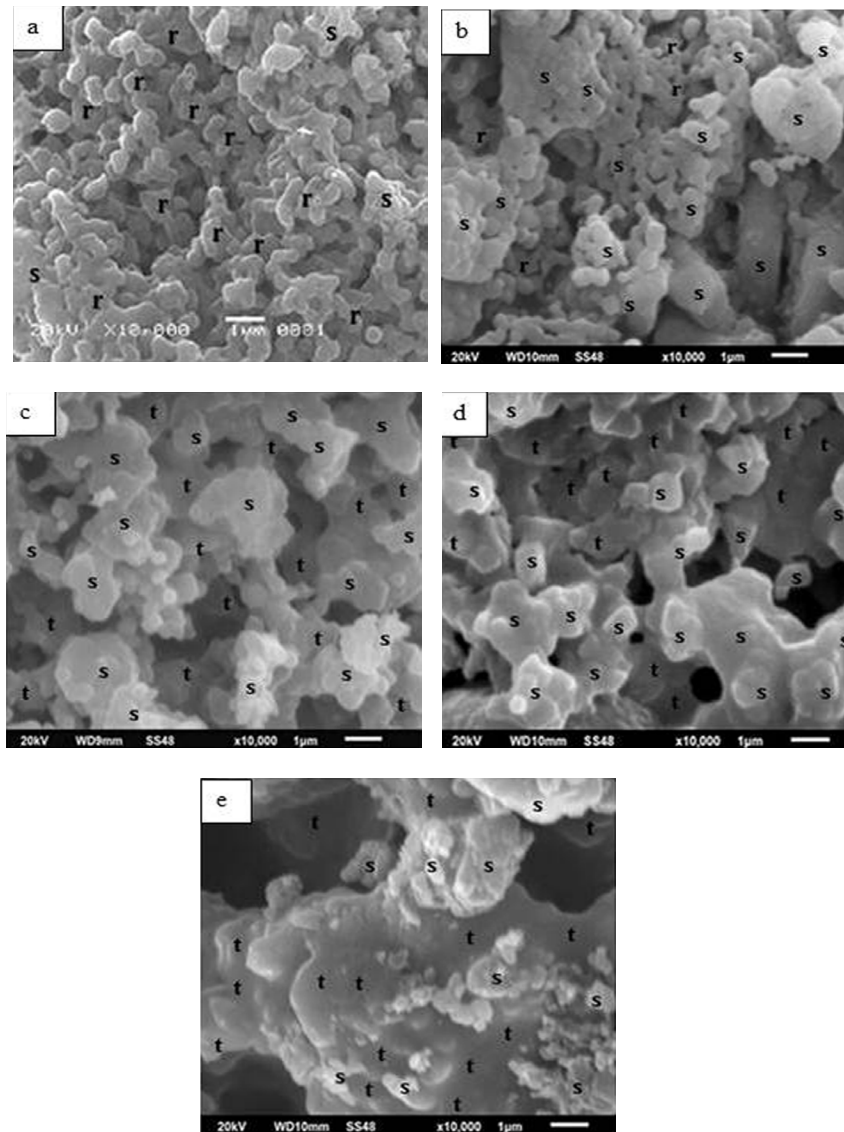


Fig. 2. SEM images of samples sintered at 1230°C whose periclase content refers to: (a) 0 %, (b) 5 %, (c) 10 %, (d) 15 % and (e) 20 %; r = α -cordierite, s = spinel, t = forsterite.

forsterite (PDF-34-0189) with $2\theta = 36.55^\circ$, and spinel (PDF-21-11520) with $2\theta = 36.91^\circ$. α -cordierite as a dominant phase is clearly detected in the sample containing no periclase (Fig. 1(a)). The spinel phase is well outlined, while α -cordierite presence is drastically decreased in the sample with 5 % periclase addition (Fig. 1(b)). The high presence of spinel is an indication that the 5 % periclase addition provides an intensive diffusive reaction between MgO and Al_2O_3 resulting in more spinel and less α -cordierite.

The α -cordierite is totally transformed into spinel and forsterite in case of 10 % periclase addition (Fig. 1(c)), which in turn leads to a spinel phase decrease. This

presumption is in accordance with the results observed with the sample containing 15 % of periclase (Fig. 1(d)). Spinel and forsterite are detected in this case, but the phase of forsterite is evidently increased. The phases considered remain practically unchanged even in case of 20 % periclase addition (Fig. 1(e)). The spinel is formed through the interaction between AlO_6 and MgO_6 octahedral [23], while the presence of forsterite results most likely from the interaction of SiO_4 and MgO_4 [24].

The surface morphologies of the samples studied are characterized by SEM. The results show a quite significant effect of the periclase addition, most likely as a result of crystallisation. The micrographs presented in

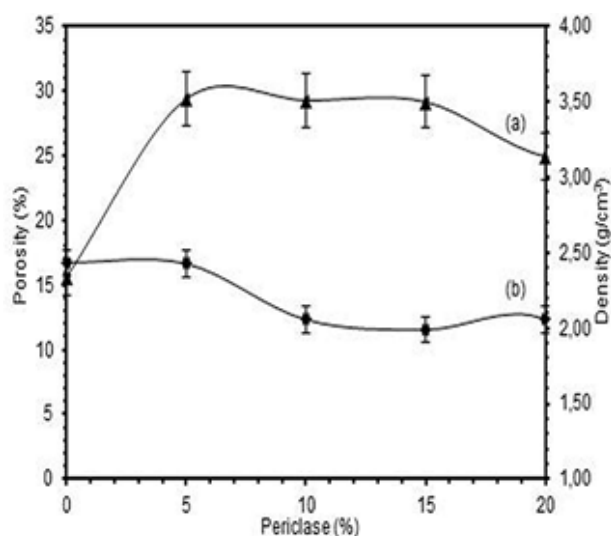


Fig. 3. Density (a) and porosity (b) as a function of the periclase content of the samples studied.

Fig. 2(a-e) illustrate the periclase effect on the size and distribution of the particles on the surface. As displayed by the images in Fig. 2(a-b), the surface morphologies of the samples are marked by the existence of particles of a different grain size and distribution.

The microstructure of the sintered sample containing no periclase (Fig. 2(a)) shows large grains with grain boundaries, while the images of the samples containing 5 % and 10 % of show no grain boundaries (Fig. 2(b-c)). In addition, it is obvious that the large grains in the sintered sample with 0 % of periclase are most likely composed of α -cordierite. This is supported by the result of the XRD analysis presented in Fig. 1(a), where α -cordierite is detected. The surface of the samples containing higher periclase content (15 % - 20 %) is occupied, as shown in Fig. 2 (d-e), by some large grains of spinel and forsterite. Both samples are marked by initiated coalescence of spinel as a result of α -cordierite crystallization. This feature suggests that with 15 % and 20 % periclase addition, α -cordierite phase continues to change leading to the formation of spinel and forsterite. This is supported by the XRD result presented in Figs 1(d,e). The formation of spinel and forsterite can be more clearly seen by inspecting the SEM micrograph of the sample containing 20 % of periclase (Fig. 2(e)).

The physical properties of the samples studied are shown in Fig. 3 and Fig. 4.

Fig. 3(a,b) shows the changes of the density and porosity of the samples as a function of their periclase

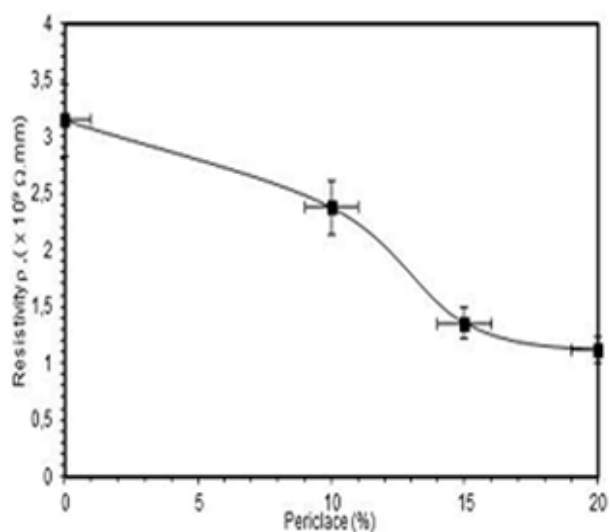


Fig. 4. Variations of the electrical resistivity as a function of the periclase content of the samples studied.

content. As evident, the density of the sample containing no periclase (Fig. 3(a)) increases drastically with the periclase content increase up to 5 %. A slow decrease is observed on further periclase increase up to 20 %. Fig. 3(a) shows that the densities of the samples increase from 2.36 g/cm³ to 3.45 g/cm³ with periclase content increase from 0 % to 5 %. It decreases slowly and reaches a value of 3.25 g/cm³ in 20 % periclase presence. The change of the density is most likely due to cordierite conversion into spinel and forsterite as evident from the XRD results (Fig. 1). These findings are in accordance with the results reported in refs. [25 - 27], which evidence that the density of the spinel and forsterite phases is higher than that of cordierite. The corresponding values cited there refer to 3.54 g/cm³, 2.96 g/cm³ and 2.3 g/cm³. The slow decrease of the porosity (Fig. 3(b)) with periclase content increase up to 5 % is attributed to decreased formation of cordierite. The porosity continues to decrease beyond this periclase content probably indicating a domination of spinel and forsterite, smaller particles distances and smaller particle sizes in the samples as a result of the higher periclase content. This is in accordance with the surface morphologies of the samples analyzed by SEM (Fig. 2(a-e)).

Fig. 4 shows the change of the electrical resistivity of the samples as a function of the periclase addition. It is evident that the higher the periclase content corresponds to lower electrical resistivity, which implies that the samples become resistance to electricity as a result

of the increased amount of spinel and forsterite. This conclusion is supported by the XRD results presented in Fig 1. This profile demonstrates that the electrical resistivity of the samples can be controlled through spinel and periclase formation. This is very useful for adjusting the suitability of the material to specified applications such as insulators and conducting elements in electronic devices.

CONCLUSIONS

This study demonstrates the effect of periclase addition on the structure and microstructure of cordierite prepared from rice husk silica. The addition of periclase from 5 % to 20 % reveals that cordierite formation is practically undetected, while spinel and forsterite are the dominant phases present. The phase transformation is found to change the characteristics of the samples referring to increased density because of cordierite conversion into spinel and forsterite as well as decreased porosity and electrical resistivity. The samples studied are considered insulators on the ground of the characteristics identified.

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