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Sinterability and thermal properties of cordierite ceramics prepared from Algerian kaolinite and magnesium hydroxide

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ABSTRACT

In this paper, we investigate the effect of MgO additions on the formation and densification behaviour of the cordierite obtained from some mixtures of Algerian kaolin and magnesium hydroxide. The sintering properties of these compositions have been evaluated by X-ray diffraction and bulk density. XRD analysis revealed that the major phase of the synthesized ceramics was cordierite along with a trace of spinel. Firing the pressed specimens at 1400°C for 1 hour yielded a dense cordierite ceramics with a relative density higher than 96%, a negligible open porosity and a lower linear thermal expansion coefficient of 2.73×10^{-6} K^{-1} between 200 and 800°C. Cordierite due to its very low coefficient of thermal expansion is considered as promising candidate for advanced applications.

KEYWORDS

Kaolin; Cordierite; Spinel; Sintering; Thermal properties

I. Introduction

Cordierite($2MgO.2Al_2O_3.5SiO_2$) ceramics present have low thermal conductivity, low dielectric constant [1,2] and excellent thermal shock resistance due to their low thermal expansion coefficient (from $2\times10^{-6}°C^{-1}$ to $6\times10^{-6}°C^{-1}$) [3,4,5]. They are widely used as a material in Kiln furniture, honey comb-shaped catalysts carriers in automobile exhaust systems and substrates in the micro electric applications [6,7]. The excellent properties exhibited by kaolin-based cordierite system make them suitable for the fabrication of catalyst support. Cordierite ceramics used for some applications need to have high density and good mechanical properties. However, cordierite is difficult to sinter without sintering aids because of the sintering temperature range which is just before its incongruent melting point [6]. Therefore, some appropriate methods were having been developed to prepare these ceramics. It is well known that sol-gel method has the advantage of an excellent control of chemical composition and the possibility of reducing the sintering temperature [8,9]. Addition of some sintering aids is another way to obtain high density cordierite materials at relative low temperature. The purpose of the present works is to study the effect of MgO additions on the formation and densification behavior of cordierite obtained from Algerian kaolinite.

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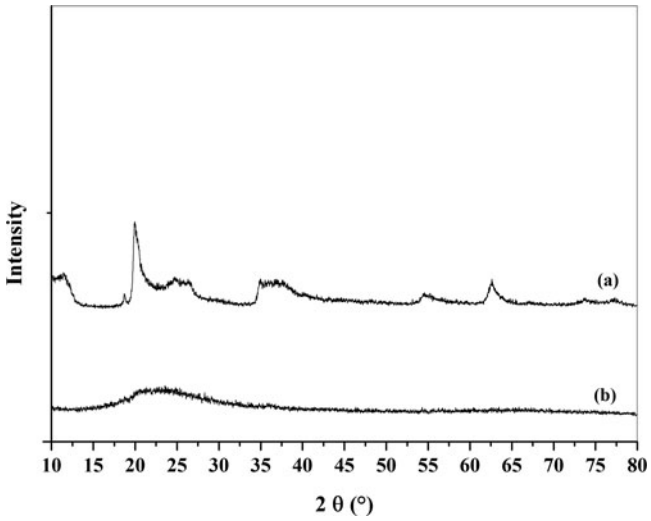

Figure 1. X-ray diffraction spectra of (a) kaolin and (b) metakaolin.

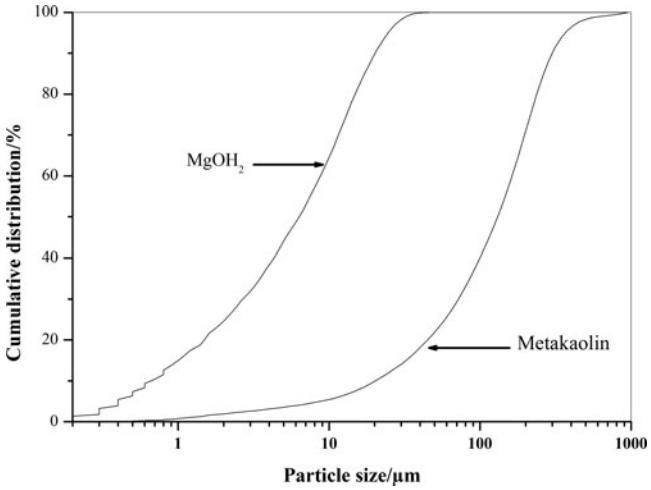

Figure 2. Particle size distribution of calcined Algerian kaolin and magnesium hydroxide

II. Experimental procedure

The starting materials used in this study were kaolin from (Djebel Debbagh - Algeria) and magnesium hydroxide. Before preparing the mixture, the crystalline kaolin was calcinated for 1 h at 700°C to yield metakaolin in amorphous state (Fig. 1). The chemical composition of metakaolin analyzed by X-ray fluorescence spectroscopy (XRF) is shown in Table 1. It contains a relative higher content of alumina. Particle size distribution of calcined kaolin and magnesium hydroxide are shown in fig 2. As it can be clearly seen, the powder of metakaolin presents

Table 1. Mean chemical composition of Algerian metakaolin calcined at 700°C for 1h.

Component	SiO_2	Al_2O_3	P_2O_5	CaO	SO_3	Fe_2O
wt%	53.056	44.405	0.027	0.170	0.020	0.064
Component	MnO	Co_3O_4	NiO	K_2O	ZnO	As_2O_3
wt%	1.549	0.136	0.093	0.022	0.088	0.371

a size distribution between 1 and 1000 μm with average particle size of ($D_{50} \sim 300$ μm). But the powder of magnesium hydroxide presents a size distribution between 0.1 and 100 μm with average particle size of (D50 $\sim$ 10 μm). After milling for 8 hours the average particle size of all mixtures is lower than 5 μm [10–12].

Metakaolin and magnesium compound were weighed in a molar ratio of $MgO.Al_2O_3.2SiO_2$ in order to compare the results. Cordierite powders were also produced with different amounts of magnesium hydroxide compound. These powders were wet-mixed and milled in distilled water for 8 h using zirconia balls in a high-energy planetary ball mill (Fritsch P6). All mixtures powder were dried at 110°C for 24 hours and then uniaxially pressed at 50 MPa in disks form with diameter of 13 mm. The green compacts were fired in air from room temperature up to 1400°C at a rate of 5°C/min in air. Above 1000°C, the firing rate was 2°C/min. After firing, the samples were cooled in the furnace.

Thermogravimetric (TG) and differential thermal analysis (DTA) tests were carried out done with the sample of kaolin heated at 20°C/min (SETARAM TG/DTA 92). The bulk density was determined by the Archimedes method. Density is defined as mass per unit volume. Any object, wholly or partly immersed in a fluid, is buoyed up by a force equal to the weight of the fluid displaced by the object. The weight of the displaced fluid is directly proportional to the volume of the displaced fluid. Crystalline phases were identified by X-ray diffractometry (Shimadzu mode 5600 with CuK_α radiation). The linear thermal expansion coefficient was measured using a NETZSCH DIL 402C dilatometer.

III. Results and discussion

III.1. Differential thermal analysis and thermogravimetric measurements

Fig. 3 shows TG/DTA and their derivative (DTG/DDTA) curves of kaolinite (heating rate of 20°C/min). Two endothermic peaks at 105°C and 508°C and an exothermic one at 984°C are present.

The first weight loss is due to the evaporation of adsorbed water corresponding entirely to the first endothermic peak; and the second weight loss in the temperature range 350–840°C is due to the dehydration of the kaolinite correlating with the second endothermic peak appeared at 508°C. The exothermic peak observed at 984°C is believed to be due to the formation of spinel or primary mullite [13, 14].

The samples were heated in a differential dilatometer up to 1200°C with a heating rate of 5°C/min. Dense α-Al_2O_3 was used as the reference material. The resulting data of length changes were differentiated using a computer program. The linear shrinkage was plotted against temperature for kaolinite at 1300°C (Fig. 4). Shrinkage of small extent first appears at 528 – 600°C intervals. This was due to loss of structural water with the formation of metakaolin. The second stage of densification starts at 915 – 972°C, and corresponds to the formation of mullite or spinel. The third stage of the densification process starts at the temperature 1128°C is due to the sintering of materails.

III.2. Sinterability

The bulk density of MgO-kaolin mixtures is changing with firing temperature and MgO content (Fig. 5):

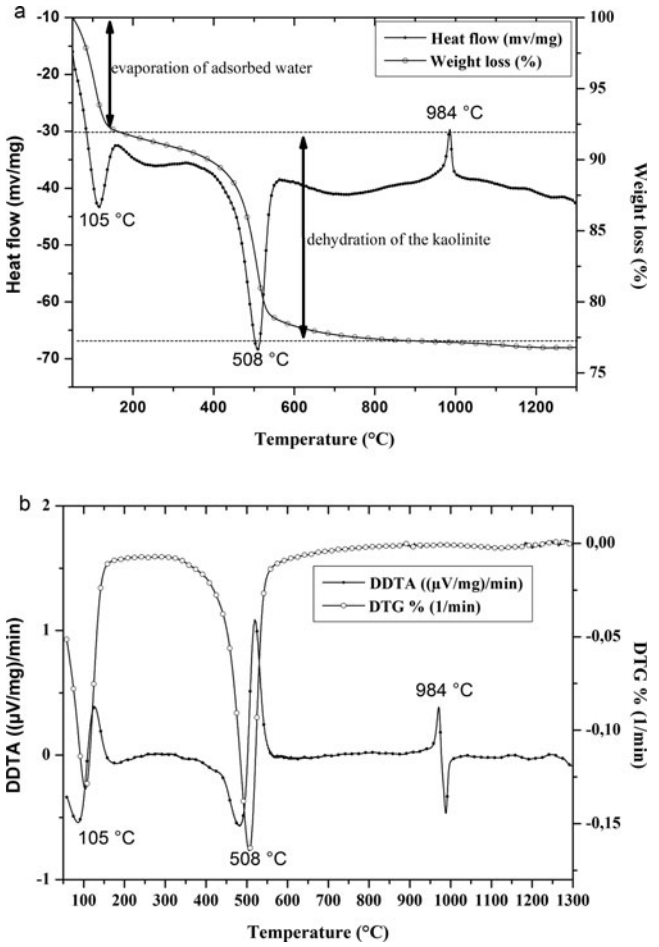

Figure 3. TG/DTA and DTG/DDTA curves for Algerian kaolin with a heating rate of 20°C/min.

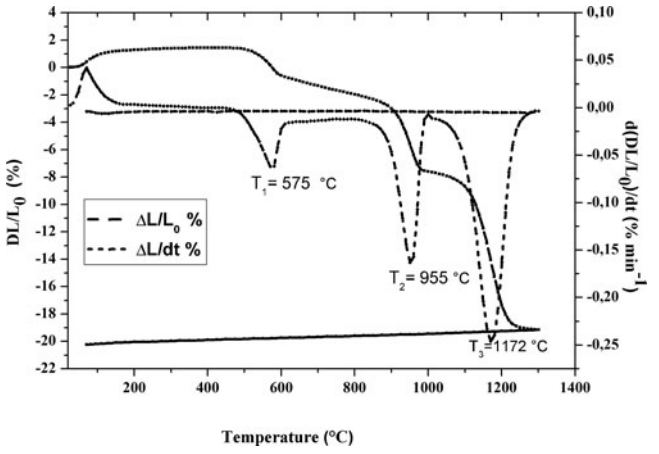

Figure 4. Linear shrinkage of kaolinite with heating rate of 5°C/min up to 1300°C.

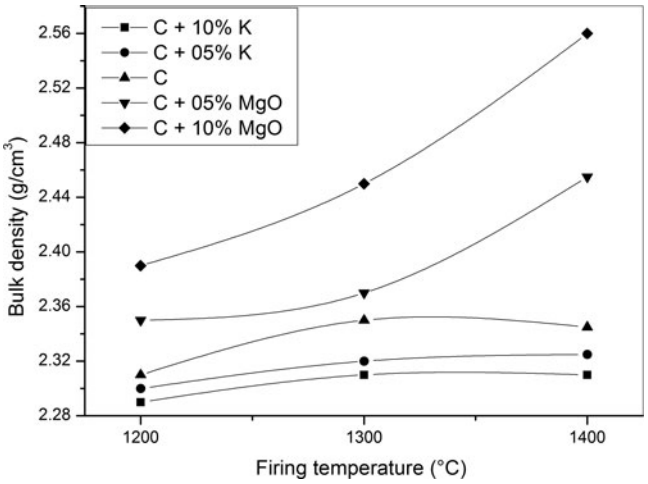

Figure 5. Bulk density of fired samples plotted against temperature and additive ratio of MgO) c: cordierite, k : kaolin.

- For the samples cordierite + 5 wt % kaolin, cordierite + 10 wt % kaolin and cordierite + 0 wt % MgO, there is a slight increase in density in 1200 – 1300°C interval, followed by a tendency toward a plateau in 1300 – 1400°C interval.
- For cordierite + 5 wt % MgO and cordierite + 10 wt % MgO samples, difference in sintering from 1300°C was significant and high relative densities were obtained (93.13% and 96.93% respectively) on 1400°C for 1 hour.

The bulk density of all samples showed a continuous increase with as the duration increases (Fig. 6). From this figure, it can be seen that increasing MgO additives allow improving the density.

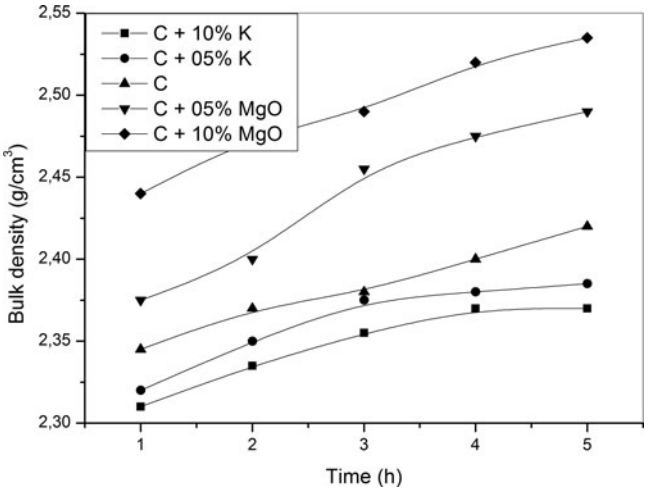

Figure 6. Bulk density of fired specimens plotted against time and additive ratio of MgO (C: Cordierite, K : Kaolin).

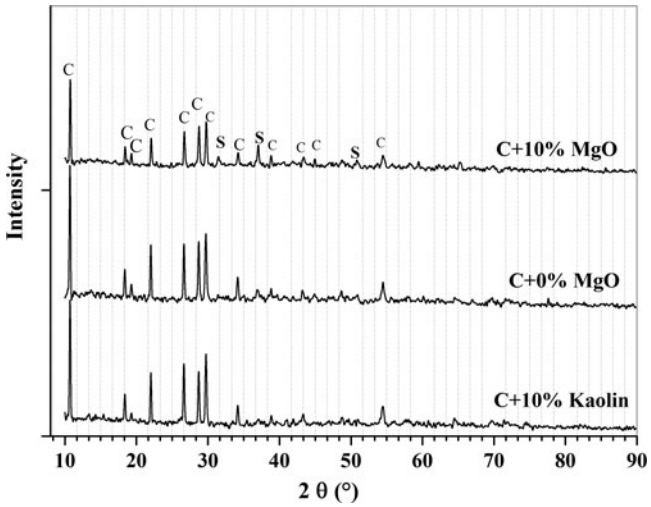

Figure 7. X-ray diffraction pattern of the samples sintered at 1400°C for 1h. (C: Cordierite, S: Spinel)

III.3. X-ray diffraction patterns

Figure 7 shows the X-ray diffraction patterns of the specimens fired at 1400°C for 1 hour. Cordierite and small amounts of Spinel were detected in all the specimens [15–20]. All the graphs show a continuous decrease in cordierite content and increase in Spinel content with increasing MgO addition.

III.4. Dilatometer measurements

The thermal expansion behavior of the samples sintered at 1400°C for 1h is shown in figure 8. The linear thermal expansion coefficient)α) of the samples containing 0 wt%, 10 wt % MgO and 10wt% kaolin fired at 1000°C are given in Table 2. it decreases with the addition of MgO.

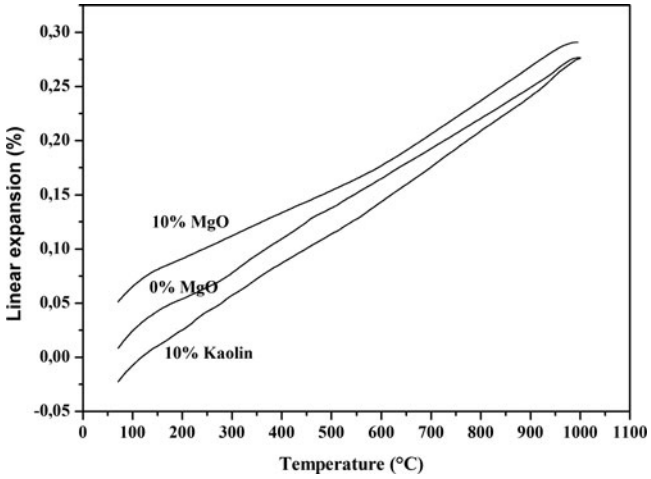

Figure 8. Thermal expansion curves of the samples sintered at 1400°C for 1h.

Table 2. Thermal expansion coefficient $\alpha.10^{-6}$ ($°C^{-1}$) of the sintered samples.

Temperature range	Cor. with 10% MgO	Cor. with 0% MgO	Cor. 10% kaolin
200–800°C	2.20	2.73	3.05
100–1000°C	2.51	2.80	3.14

IV. Conclusion

The effects of MgO addition on the sintering behavior of cordierite powders, oxide magnesium and Algerian kaolin were studied in this work. The results are summarized as follows:

- The bulk density of all samples increases with increase in MgO addition.
- Increasing MgO content of the starting composition decreases sintering temperature.
- Two phases (cordierite and spinel) were detected in all samples.
- The linear thermal expansion coefficient of the samples sintered at 1400°C for 1h decreases with the addition of MgO.

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