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An investigation on the properties and microstructure of mullite-bonded cordierite ceramics

M.Nouri. Khezrabadi[a], R. Naghizadeh[b], P. Assadollahpour[a] and S.H. Mirhosseini[a]
[a]*Academic Center for Education, Culture and Research, Yazd Branch, Iran*
[b]*Iran University of Science and Technology (IUST), Tehran, Iran*

The fabrication of a mullite-bonded cordierite body suitable for use as kiln furniture has been investigated in this article. First of all cordierite powder was synthesized by mixing talc, kaolin and alumina as starting materials, pressing and firing up to 1325°C. The amount of cordierite phase was estimated by an X-ray diffraction technique. The synthesized cordierite was then mixed with certain amounts of kaolin and alumina as the starting materials for mullite formation and the mixture was shaped by a pressing method, following by firing at different temperatures. The changes of density, porosity and cold crushing strength of the samples were measured as a function of heat treatment temperature. The results showed that these properties improve by a rise in temperature. The mullite formation within the samples was confirmed by an XRD method. The thermal expansion coefficient of the samples fired at 1350°C was determined by a dilatometer having an average value of 3.18×10^{-6} (1/K), which indicates an excellent thermal shock resistant for the samples. The microstructure of the fired samples was studied using scanning electron microscopy (SEM), which showed interesting results.

Keywords: Cordierite, Mullite, Physical properties, Micostructure.

Introduction

Natural cordierite having a chemical composition of $Mg_2Al_4Si_5O_{18}$ is a magnesium aluminosilicate mineral, occurring very rarely in nature. Cordierite ceramics show several superior high temperature properties such as excellent thermal shock resistance and chemical stability at elevated temperatures. In particular, the superior thermal shock resistance property enables them to be applied as kiln furniture [1, 2]. However, the relatively poor structural durability of cordierite ceramics should be improved for reliable long-term operation. Because mullite ($Al_6Si_2O_{13}$) ceramics have a high melting temperature, good resistance against creep deformation, and a moderate thermal expansion coefficient, the combination of cordierite with mullite would provide an advanced ceramic body with less degradation of the thermal shock resistance property. Additionally, the elongated mullite grains can be formed by the employment of an appropriate fabrication procedure [3-6] which may cause improved mechanical properties in the resulting ceramic body.

Cordierite mullite ceramics find increasing applications as refractory materials in the context of recent developments in fast-firing techniques of ceramic products. These refractories exhibit, in general, a complex microstructure characterized by crystalline phases of different thermal expansion coefficients and elastic modulii and a residual silicate glassy phase [7].

In the current study a mullite-bonded cordierite ceramic has been prepared using a mixture of synthesized cordierite and starting materials for the in-situ formation of mullite phase. The cordierite used in this study has been synthesized by mixing starting materials including talc, alumina and kaolin.

Experimental Details

Cordierite powder was synthesized by mixing talc, kaolin and alumina as starting materials. The mixture was pressed into cylindrical shapes by a hydraulic press under pressure of 20 MPa and fired at 1300 and 1325°C. The formation of cordierite as the major phase was studied using X-ray diffraction.

The synthesized cordierite was then mixed with certain amounts of kaolin and alumina as the starting materials for in-situ formation of mullite within the samples. The mixture was pressed into cylindrical shapes under pressure of 50 MPa, following by firing at 1250, 1300, 1350 and 1400°C. The formation of mullite within the fired samples was also studied by X-ray diffraction. The density and porosity of the samples and their mechanical properties were measured according to ASTM C0020-00 and ASTM C0133-03 respectively.

In order to estimate the thermal shock resistance of the samples, the thermal expansion coefficient of the

*Corresponding author:
Tel : 0098-351-8219360
Fax: 0098-351-8219361
E-mail: Nouri@acecr.ac.ir

sample fired at 1350°C was measured by a NETZSCH 402EP dilatometer.

Finally a microstructural study of the samples fired at 1300 and 1400°C was performed using scanning electron microscopy (SEM).

Results

Figure 1 shows the X-ray diffraction patterns of the synthesized cordierite powder. The presence of cordierite as the major phase is obvious in this pattern. Alumina and spinel also are present as minor phases.

Figure 2 shows the X-ray diffraction patterns of the cordierite-mullite samples after firing at different temperatures. It can be seen that the amount of mullite phase at 1250°C is not considerable, but it shows an increase as the temperature rises to more than 1300°C. Simultaneously the intensity of alumina peaks decreases indicating that transformation of alumina to mullite has happened due to the reaction with silica. The silica comes from structural decomposition of kaolin at high temperature.

Figures 3 and 4 show the changes of physical and mechanical properties of the samples as a function of temperature. Due to better sintering the highest density and lowest porosity belong to the samples fired up to

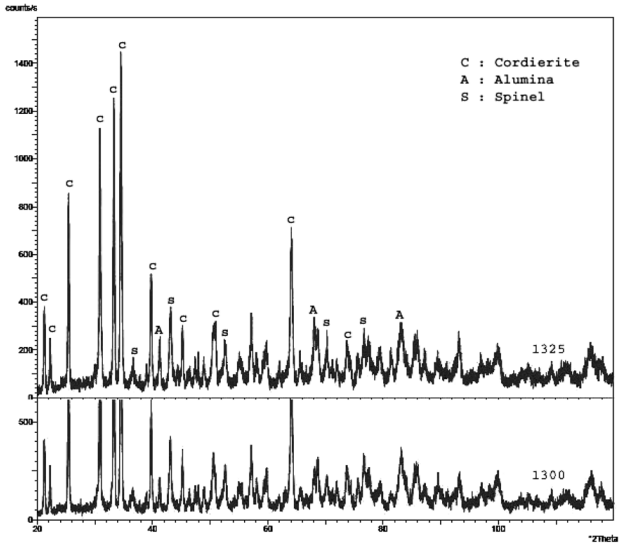

Fig. 1. X-ray diffraction patterns of cordierite powder.

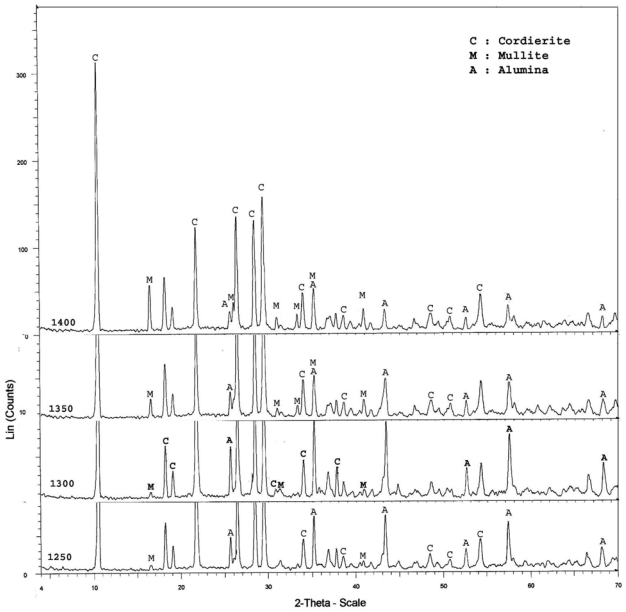

Fig. 2. X-ray diffraction patterns of cordierite-mullite samples after firing at different tempratures.

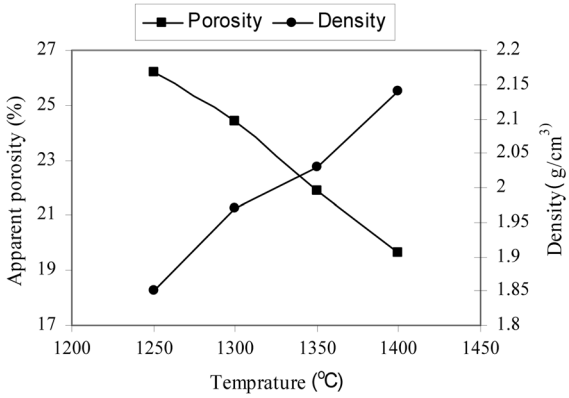

Fig. 3. Variations of density and porosity as a function of temperature.

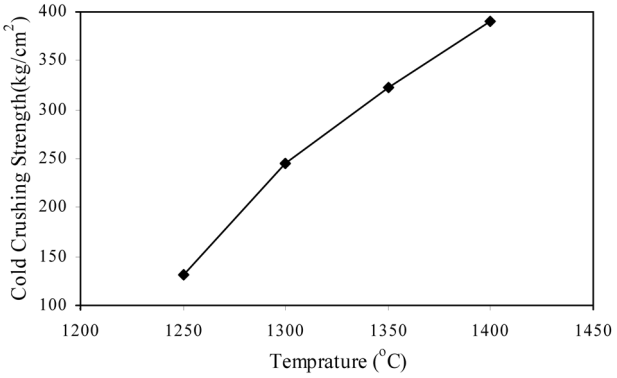

Fig. 4. Changes of mechanical properties as a function of temperature.

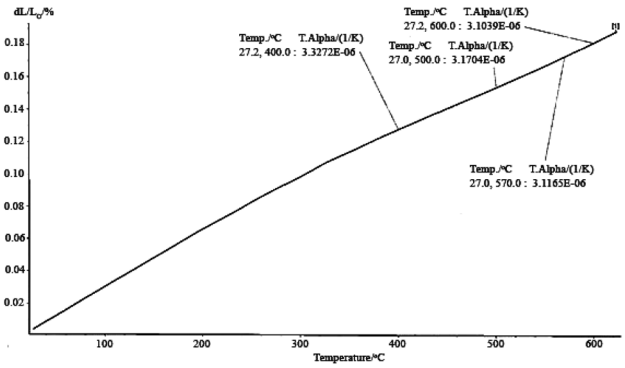

Fig. 5. Thermal expansion coefficient curve of the samples fired at 1350°C.

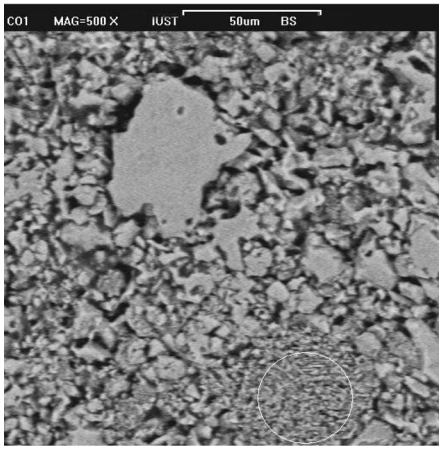

Fig. 6. Scanning electron micrograph of a sample after firing at 1300°C.

1400°C. They also show the highest cold crushing strength, which is related to the presence of larger amount of mullite at this temperature. Mullite has a needle-like structure that give rise to high mechanical strength.

Figure 5 shows the thermal expansion coefficient curve of the sample fired at 1350°C. The length changes have been measured from 25-600°C. The average thermal expansion coefficient in this range is nearly 3.18×10^{-6} (1/K). Regarding the thermal expansion coefficients of cordierite and mullite which are 1.7×10^{-6} and 5.3×10^{-6} (1/K) respectively, the cordierite mullite samples have a reasonable thermal expansion coefficient. This low thermal expansion coefficient leads to an excellent thermal shock resistance in practice.

Figure 6 shows the microstructure of a sample after firing at 1300°C. Energy dispersive spectroscopy (EDS) analysis of light particles demonstrated that they are the cordierite phase. In the lower part of the figure there is an island-like region apparently different from other parts, which has been marked with a white circle. Figure 7 shows this region at a larger magnification. EDS analysis of this part showed it to be the mullite phase, although this could be surmised from the needle-like structure of the region. This needle-like structure implies that the mullite phase has been nucleated from the melt. The melt is a result of the reaction between free silica and the alkalis that are present as impurities in the raw materials. Actually, the mullite formation process within the samples during firing can be described as follows. First of all, kaolin turns into metakaolin by the reaction:

$$Al_2O_3.2SiO_2.2H_2O \xrightarrow{450\text{-}550^oC} Al_2O_3.2SiO_2+2H_2O$$

At 925°C an exothermic reaction takes place and a type of spinel is formed:

$$Al_2O_3.2SiO_2 \xrightarrow{925^oC} 2Al_2O_3.3SiO_2+SiO_2$$

Between 1050 and 1100°C amorphous silica exits from the spinel structure and a type of mullite known as primary mullite is formed, which has a flake-like morphology. The related reaction is :

$$3(2Al_2O_3.3SiO_2) \xrightarrow{1050^oC} 2(3Al_2O_3.2SiO_2)+5SiO_2$$

The released silica then reacts with alkalis and a melt phase is created. With a rise in temperature alumina and the remaining silica dissolve in the melt and, after saturation, mullite crystals nucleate from the melt which are known as secondary mullite. At first the amount of the mullite phase is not very much and so this phase appeares as island-like regions.

Figure 8 shows the microstructure of samples after firing at 1400°C. There are not any island-like regions in this figure and EDS analysis showed that mullite is widespread between cordierite particles due to island growth. This mullite bond is responsible for the high mechanical strength of the samples fired at this high temperature.

Figure 9 shows a part of Fig. 8 at a higher magnification but the same magnification as Fig. 7. A comparison between Figs. 7 and 9 indicates that growth of

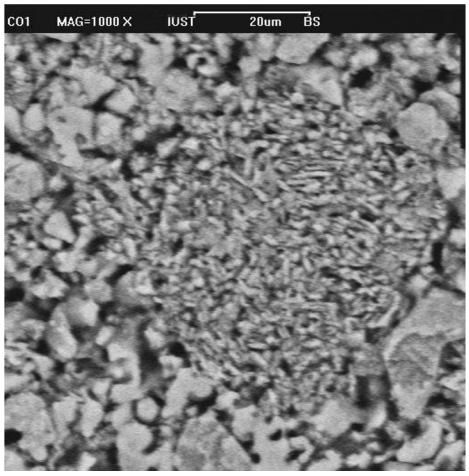

Fig. 7. Lower part of figure 6 at a larger magnification.

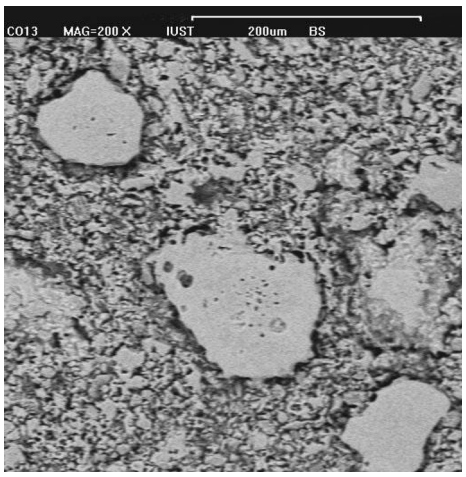

Fig. 8. Scanning electron micrograph of samples after firing at 1400°C.

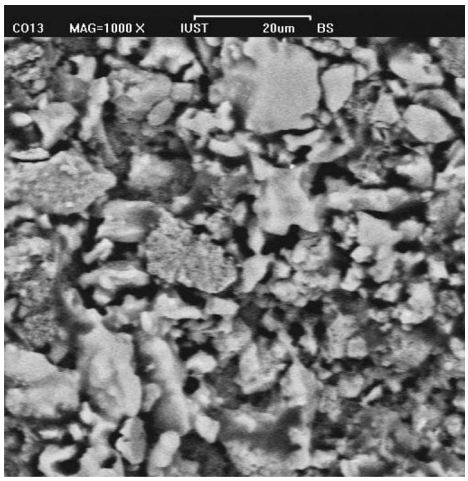

Fig. 9. A part of figure 8 at a higher magnification.

crystals has occurred at high temperature.

Conclusions

1. Synthesizing cordierite powder and mixing it with a certain amount of kaolin and alumina, leads to the in-situ formation of mullite, and a mullite-bonded cordierite ceramic body was fabricated.

2. The optimum temperature for firing of the samples is 1400°C which leads to the best physical and mechanical properties.

3. This mullite-bonded cordierite body has a thermal expansion coefficient of about 3.18×10^{-6} (1/K) giving rise to an excellent thermal shock resistance in practice.

4. Microstructural studies showed that mullite is formed as island-like regions at first but it becomes widespread in the body with a rise in heat treatment temperature.

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