Chiang Mai J. Sci. 2008; 35(1) : 6-10
www.science.cmu.ac.th/journal-science/josci.html
Contributed Paper

The Preparation of Cordierite-Mullite Composite for Thermal Shock Resistance Material

Phatthamon Kiattisaksophon [a], Sukdiphon Thiansem* [b]
[a] Department of Industrial Chemistry,Faculty of Science, Chiang Mai University, Chiang Mai, 50200, Thailand.
[b] Silicate Technology Center, Faculty of Science, Chiang Mai University, Chiang Mai, 50200, Thailand.
*Author for correspondence; e-mail: Sukdipown@hotmail.com

Received: 20 September 2007
Accepted: 15 October 2007

ABSTRACT

In this study aims to prepare cordierite-mullite composite to be use as a thermal shock resistance materials. Commercial available cordierite and mullite powders were used as starting raw materials with various ratios up to 100 wt% and were mixed together by ball milling in order to prepare a composite material. 0.6 wt% polyvinyl alcohol (PVA) was also added in the mixtures. The specimen was pressed under 1,500 kg/cm^2 and sintered between 1250^{o}C and 1400^{o}C with a temperature interval of 50^{o}C. The thermal expansion coefficient was measured and observed by loop of thermal behavior when heated up to maximum temperature and cooled down to room temperature. Thermal behavior was studied. Thermal shock resistance was measured following ASTM method. The microstructure of composite has been characterized by scanning electron microscope (SEM). The result revealed that the percentage of cordierite-mullite at 70:30 wt% and sintered at 1400 ^{o}C showed good results in both of physical and thermal properties.

Keywords: Cordierite, Mullite, Thermal expansion, Thermal shock resistance.

1. INTRODUCTION

Cordierite (2MgO. 2Al_2O_3. 5SiO_2) and mullite (3Al_2O_3.2SiO_2) represent technically important ceramics which are applicable in a variety of areas. The cordierite ceramic due to its low dielectric constant (e = 5-6), high resistivity (r>10^{12} Ωcm), high chemical durability, high resistance to thermal shock and very low thermal expansion coefficient (a = 1-2 x 10^{-6} C^{-1}) are a promising candidate in many applications. Some examples of the applications are electrical porcelains, catalytic converter substrates for exhaust gas control in automobiles, heat exchanger for gas turbine engines, industrial furnaces, packing materials in electronic packing, refractory coating on metals, integrated circuit substrate, etc. However, cordierite has the disadvantage of its inferior mechanical properties. Moreover, cordierite ceramics are difficult to sinter without any sintering aids because its help the densification by liquid-phase process. Unfortunately, the electrical and thermal properties of cordierite are degraded by sintering aids [1-3, 6]. Mullite is used as structural materials due to its excellent mechanical properties even at high temperatures and is also considered a suitable material for electronic packages [4,5]. By formulating cordierite-mullite composites is possible to improve the mechanical behavior and thermal properties.

The objective of this paper aims to study the best fraction of the binary mixtures between mullite and cordierite and to observe the best condition for the preparation of cordierite-mullite composites to be used as refractories as well.

2. Materials and Methods

The starting raw materials used in this study were commercially available powders of cordierite and mullite (Northern-Chemical and Glassware Company, Chiang Mai, Thailand). As-received cordierite and mullite powder were milled by ball mill for 8 h, sieving (150 mesh) and various ratio up to 100 wt% (Table 1). Cordierite-mullite mixtures were performed by homogenizing the powder in the speed mil and added 0.6 wt% polyvinyl alcohol (PVA) in the mixtures; help forming. Then, they were uniaxial pressed at 1,500 kg/cm^2 and sintered in an electrical furnace at 1250 ^{o}C and 1400 ^{o}C with a temperature interval of 50 ^{o}C for 2h using heating rate of 5 ^{o}C/min.

Table 1. Ratio of cordierite and mullite: C = cordierite, M = mullite.

CM	1	2	3	4	5	6	7	8	9	10	11
C	0	10	20	30	40	50	60	70	80	90	100
M	100	90	80	70	60	50	40	30	20	10	0

Bulk density, apparent porosity and water absorption were determined according to ASTM, C 3733-88. Thermal expansion was determined with a heating rate of 10 ^{o}C/min up to 1100 C using an automatic dilatometer (dilatometer, L75). The microstructures of sintered materials were observed by scanning electron microscopy (SEM). Thermal shock resistance test was carried out according to the ASTM, C 1171-91 general method.

3. Results and Discussion

All mixes under investigation present the different properties of the fired bodies. The fired bodies that sintered at 1250 ^{o}C and 1300 ^{o}C are generally highly porous; the apparent porosity varied between 22 and 35% and the water absorption varied between 11 and 17%. While, the fired bodies that sintered at 1350 ^{o}C and 1400 ^{o}C bring to decreasing the apparent porosity and the water absorption when the ratio of cordierite much more 60 wt% (Figure1 and 2).

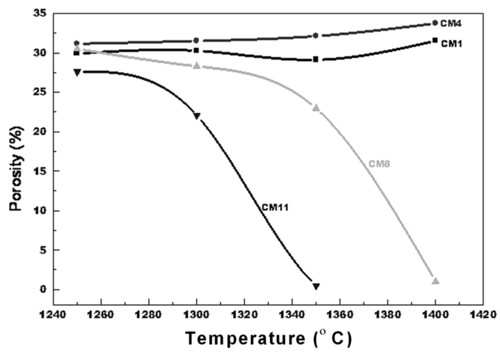

Figure 1. Change in apparent porosity with firing temperature.

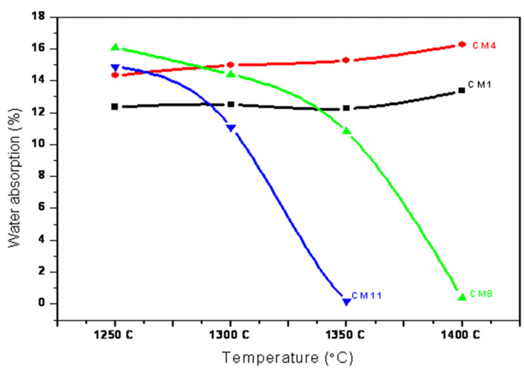

Figure 2. Change in water absorption with

Highly bulk densities were obtained at 1350 °C and 1400 °C for all bodies containing cordierite over 60 wt%. This might be the cordierite came to be the glassy phase and filled into mullite porous that affective to high density. So, the selected mixtures are CM1, CM4, CM8 and CM11. The bulk density values lie between 1.85 and 2.6 g/cm^3 (Figure 3).

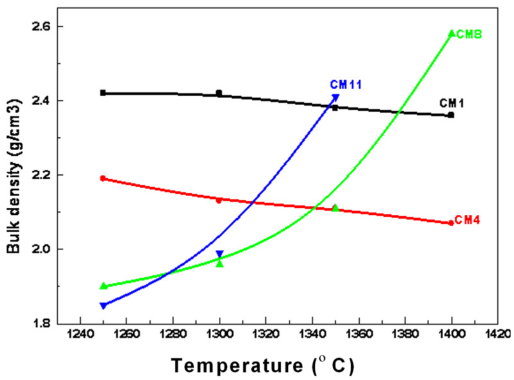

Figure 3. Change in bulk density with firing temperature.

The specimen of cordierite-mullite composite (CM1, CM4, CM8, CM11) were measured by dilatometer. The cordierite-mullite composite (CM8) showed the best behavior for all temperature. The specimen that fired at 1400 °C has the best thermal properties observed by loop of thermal behavior when heat up at temperature 1100 °C and cool down at room temperature. When the material had thermal stress, it did not storage stress in the body. The thermal expansion coefficient showed the values in Table 2. The behavior of thermal property showed in Figure 4.

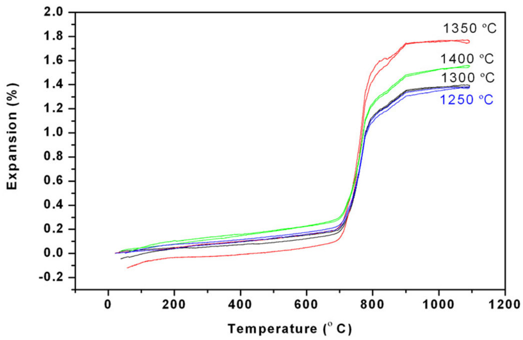

Figure 4. Thermal expansion behavior of cordierite-mullite at 70:30 wt% (CM8) in the different temperature.

Table 2. Thermal expansion coefficient of cordierite-mullite at 70:30 wt% (CM8) in the different temperature.

Fired Temperature (°C)	Thermal expansion coefficient ($\times 10^{-6}$ °C^{-1})
1250	3.10
1300	3.16
1350	4.52
1400	2.98

For this result, CM8 were selected to test thermal shock resistant following ASTM,C 1171-91. The body (CM8) can resist up to 8 cycles. At the 9 cycles, it showed the spot of microcracks in the body that observed by the eyes (Figure5).

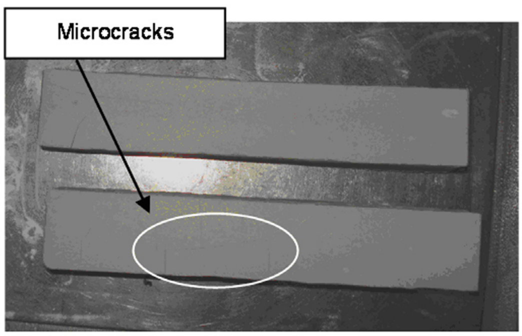

Figure 5. The spot of microcracks in the body of cordierite-mullite at 70:30 wt% (CM8) at the 9 cycles.

SEM micrograph of CM8 fired at 1250 °C (Figure 6) showed the orientation of grains mullite preserved in their original size as identified from the fine microstructure formed of 0.3□m mullite crystals. The mixture fired at 1300 °C showed grain growth of mullite crystals. Cordierite prisms crystallizing out as a reaction product at the mullite grain boundaries, due to the diffusion of Mg^{2+} ions from the neighboring cordierite grain or from the matrix. The substitution of Mg^{2+} ions for Al^{3+} ions was presenced the dislocations resulted in lattice strain (distortion). Therefore Mg^{2+} ions can give destruction of mullite crystal that it can penetrate into the mullite lattice as shown in Figure 7.

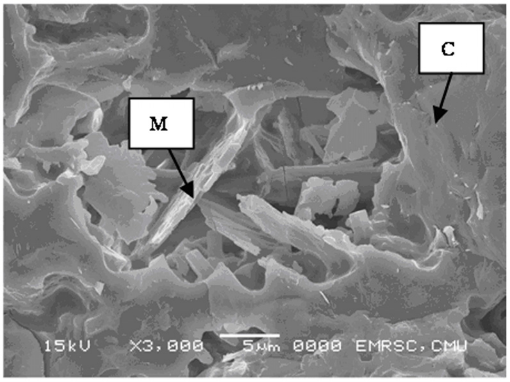

Figure 6. The orientation of grains mullite with the fine microstructure of the mullite crystals when fired at 1250 °C.

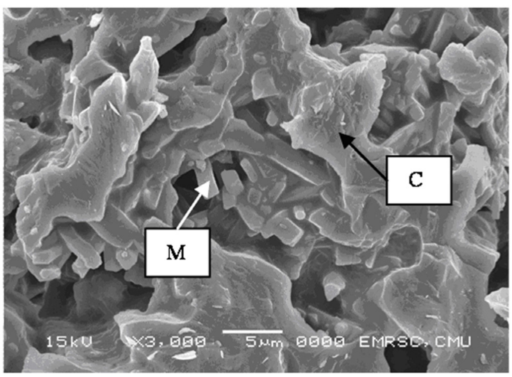

Figure 7. The destruction of mullite crystal when fired at 1300 °C.

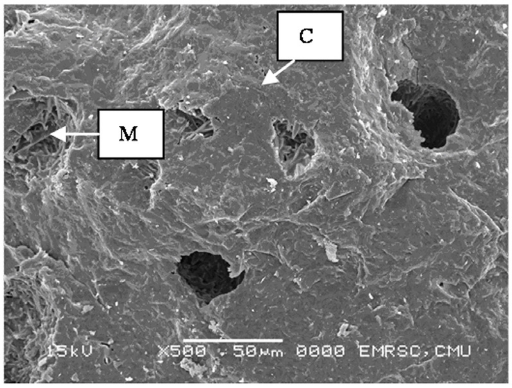

Figure 8. The overview of the densification body and variation in the size of pores when fired at 1400 °C.

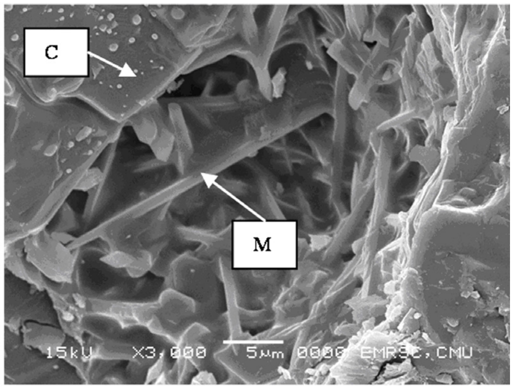

Figure 9. The orientation of mullite crystal in pores of body, as the network, when fired at 1400 °C.

The mixture fired at 1400 °C showed the densification of the body and variation in the pore size (Figure 8). The orientation of mullite crystal in pores of body exhibited as the network (Figure 9). Cordierite prism was formed in mullite matrix and melted in glass phase; filled in pores of mullite crystal structure. The pores of body were improved thermal behavior. When the materials received thermal stress, it could reduce thermal stress by pores and keep the deformation of materials. So the product, containing raw proportion of 70:30 cordierite-mullite and fired at 1400 °C, was the best condition for used as the thermal shock resistance materials.

4. CONCLUSIONS

The fraction of cordierite and mullite suitable for used as the thermal shock resistance materials is 70:30 wt% : cordierite-mullite and fired at 1400 ^{o}C. Because there achieve the best physical and thermal properties. Bulk density is 2.58 g/cm^3, apparent porosity is 1.00 % and water absorption is 0.39 %. The properties associated with thermal shock resistance and thermal properties; highly, porosity and water absorption are affected to reduce thermal shock resistance. Thermal expansion coefficient showed the value of 2.98 x 10^{-6} $^{o}C^{-1}$ that is very low. For thermal shock resistance testing, it exhibited 8 cycles. Finally, The SEM photograph showed densification of the body containing cordierite and mullite structure. These affecting to improve thermal shock resistant and spot crack for the body.

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