

PRODUCTION OF CORDIERITE-MULLITE CERAMIC COMPOSITE FROM OLIVINE

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Abstract

In this study, reaction sintered cordierite-mullite production from olivin, halloysite and alumina raw materials was presented. Experimental studies essentially consist of three steps. In the first step mixture containing 23,86 w.% olivine, 71,60 w.% halloysite and 4,54 w.% alumina powders was shaped using dry pressing technique. In the second step the shaped samples were sintered at 1200, 1300 and 1400°C for 2 hours. In the third step firing shrinkage, water absorption and three point bending strength tests and XRD analysis were applied to the sintered samples.

Keywords: CORDIERITE, MULLITE, OLIVINE

Introduction

Cordierite ($2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$) is one of the important phases of the $\text{MgO-SiO}_2\text{-Al}_2\text{O}_3$ system. The cordierite ceramics due to their low thermal expansion coefficient, high chemical durability, low dielectric constant, high resistance to thermal shock and high refractoriness are 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 substrates, etc. [1–2]. There are several methods such as solid-state reaction, sol-gel and crystallization of glass to synthesize cordierite phase. Among these methods, the conventional methods for the synthesis of cordierite phase which includes the solid-state sintering of individual oxides of magnesium, aluminum and silicon corresponding to the chemical composition of cordierite, crystallizing the glasses or the sintering of the natural raw materials such as talc and china clay with low silicon content [1]. Chemical routes such as the sol-gel technique generally yield high purity cordierite. Natural raw materials are often used in industrial production of cordierite ceramics for refractory application. Some of the starting raw materials to form cordierite phase reported in literature include (i) a mixture of magnesium compounds and kaolinite [2], (ii) alkaline-earth-aluminosilicate glass, kaolin, alumina and magnesite [3], (iii) talc, calcined alumina and fly ash [4], (iv) kaolin, talc, silica and alumina [5], (v) talc, kaolinitic clay and gibbsite [6], (vi) kaolin, talc and magnesium oxide [7], (vii) talc, kaolin, silica, sepiolite and feldspar [8], and (viii) kaolin and talc [9]. Cordierite phase has three polymorphic forms: (i) the stable high-temperature disorder form, also known as indialite (α - or hexagonal-cordierite) which is stable below 1450°C, (ii) β -

cordierite, called orthorhombic cordierite stable between 1450 oC and its melting point (1460°C), and (iii) μ -cordierite, called metastable cordierite phase, the low-temperature ordered orthorhombic form [1], which can be prepared only under special conditions.

The combination of inexpensive raw materials and high temperature physical properties makes mullite a candidate materials for many uses. Mullite has properties of low thermal expansion, high melting point, creep resistance and chemical inertness. Because of these properties, mullite has extensively using area from refractory brick to electronic component [10,11].

The main objective of this study, was to produce cordierite-mullite composite phases by conventional sintering processes. Moreover, the influence of the sintering temperatures on the formation of cordierite-mullite phases by using starting materials that contain olivine, halloysite and alumina powders were investigated.

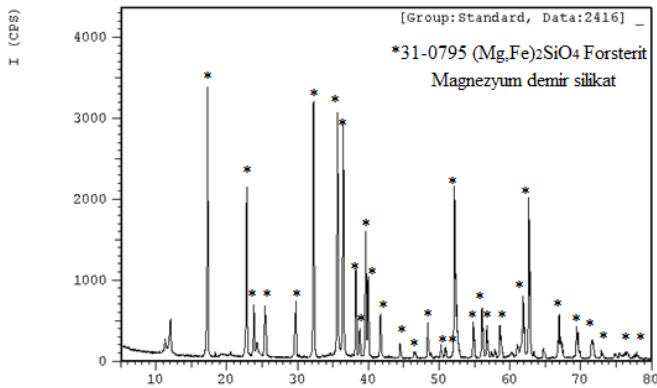
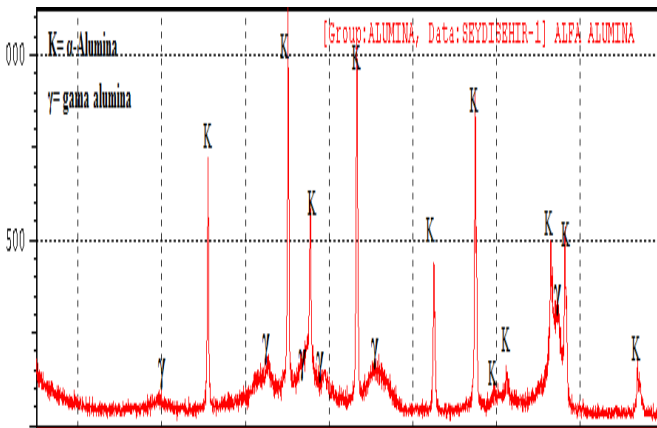
2. Experimental

2.1. Materials

The following raw materials were used in the investigation: Olivine from a deposite located Bursa-Orhaneli areas (Turkey), It is a raw olivine ore, crushed and screened. It contains forsterite and fayalite minerals Chemical and mineralogical analysis of the raw olivine ore are presented in Table 1 and Fig. 1. Alumina powder (supplied by Etibank Aluminum Factory Seydişehir, Turkey), minus 75 μm , 97.59 w.% Al_2O_3 and 1.68 w.% L.O.I. (Table 1 and Fig 2). Halloysite ore supplied by ESAN Eczacıbaşı.(Table1).

Table 1. Chemical analysis of the materials.

Chemical Compositon	Olivine	Halloysite	Alumina
SiO ₂	39	48	0.02
Al ₂ O ₃		35,5	97.59
Fe ₂ O ₃		0,70	0.017
FeO	7-8		
TiO ₂		0.10	0.079
CaO		0,10	
MgO	49	0,20	
K ₂ O		0,50	
Na ₂ O		0,30	0.42
L.O.I			1.68

**Figure 1.** XRD Patterns of olivine ore.**Figure 2.** XRD Patterns of Seydişehir alumina.

2.2. Experiments

The composition of the Cordierite phase consists of MgO:Al₂O₃:SiO₂ in weight ratios 13.7:34.9:51.4. Calculated stoichiometric ratios for the formation of cordierite phase were 23,86 w.% olivine, 71,60 w.% halloysite and 4,54 w.%. The stoichiometric cordierite batch was dry mixed in a ball milling unit with alumina balls for 2 hours. Green compacts were formed by using uniaxial press at 100 MPa pressure. The shaped samples were sintered under air atmosphere at 1200, 1300 and 1400°C with a heating rate of 7.5°C min⁻¹ for 2 h. Firing shrinkage, water

absorption, porosity and three point bending strength tests were applied to the samples. The phase composition of the sintered samples was determined by standard powder X-ray diffractometer (Schmadzu XRD 6000, Japan).

3. Results and Discussions

3.1. Physical and Mechanical Test Results

Physical and mechanical results of the samples are given in Table 2.

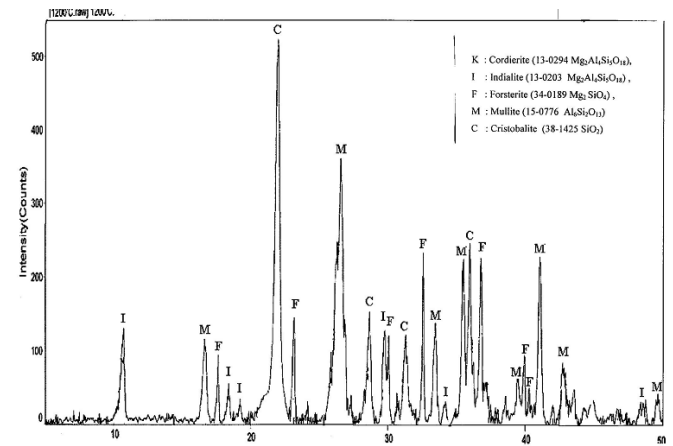
Table 2. Physical and mechanical test results of the samples

Sintering temperature	1200°C	1300°C	1400°C
Water Absorption (%)	8.61	7.78	4.06
Apparent Porosity (%)	19.69	16.27	7.98
Bulk Density (gr/cm ³)	2.29	2.09	1.97
Apparent Density (gr/cm ³)	2.85	2.50	2.09
MOR (kg/cm ²)	86.61	88.10	103.76

Water absorption, apperent porosity, bulk density and apperent density values decreased with the increasing sintering temperature. Closed porosity and three point bending strength (MOR) values increased with the increasing sintering temperature.

3.2. Phase Analyses

XRD phase analysis results of samples sintered at 1200, 1300 and 1400°C were presented at fig 3-5.

**Figure 3.** XRD patterns of the sample sintered at 1200°C

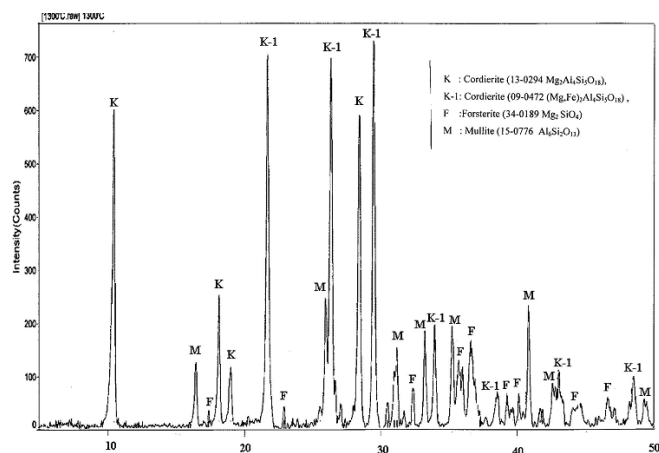


Figure 4. XRD patterns of the sample sintered at 1300°C

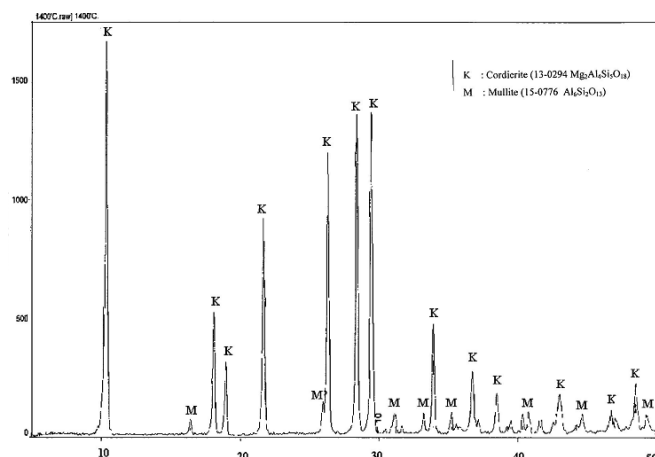


Figure 5. XRD patterns of the sample sintered at 1400°C

The X-ray diffraction patterns of samples sintered at 1200°C indicated that the presence of cordierite (13-0294 $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$), indialite (13-0203 $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$), forsterite (34-0189 Mg_2SiO_4), mullite (15-0776 $\text{Al}_6\text{Si}_2\text{O}_{13}$) and cristobalite (38-1425 SiO_2) phases as shown in Fig. 3.

The X-ray diffraction patterns of samples sintered at 1300°C indicated that the presence of cordierite (13-0294 $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$), cordierite (09-0472 $(\text{Mg,Fe})_2\text{Al}_4\text{Si}_5\text{O}_{18}$), forsterite (34-0189 Mg_2SiO_4), and mullite (15-0776 $\text{Al}_6\text{Si}_2\text{O}_{13}$) phases as shown in Fig. 4. Most of the indialite, forsterite, and cristobalite phases transformed to cordierite phase which are sintered at 1300°C.

The X-ray diffraction patterns of samples sintered at 1400°C indicated that the presence of cordierite (13-0294 $\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$) and mullite (15-0776 $\text{Al}_6\text{Si}_2\text{O}_{13}$) phases as shown in Fig. 5. Intensities of the peaks of cordierite phase increased for the samples sintered at 1300 and 1400°C.

XRD patterns of the samples sintered indicate that the ratio of transformation of cordierite phase increases with the increased sintering temperature. All of the characteristic peaks of cordierite phase are observed for the samples sintered at 1200, 1300 and 1400°C by transformation of secondary crystalline phases such as forsterite, cristobalite phases. Mullite phase intensity decreased at 1400°C.

4. Conclusions

Cordierite and mullite phases in powder form were successfully synthesized by using raw materials including olivine, halloysite and alumina. Cordierite phase transformation was fully completed at 1400°C and mostly achieved with minor mullite phase.

Water absorption, apparent porosity, bulk density and apparent density values decreased with the increasing sintering temperature. Closed porosity and three point bending strength values increased with the increasing sintering temperature.

5. Literature

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