SYNTHESIS AND CHARACTERISATION of CORDIERITE ($\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$) CERAMICS BASED on THE RICE HUSK SILICA

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ABSTRACT

Cordierite ($\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$) ceramics was synthesized and characterised by utilising silica extracted from rice husk. The preparation of cordierite ceramics were conducted by mixing magnesium and aluminium nitrate hydrate with sol silica extracted from rice husk. The samples were sintered at 1300, 1400 and 1500°C. DTA and XRD studies confirmed that cordierite was formed through the intermediate phases of crystoballite, corundum ($\alpha\text{Al}_2\text{O}_3$), spinel ($\text{MgAl}_2\text{O}_4$), and periclase (MgO), respectively. The presence of irregular morphology of a tiny crystallise with wide grain size distribution on the surface is clearly revealed by scanning electron microscopy (SEM) image. DTA results performed that thermal stability of cordierite phase increases with increasing in temperature of sintering.

Keywords : Cordierite, rice husk, sintering, structure, thermal

INTRODUCTION

Cordierite ($\text{Mg}_2\text{Al}_4\text{Si}_5\text{O}_{18}$) is a promising candidate material which has a wide range of uses and application as substrates in the microelectronic application because of their low dielectric permittivity ($\varepsilon$) = 4 at 1 MHz. In addition, decreasing cordierite sintering temperature is also essential for circuit material, since higher temperatures would cause the circuit materials to be oxidised. Therefore, various techniques, such as slip casting, chemical precipitation, sol-gel, solid-reaction, and melting have been investigated in order to decrease sintering temperature of cordierite. The sol-gel process has enabled the production ceramics material of high purity, homogeneous and with superior properties at lower temperature than the others techniques. The processing of low temperature sintering of cordierite ceramics has attracted attention due to the cost reduction and high homogeneous.

Considerable effort has been attracted to the study of cordierite as component of refractory material, high thermal and chemical stability. Cordierite also has a general use in thermal application because of its low thermal expansion, e. i substrate of catalyst for exhaust gas emissions control in automobile, supports heat exchangers for gas turbine enggine and furnace. In addition, cordierite is also used as an integrated circuit substrate because of low dielectric constant i.e, approximately, $\varepsilon$ = 5-6. Many researchers have investigated that rice husk are an excellent source of high-grade ampohous form silica and fine particle size, rice husk has become an important competitive material for preparation of number silica ceramics such as cordierite ceramics, solar grade silicon, silica carbide, silica nitride, magnesium-silica, lithium-aluminium-silica, zeolite, cements surfactant.

In the present study, an effort has been made to find the value addition to rice husk in synthesizing advanced material like cordierite. The cordierite ceramics was synthesized starting from rice husk...
and other water-based precursor materials following a simple step wet-chemical process. The cordierite obtained from rice husk as source of silica were characterised by crystalisation (XRD), microstructural (SEM) and thermal analysis (DTA/TGA).

**EXPERIMENTAL METHOD**

*Figure 1* shows the flow diagram for the synthesis of cordierite ceramics from rice husk using sol-gel method. For the preparation of sols with cordierite composition, i.e, \((\text{Mg(NO}_3\text{)}_2)_{2.6.12}\text{H}_2\text{O}:\text{Al(NO}_3\text{)}_3\text{.9.15 H}_2\text{O}:\text{SiO}_2\) sol from rice husk (2:2:5) \(^{26}\), two steps were conducted, (i) preparation of bi-component sol of magnesia-alumina with their stochiometric compositions, and (ii) preparation of three-component sol magnesia-alumina-silica with their stochiometric composition. In the first step, the precursor material for preparation of magnesia-alumina (2:2 mole ratio) sol were hydrated aluminium nitrate \((\text{Al(NO}_3\text{)}_3\text{.9.15 H}_2\text{O})\) and hydrated magnesium nitrate \((\text{Mg(NO}_3\text{)}_2)_{2.6.12}\text{H}_2\text{O}\) were dissolved in measured volume of deionised water to make a solution of 1 M and pH of 3 by addition of concentrated ammonia solution under stirring. The cordierite sol was continuously heated at 90° C under stirring to obtain the gel powder. The gels were ground to make 600 mesh powders pressed in a metal die at 2 ton to yield cylindrical pellet. These samples were sintered at 1300, 1400 and 1500° C.

![Flow Diagram](image)

*Figure 1. A flow diagram for the synthesis cordierite from rice husk silica*

X-ray diffraction (XRD) patterns for all samples were conducted with an automated Shimadzu using CuKα produced at 30 kV and 30 mA over the 2θ range 5°-100°. Microstructural analysis was conducted with scanning electron microscopy (SEM) Philips-XL on polished and thermally-etched samples. The sintered samples were cold-mounted into an epoxy resin. Differential Thermal
analysis (DTA) measurements on the samples were carried out on a netzsch instrument. Specimens weighing 5 mg were put in platinum crucible and heated to the temperature of 1400°C.

RESULTS AND DISCUSSIONS

X-ray Diffraction (XRD) Analysis. Figure 2. shows the x-ray diffraction (XRD) pattern for the cordierite sample without sintering. Qualitative XRD was conducted by comparing the diffraction lines with the standard PCPDF files using search-match method. A search-match method using PDF file numbers are crystoballite (PDF-39-1425) with 2θ = 21.9º, corundum (PDF-42-1268) with 2θ = 35.5º, cordierite (PDF-13-0293) with 2θ = 10.3º, spinel (21-1152) with 2θ = 36.8, and periclase (45-0946) with 2θ = 43.4º (PCPDF, 1997). The phases of crystoballite and corundum were the major phases observed for the sample without sintering (Figure 2). This result can be attributed to the incomplete reaction of the raw material to form cordierite under the test condition without sintering, which agrees well with the results obtained by

Figure 3. shows the x-ray diffraction pattern for study the formation of the sample for sintered MgO: Al$_2$O$_3$:SiO$_2$ in ratio 2:2:5 at various temperatures. The experiment was conducted by sintering the raw materials at various temperatures of 1300°C, 1400°C and 1500°C for 6 hours. Various phases were found for this experiment. The conventional solid state reaction process for formation of cordierite compound was based on the following reaction:

\[ \text{SiO}_4 + \text{AlO}_4 \rightarrow \text{Si(Al)}_3\text{(OSi)} \]

(1)

\[ \text{Si(OAl)}_3\text{(OSi)} + \text{MgO} \rightarrow \mu\text{-cordierite} \]

(2)

\[ \text{AlO}_6 + \text{MgO} \rightarrow \text{MgAl}_2\text{O}_4 \]

(3)

\[ \mu\text{-cordierite} + \text{MgAl}_2\text{O}_4 \rightarrow \alpha\text{-cordierite} \]

(4)

However, the reaction processes were usually more complex. Sintering at 1300°C for 6 hours (Figure 3) shows that the phases observed were cordierite (Mg$_2$Al$_4$Si$_5$O$_{18}$), crystoballite (SiO$_2$), corundum (α-Al$_2$O$_3$), and spinel (MgAl$_2$O$_4$). The peak intensities for crystobalite, corundum and spinel were decreased with increasing the sintering temperature, but crystalloballite disappeared by 1400°C and 1500°C, followed by appearance of periclase at 1400°C. The significant result observed for the phase of cordierite was increased with increasing the sintering temperature. The formation of cordierite is believed to occur through exothermic reaction according to the reaction 1, 2, 3, and 4 above.

Figure 2. X-ray diffraction pattern of cordierite without sintering using CuKα radiation. The peaks are labeled K for crystobalite (SiO$_2$) and D for corundum (α-Al$_2$O$_3$).
Figure 3. X-ray diffraction patterns of the sintered cordierite at a) 1300°C, b) 1400°C and c) 1500°C using CuKα radiation. The peaks are labeled C for cordierite (Mg₂Al₄Si₅O₁₈), K for cristobalite (SiO₂), D for corundum (α-Al₂O₃), S for spinel (MgAl₂O₄), and P for periclase (MgO).

Microstructural Analysis. The scanning electron microscopy micrographs for the sample cordierite are produced in Figure 4. Microstructure results confirmed the present of irregular and agglomerated morphology of the solid and compact particles, due to the fine particle size. Also, there appeared to wide size distribution of particles on the surface of the samples.

Figure 4. The scanning electron microscopy (SEM) images of cordierite samples, (a) without sintering, (b) 1300°C, (c) 1400°C, (d) 1500°C.
Differential Thermal Analysis. DTA analyses were also performed on the same samples. The results of DTA measurement for the sample without sintering and sintering at 1300ºC, 1400ºC and 1500ºC are shown in Figure 5 (a), (b) (c) and (d). Three broad endothermic peaks at approximately 165.43ºC, 501.66, 697.55 and the other small peak at 337.34 were detected on the sample without sintering (Figure 5 a). Two peaks at 97 and 186 are found on the samples sintering of 1300ºC and 1400ºC (Figure 5 b and c), but not at 1500ºC (Figure 5 d).

The first endothermic peak temperature i.e 97 and 165.43 (Figure 5 a, b and c) can be attributed to the removal of absorbed water and volatiles present as Na-OH, and C-O on the sample, which agree well with the results obtained by\(^{37,6}\). The second endothermic peak at 337.34, 501.66, and 697.55 (Figure 5 a) corresponding to the presence of Si-OH (silanol)\(^ {38}\), Si-O-Si\(^ {39}\), and Mg-O-Al-Si\(^ 5\), due to the structural hydroxyl groups. All of the endothermic peak intensities decreased on the further sintering after1500ºC, there is no peak on the DTA curve, suggesting a completely dehydrated sample. It clears that there occurred the crystallisation process to form cordierite as shown in the XRD results.

Figure 5 Thermal analysis for the cordierite samples (a) without sintering, (b) 1300ºC, (c) 1400ºC, (d) 1500ºC.

CONCLUSIONS

The synthesis and characterisation of cordierite ceramics based on the rice husk silica have been successfully demonstrated. XRD result on the sample without sintering shows the presence of cristoballite (SiO\(_2\)), and corundum (\(\alpha\)Al\(_2\)O\(_3\)). It is suggesting that raw material was not reacting to form cordierite on the sample without sintering. Cystallisation of cordierite phase starts to form at temperature sintering of 1300ºC, 1400ºC and 1500ºC. The formation of cordierite phase forms through intermediate phases of cristoballite (SiO\(_2\)), spinel (MgAl\(_2\)O\(_4\)) and corundum (\(\alpha\)Al\(_2\)O\(_3\)). Phases of cordierite and (\(\alpha\)Al\(_2\)O\(_3\)) were found for all of sample sintering, but periclase (MgO) phase was formed on the sample sintering temperature at 1400ºC. Irregular morphology of the solid compact phases with wide grain size distribution on the surface is clearly revealed by scanning electron microscopy (SEM) image. As expected, thermal stability of cordierite phase increases with increasing in temperature of sintering.

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