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Preparation of refractory cordierite using amorphous rice husk silica for thermal insulation purposes

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ABSTRACT

This study aims to investigate the effect of sintering temperatures on the phase formation and physical characteristics of refractory cordierite prepared from rice husk silica, Al_2O_3, and MgO powders. The samples were subjected to sintering temperatures of 1050–1350 °C, and development of structures was characterized using Fourier Transform Infrared (FTIR) spectroscopy, X-ray diffraction (XRD) coupled with Rietveld analysis, scanning electron microscopy (SEM) and dilatometry. The results obtained indicated the significant role of sintering temperatures on phase transformation of spinel and cristobalite into cordierite, in which at sintering temperatures of 1230–1350 °C the cordierite emerges as a dominant phase, while spinel and cristobalite are practically undetected. Formation of cordierite was followed by decrease in density, porosity, and thermal expansion coefficient, while for hardness and bending strength the opposite was true. Thermal expansion coefficient of the sintered sample at 1350 °C is 3.3×10^{-6}/°C and the XRD analysis demonstrated that the main crystalline phase is cordierite. Based on these characteristics, the samples are considered as insulator, suggesting their potential use in refractory devices.

1. Introduction

Cordierite ($Mg_2Al_4Si_5O_{18}$) ceramic is well known as an excellent insulator and high-thermal resistant material, due to its low dielectric constant as well as thermal expansion coefficient. In previous study [1,2], it was reported that thermal expansion of cordierite is 2.2×10^{-6}/°C, while the others reported the value of around $1–4\times10^{-6}$/°C [3] and $0.8–2\times10^{-6}$/°C [4,5]. Another interesting property of cordierite which makes this material a great importance as refractory material is its very high melting temperature (1460 °C), which is the highest among silicate glass–ceramics [6]. In addition, cordierite exhibits excellent thermal shock resistance [7,8] and high chemical stability [3]. Cordierite can be found in three polymorphic forms, depends on temperature, *i.e.* α-cordierite at high temperature, while β-cordierite and μ-cordierite at low temperature [9,10]. This polymorphism characteristic means that sintering of the substrate at high temperature will lead to conversion of μ-cordierite and β-cordierite into α-cordierite, as the most stable phase. With such properties, cordierite ceramic is considered as a very promising structural materials, suitable for various applications such as catalyst carriers for exhaust gas purification, heat exchanger for gas turbine engines [7,11], refractory for furnaces, electrical and thermal insulation, filter, membranes and heating elements [12,13].

Recognizing the important roles of cordierite in various industrial areas, production of cordierite has been continuously explored, and in general, it is found that the formation of cordierite phase is strongly dependent on the chemical composition, the types of raw material, the presence of impurities, and the preparation methods applied. Many attempts have been devoted to prepare this material from different raw materials using different methods. In previous study [14], solid-state reaction was applied to synthesize cordierite from Al_2O_3, MgO, and SiO_2. In another study [15], cordierite was synthesized from serpertine, kaolinite, and alumina as raw materials, and found that the formation of cordierite was achieved at sintering temperature of 1350 °C. The same formation temperature was reported by others [16] using the

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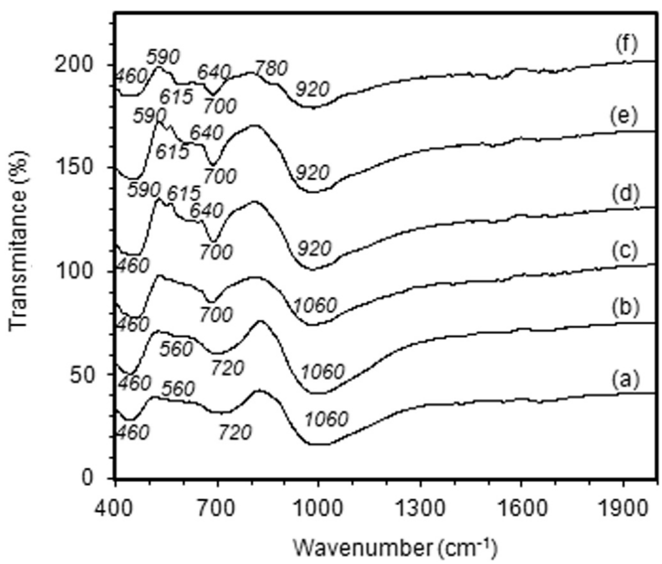

Fig. 1. FTIR spectra of sintered samples at different temperatures (a) 1050 °C, (b) 1110 °C, (c) 1170 °C, (d) 1230 °C, (e) 1290 °C and (f) 1350 °C.

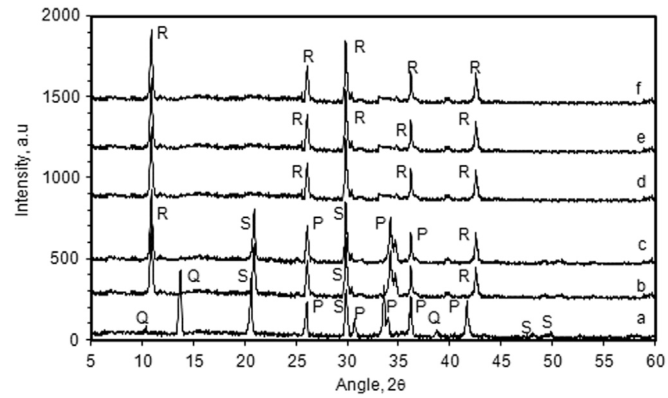

Fig. 2. The x-ray diffraction patterns of the sintered samples at different temperatures (a) 1050 °C, (b) 1110 °C, (c) 1170 °C, (d) 1230 °C, (e) 1290 °C and (f) 1350 °C. P: spinel, Q: μ-cordierite, R: α-cordierite, S: cristobalite.

same method but different raw materials, including andalusite and stevensite. Previous study also showed that α-cordierite, $MgAl_2O_4$/spinel and cristobalite formed at 1300 and at 1350 °C, while at 1375 °C the only phase observed was α-cordierite [9]. Several other studies have reported the synthesis of cordierite from a variety of raw materials such as alumina, kaolinite and talcum [17], fumed silica, bauxite, and talcum [18], talcum, kaolinite, fieldspar, and sepiolite [1, 19], and stevensite-rich clay and andalusite using oil shale as a natural pore-forming agent [20].

Beside solid-state reaction, another method that has been extensively applied is sol–gel method. In previous study [21], this method has been applied to prepare cordierite using silicon alkoxide, chelated aluminum sec-butoxide, and magnesium acetate as starting precursors, and found that the initiation of the μ→α cordierite transformation took place in the temperature range of 1000–1100 °C and α-cordierite was produced at 1200 °C. The study using a mixture of aluminum isopropoxide, magnesium ethoxide, and tetraethylorthosilicate in absolute ethanol revealed that μ-cordierite crystallized at temperature range of 950–1000 °C accompanied by the formation of spinel in small amount, and transformation of μ into α-cordierite started at about 1100 °C [22]. In another study [23] synthesis of cordierite from aluminum chloride, magnesium chloride, and tetraethylorthosilicate was reported. The results obtained indicate that crystallization of μ-cordierite occurred at temperatures between 900 and 1000 °C, and transformed into α-cordierite at temperatures in the range of 1400–1450 °C. Slightly different result was reported by others [24], who used silicic acid, magnesium and aluminum salts, as raw materials, in which it was found that μ-cordierite crystallization occurred at 900 °C, followed by the formation α-cordierite at temperature of 1200 °C and complete transformation of μ into α-cordierite at 1350 °C.

In general, the characteristics of refractory cordierite for thermal insulator are influenced by complex relationship between microstructure and crystalline phases having different thermal expansions when subjected to high temperature and under thermal shock condition. In overall, refractory material should exhibit high thermal shock resistance, high fracture toughness, and low thermal expansion. For these reasons, thermal resistance parameters and thermal shock behaviors of refractory cordierite have been the subject of extensive studies. Previous researchers [7,25] have attempted to synthesize cordierite with excellent thermal shock resistance by subjecting the raw materials into rapid and severe changes in temperature. They found that microstructure and sintering temperature strongly influenced the fracture toughness and densification of cordierite. Considerable effort has also been devoted to study the use of cordierite as the component of refractory material for high thermal applications [5,26], and found that the fracture toughness increases with increasing sintering temperature from 1250 to 1300 °C. Suzuki et al. 1992 [27] have successfully prepared cordierite with high purity and homogeneity, after the sample was sintered at 1300 °C

Related to raw materials for preparation of ceramics, rice husk is a very attractive source of silica, primarily since this agriculture residue is abundantly available, renewable, high silica content, and simple extraction of the silica from the husk. In our previous investigations, active silica from rice husk was obtained by simple acid leaching, and the silica has been used to produce several ceramic materials include borosilicate [28], cordierite [29], carbosil [30], aluminosilicate [31], and mullite [32,33]. The potential of rice husk as an excellent source of high-grade amorphous silica has also been investigated in many other studies [34–36]. Furthermore, this silica has been utilized for preparation of various valuable materials such as solar grade silicon [37], silica carbide [38], magnesium–alumina–silica [39], and lithium–aluminum–silica [40].

To take advantage of the its availability and excellent properties, this present study is aimed to evaluate the potential of rice husk silica as an alternative to commonly used silica for production of cordierite as refractory material for thermal insulation purposes using solid-state reaction. The precursors produced then subjected to thermal treatment in order to investigate the phase development and physical properties. To gain insight on several basic characteristics, the samples were characterized using various techniques include FTIR spectroscopy for functionality analysis, XRD technique for structure investigation, and SEM technique for microstructure investigation.

2. Experimental methods

2.1. Materials

Raw husk used as a source of silica was from local rice milling industry in Bandar Lampung Province, Indonesia. Al_2O_3, MgO powders, KOH, HCl, and absolute alcohol (C_2H_5OH) were purchased from Merck (kGaA, Damstadt, Germany).