



Fabrication and characterization of single phase cordierite honeycomb monolith with porous wall from natural raw materials as catalyst support

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Abstract

A low-cost, single phase cordierite honeycomb monolith whose walls are highly porous has been prepared by the extrusion method using local natural raw materials, as kaolinite and magnesite from Saudi Arabia without need of any organic additives. The characterization of the achieved monoliths has focused on the phase evaluations and microstructures development depending on batch compositions and sintering conditions. Thermal expansion coefficients, microstructures and crystalline structures of the obtained samples have been determined by X-ray diffraction, scanning electron microscopy, FT-IR and thermal dilatometry. Crystalline phases have been also determined by Rietveld analysis. The results show that, synthesis condition of single phase cordierite is extremely narrow by means of both chemical composition and sintering temperature. Single phase cordierite could not have been obtained by using stoichiometric composition for any sintering conditions. Also changing of the sintering temperature as low as 5 °C has affected the crystalline phases. Only single phase cordierite has been synthesized by sintering the non-stoichiometric composition at 1320 °C for 3 h where the linear thermal expansion coefficient of the sample is $2.07 \times 10^{-6}/^{\circ}\text{C}$. Also the fabricated monolith has kept its structure after thermal shock of 300 cycles at 450 °C.

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1. Introduction

The atmosphere has seriously been affected by the automobile exhaust gases which includes nitrogen oxide (NO_x), carbon monoxide (CO), some burned and unburned hydrocarbons. The presence of the hazardous gases causes serious environmental problems such as acid rains, photochemical smog, ozone depletion and global warming [1–2]. For about 2 decades, one of the main issues of researchers has been the removal of these harmful pollutants. In case, automobile exhaust combustion has been assumed as an effective method to reduce the emissions of the harmful compounds using a

catalytic converter which converts toxic pollutants into less toxic pollutants by catalyzing a redox reaction [3–6].

Honeycomb monolith type catalytic converters which are more advantageous than powder catalysts by means of (i) formation of thin layer with high active surface area, (ii) less amount of catalyst loaded on the monolith surface, (iii) easy separation and (iv) complete recovery of the catalyst from the reaction mixture and (v) low resistance to gas flow have been widely used in heterogeneous catalysis, especially in gas phase catalytic reactions [7–8]. These types of catalytic converters are obtained by depositing a catalytic layer onto a non-catalytic substrate. All catalytic reactions occur on the catalytic layer and require mechanical performance supplied by the substrate. So, researches in the field generally focus on the effective (catalyst) layer. In literature, there are many of research results published about the fabrication and characterization of catalytic layer as zeolite [9], copper and manganese

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[10], carbon nanotube [11], alumina [12], titanium silicate [13], zirconium dioxide [14], iron oxide [15].

Catalytic converters are the materials working in arduous conditions especially in case of thermal shock. Heat of the exhaust gas rises up to around 700 °C within seconds and when the engine stops, it decreases to room temperature and this cycle runs thousands of times for the life of the catalytic converter. The thermal shock characteristic of the converter is directly related with the substrate. Thus, fabrication of substrate is also as important at least as the effective layer for the life of the converter.

Cordierite ($2\text{MgO} \cdot 2\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$) is a good candidate as catalytic converter substrate depending on its prevailing properties as (i) low thermal expansion coefficient, (ii) low dielectric constant, (iii) high chemical durability and (iv) high refractories. Three forms of cordierite are known to exist: indialite and both beta- and micro-cordierite. Indialite is the only stable form at high temperatures and is the only one achievable or naturally found in ceramic bodies. Beta- and micro-cordierite can only be formed under special conditions.

It is situated in the primary crystallization field of mullite and has a chemical composition (weight%) of $\text{MgO}=13.8$, $\text{Al}_2\text{O}_3=34.8$ and $\text{SiO}_2=51.4$. Thus, after sintering in most extractions, minor phases such as mullite, corundum, spinel and forsterite, have remained along with the cordierite phases [16–19]. Thermal expansion coefficient of mullite ($5.4 \times 10^{-6}/^\circ\text{C}$), corundum ($8.4 \times 10^{-6}/^\circ\text{C}$), spinel ($9.2 \times 10^{-6}/^\circ\text{C}$) and forsterite ($2.8 \times 10^{-6}/^\circ\text{C}$), impurity phases, are higher than the thermal expansion coefficient of the cordierite ($2.0 \times 10^{-6}/^\circ\text{C}$) phase. Depending on the big difference on the thermal expansion leads a decrease in the life of the cordierite monolith.

The purpose of this study is the fabrication and the characterization of single phase cordierite monolith which is the primary requirement of the catalytic converter substrates by means of life time in real applications from local natural, low cost, raw materials that are extensively developed in Saudi Arabia.

2. Materials and methods

The starting raw materials used in this study are magnesite and kaolinite from Saudi Arabia. The chemical compositions of the raw materials are given in Table 1. Five different batches (Table 2) have been designed to achieve single phase cordierite monolith. The chemical composition of batch 3 (C-3) is very close to the stoichiometric composition of natural cordierite, whereas the other batches have excess (C-4 and C-5) or less (C-1 and C-2) magnesite.

The mixtures have been dispersed in de-ionized water and wet-milled with alumina balls of 5 mm \varnothing for 30 min within a castimide-coated planetary ball mill at 300 rpm. The wet slurries have been feed to a filter press to obtain plastic paste. Plasticity of the paste for extrusion has been determined by Pfefferkorn plasticity test method and the pfefferkorn level of all batches has been set as 33 mm of height by controlling the humidity of the paste.

Green monolith bodies have been prepared using piston type extruder by applying 100 bars with an extrusion speed of around 1 cm/s. The shaped samples have been dried at room temperature for 24 h and following at 105 °C for 24 h. Then they have been placed onto an alumina plate and then sintered in air atmosphere up to 1150 to 1350 °C for 3 h at a rate of 5 °C min^{-1} .

The phases of the produced samples have been determined by XRD (Rigaku-MiniFlex-Japan) and FT-IR (Bruker-Alpha) analysis. The XRD analyses have been performed at a speed of 2°/min with 0.01° steps and using $\text{CuK}\alpha$ radiation between 5° and 70°. Additionally, crystalline phases have been also determined by Rietveld analysis. The chemical compositions and particle size distributions of the raw materials have been determined using XRF (ALS Minerals, Canada) and a laser particle size analyzer (SHIMADZU-SALD-301V), respectively. The thermal characteristics of the batches prepared have been determined by differential thermal analysis (DTA; Shimadzu DTG-60H, Japan). The microstructure of the obtained samples has been recorded via scanning electron microscopy (Mini-SEM; Micro Optic). The thermal expansion coefficients have been obtained using a dilatometer (Netzsch-DIL 402PC, Germany).

Thermal shock character of the prepared samples has been determined for 400 cycles (1 heating and cooling process). Hot air with a temperature of 450 °C has been feed inside of the obtained samples with a rate of 2 l/min for 3 min. Then the sample was left for cooling for 5 min.

Table 2
Chemical composition of batches.

Sample No	Raw material (g)		Chemical composition (%)		
	Magnesite	Kaolinite	MgO	Al ₂ O ₃	SiO ₂
C-1	12.84	102.76	11.96	32.02	51.35
C-2	13.84	102.76	12.87	32.01	51.35
C-3	14.84	102.76	13.78	32.00	51.35
C-4	15.84	102.76	14.69	31.99	51.35
C-5	16.84	102.76	15.60	31.98	51.35

Table 1
Chemical composition of raw materials.

Raw Materials	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	Cr ₂ O ₃	TiO ₂	LOI
Kaolinite	49.3	30.87	1.48	1.24	0.26	0.17	0.12	0.02	2.8	13.0
Magnesite	2.12	0.29	0.04	1.62	89.54	0.09	0.11	0.01	0.01	4.47

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