



In situ formation of sintered cordierite–mullite nano–micro composites by utilizing of waste silica fume

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ABSTRACT

This study aims at in situ formation of sintered cordierite–mullite nano–macro composites having high technological properties using waste silica fume, calcined ball clay, calcined alumina, and magnesia as starting materials. The starting materials were mixed in different ratios to obtain different cordierite–mullite composite batches in which the cordierite contents ranged from 50 to 100 wt.%. The batches were uni-axially pressed at 100 MPa and sintered at 1350, 1400 and 1450 °C to select the optimum temperature required for cordierite–mullite nano–macro composites formation. The formed phases were identified by X-ray diffraction (XRD) pattern. The sintering parameters in terms of bulk density (BD) and apparent porosity (AP) were determined. The microstructure of composites has been investigated by scanning electron microscope (SEM). Cold crushing strength (CCS) of the sintered batches was evaluated. The result revealed that the cordierite–mullite nano–macro composites were in-situ formed at 1400 °C. The batch containing 70 wt.% cordierite showed good physical and mechanical properties.

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1. Introduction

Fumed silica is a waste material produced from silicon metal and ferro-silicon alloy industry [1]. It contains 94–97 wt.% of silicon dioxide (SiO₂) and formed during the reduction of pure quartz (SiO₂) by carbon, in the presence of iron, on heating in electric furnaces up to 1750 °C. A portion of silicon monoxide (SiO) gas escapes during this reduction process through the furnace charge with waste gases, which is oxidized in air, cooled and separated as microsilica dust by electrostatic filters or filter-bags. This fume has to be collected to prevent environmental pollution. Owing to the fine particle size, high SiO₂ content and large surface area of the fume, it can be used as a pozzolan to improve the properties of concrete [1,2]. Furthermore, silica fume has been found to improve the compressive strength, bond strength, and abrasion resistance of concrete. Also, it reduces the permeability and thus helps in protecting steel from corrosion [1–6].

In this work, we studied the feasibility of using silica fume as a source of silica in presence of calcined ball clay, MgO and calcined alumina to fabricate cordierite–mullite nano–micro composites.

Cordierite ceramics consists of the 2MgO:2Al₂O₃:5SiO₂ ternary system components and crystallizes in an orthorhombic, pseudo-hexagonal system, its density is 2.53 g/cm³ and melting point

1470 °C [7]. Mullite (3Al₂O₃·2SiO₂) is another important ceramic material which has an orthorhombic crystal lattice, density 3.17 g/cm³ and melting point 1810 °C [8]. The cordierite ceramics are a promising candidate materials in many applications due to its low dielectric constant ($\epsilon = 5-6$), high resistivity ($r > 10^{12} \Omega \text{ cm}$), high chemical durability, high resistance to thermal shock and very low thermal expansion coefficient ($\alpha = 1-2 \times 10^{-6} \text{ C}^{-1}$). Some examples of its 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. [9,10]. However, cordierite has the disadvantage of its inferior mechanical properties and thermal conductivity compared to other ceramics such as alumina, thus it has relatively shorter service life under thermal fatigue and its ability to dissipate heat is relatively low [11]. Moreover, cordierite ceramics are difficult to sinter by solid state process. Liquid phase sintering, by addition of sintering aids, is not used for cordierite ceramics densification because electrical and thermal properties of cordierite are degraded by sintering aids [9,10,12,13]. Mullite is used as structural material due to its excellent mechanical properties even at high temperature and is also considered as suitable material for electronic packages [14,15]. By formulating cordierite–mullite composites, it is possible to improve the mechanical behavior and thermal properties.

The objective of this paper is to determine the best composition (having higher technological properties) of sintered binary mixtures between nano cordierite and mullite obtained by

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utilizing waste silica fume in presence of calcined ball clay, MgO and calcined alumina.

2. Materials and experimental procedures

2.1. Materials

The starting materials used in this work were fumed silica acquired from Egyptian Company for Ferro-Alloys (Edfu, Aswan, Egypt), a commercial calcined alumina (α -Al₂O₃) powder with medium particle size $\leq 10 \mu\text{m}$ from Alexandria Co. for refractories, calcined ball clay (Aswan area, Egypt) and a highly pure magnesia (VWR International Ltd., England). The chemical compositions of the starting materials are given in Table 1.

2.2. Preparation and characterization of nano cordierite–mullite composites

Six batches containing 50, 60, 70, 80, 90, and 100 wt.% of cordierite were prepared and designed as CM1, CM2, CM3, CM4, CM5 and CM6, respectively. Their compositions are represented in Table 2. Each batch was carefully weighed and mixed by ball mill for 2 h. The starting powders were uni-axially pressed at 100 MPa to obtain pellets of 1 inch in diameter and 0.5 inch in thickness. The pellets were sintered in air at temperature ranging between 1350 °C and 1450 °C, with firing rate of 5 °C/min from room temperature up to 1000 °C, and 2.5 °C/min up to peak temperature and holding for 2 h. The sintering parameters in terms of bulk density and apparent porosity of the sintered compacts were determined after each sintering temperature according to ASTM-C20 method. The changes in phase composition of the sintered batches were identified by X-ray using a Philips 1730 diffractometer with Ni filter, Cu K α radiation at a scan speed of 0.5 min⁻¹. Microstructure of the fracture surfaces or chemically etched-surfaces of sintered compacts was examined using scanning electron microscope (Philips XL 30) after coating with gold thin films. Some samples were chemically etched for 0.5 min with hydrofluoric acid, washed with distilled water and dried before scanning. Cold crushing strength (CCS) of the sintered batches was

Table 1
Chemical composition of the starting materials (wt.%).

Oxide	Silica fume	Alumina	Calcined ball clay	Magnesia
SiO ₂	94.53	0.74	50.62	–
Al ₂ O ₃	0.79	98.20	31.96	–
TiO ₂	–	0.23	1.99	–
Fe ₂ O ₃	2.22	0.41	2.96	–
CaO	0.48	0.27	0.32	–
MgO	1.12	0.07	0.14	99.98
K ₂ O	0.02	0.05	0.12	–
Na ₂ O	0.03	0.10	0.09	–
SO ₃	0.10	–	–	–
L.O.I	1.25	–	11.92	–

Table 2
Batch design of the investigated samples CM1–CM6 (wt.%).

Batch no.	Batch compositions				Calculated mineral compositions	
	Calcined ball clay	MgO	SiO ₂	Al ₂ O ₃	Cordierite	Mullite
CM1	37.98	6.84	20.5	35.91	50	50
CM2	45.57	8.20	18.6	28.73	60	40
CM3	53.17	9.57	16.7	21.55	70	30
CM4	60.76	10.94	14.8	14.36	80	20
CM5	68.36	12.30	12.9	7.18	90	10
CM6	75.95	13.67	11	–	100	0

carried out using a hydraulic machine (SEIDNER, Riedlinger, Germany), having a maximum loading capacity of 600 kN.

3. Results and discussion

3.1. Phase composition of the starting materials and sintered composites

X-ray diffraction (XRD) patterns of the fumed silica and calcined alumina are shown in Fig. 1a and b. XRD analysis reveals that silica fume is amorphous because there are no crystalline peaks except a weak and very broad peak near 21° (Fig. 1a). Fig. 1b exhibits the X-ray diffraction patterns of the used calcined alumina. All detectable diffraction peaks are corresponding to those of α -Al₂O₃ (corundum).

To follow up the phase composition changes after sintering, X-ray diffraction patterns (Fig. 2) was conducted for CM1 batch sintered at 1300, 1350 and 1450 °C. From the figure, it is indicated that CM1 batch sintered at 1300 °C composed mainly of cordierite and mullite minerals, in addition to corundum, cristobalite and quartz phases. By increasing the sintering up to 1350 °C, the peak intensity of cristobalite and quartz has been decreased with remarkable increase in the peak intensity of mullite and cordierite. Such decrease in peak intensity of cristobalite and quartz may be attributed to their reaction with MgO and Al₂O₃ forming cordierite and mullite. Katsuhiko et al. [16] proposed that the reaction between MgO and kaolin took place by the formation of amorphous phase with simultaneous diffusion of Mg²⁺ and O²⁻ ions into the decomposed kaolin. Sorrell [17] reported that the dehydrated kaolinite does not react directly with magnesium oxide to form cordierite and the availability of mullite is a key factor in the formation of cordierite where its formation is accompanied by a decrease in mullite content. Similar conclusion was drawn by Ibrahim et al. [14], where the early formed mullite at 1250 °C gave the way to cordierite crystallization which promoted by firing at 1350 °C. Such previous published work [14,16,17]

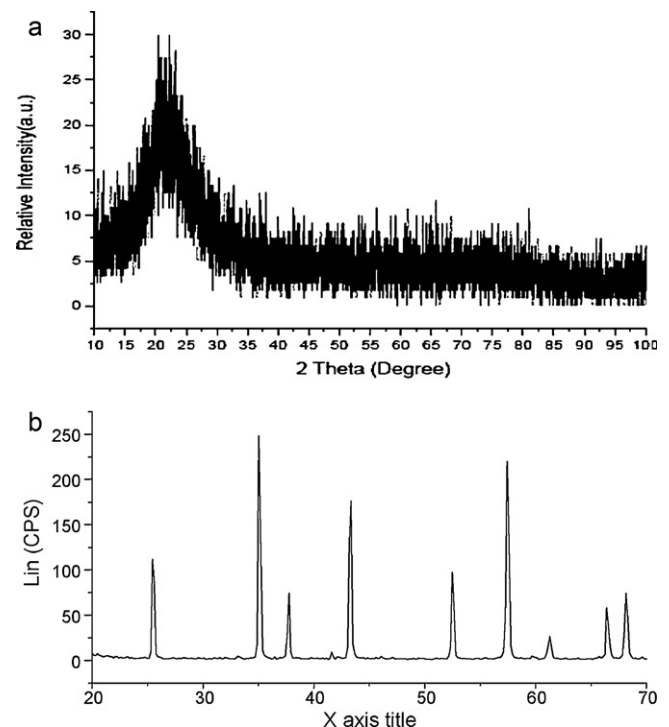


Fig. 1. XRD patterns of silica fume (a) [4] and calcined alumina (b).

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