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Characterization of Cordierite Synthesized from Egyptian Kaolin and Talc

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THE SYNTHESIS of cordierite from Egyptian Kaolin, Talc and Al $(OH)_3$ by solid state reaction at different temperature 1250 - 1350 ^oC has been studied. The samples of the synthesized cordierite obtained from different compositions of raw materials were characterized by X-Ray Diffraction (XRD) and Scanning Electron Microscope (SEM). The XRD analysis showed α-cordierite as a major phase for samples calcined at 1350 ^oC for 1 hr. The morphology of the surface of prepared cordierite appeared to be homogenous, smooth and non pours. Density and porosity of the prepared cordierite were also determined.

Keywords: Solid state reaction, Cordierite, Talc and Kaolin.

Cordierite ceramics have been extensively studied in the last decades, due to their excellent properties, low thermal expansion coefficient, excellent thermal shock resistance, low dielectric constant, high volume resistivity, high chemical durability, high refractoriness and high mechanical strength. Therefore, they are widely used as honeycomb-shaped catalyst carriers in automobile exhaust systems, as substrate material for integrated circuit boards and as refractory materials[(1–6)].

Occurring of natural cordierite is very rare in nature[(7)]. Karkhanavala and Hummel investigated three polymorphs of cordierite as far back as 1953, a stable low-temperature (β) form, a metastable low-temperature (μ) form, and a stable high-temperature (α) form. The high-temperature form can be obtained by solid-state reaction of batch material at 1300^{o}C to 1460^{o}C, or by crystallization of the glass between 1050^{o}C and 1460^{o}C. The metastable low temperature form is not easily developed and requires many hours of crystallization of finely powdered glass at temperatures around 800^{o}C and 900^{o}C. The stable low-temperature form

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is developed only by hydrothermal treatment of glass, the μ form, or the α form at temperatures below 830°C[1].

The most common method to synthesize cordierite is the high temperature reaction in the solid state, although chemical methods, such as co-precipitation reactions, solution combustion or sol- gel technology have been proposed in order to decrease the synthesis temperature and to improve physical properties. Among these methods, sintering of oxide powders through solid state reactions is the most popular.

Some of the starting raw materials reported in literature include (i) a mixture of magnesium compounds and kaolinite[1], (ii)alkaline-earth-aluminosilicate glass, kaolin, alumina and magnesite[2], (iii) talc, calcined alumina and fly ash[3], (iv) kaolin, talc, silica and alumina[4], (v) talc, kaolinitic clay and gibbsite[5], (vi) kaolin, talc and magnesium oxide[6], (vii) talc, kaolin, silica, sepiolite and feldspar[8], and (viii) kaolin and talc[9].

In the present work, solid state methods was employed for the preparation of cordierite, starting with different compositions of kaolin, Talc and alumina.

Experimental

Materials

The raw materials used in this work were Kaolin (Egypt), talc (Egypt) with the composition shown in Table 1, determined by X-ray fluorescence and aluminum hydroxide (Merck).

TABLE 1. The chemical compositions of Kaolin and talc (wt.%), determined by X -ray fluorescence.

Materials	SiO_2	TiO_2	Al_2O_3	Fe_2O_3tot	MnO	MgO	CaO	Na_2O	K_2O	NiO	Cr_2O_4	SO_3	L.O.I
Kaolin	50.81	2.46	31.69	2.13	0.01	0.41	0.74	0.05	0.08	-	-	0.2	11
Talc	58.65	0.02	1.14	5.52	0.04	26.63	1.26	-	-	0.26	0.16	0.08	6.1

The most common mineral in the production of cordierite[10-12] is kaolin ($Al_2O_3.2SiO_2.2H_2O$) that used as a source of SiO_2 and Al_2O_3, also increases the plastic behavior of the batch. Talc ($3MgO.4SiO_2.H_2O$) has also been used as raw material for cordierite synthesis in many studies, it used as the source of MgO and SiO_2[11-13]. The Raw materials were chosen as components in this study because of its low impurity content and they are local product.

Preparation of cordierite

In the literature, based on the SiO_2-MgO-Al_2O_3 diagram[14] some researchers have employed a 1:1:2 mole ratio[11, 12] while some others have used a 2:2:5 mole

ratio $MgO\text{-}Al_2O_3\text{-}\ SiO_2$ for cordierite[10,15] . In this work, the raw materials were mixed in proper amounts to obtain 2:2:5 stoichiometric cordierite mixtures.

Five batch compositions of kaolin and talc were mixed with different amount of aluminum hydroxide to attain 2:2:5 mole ratio of $MgO\text{-}Al_2O_3\text{-}\ SiO_2$ for cordierite composition as listed in Table 2. Then, the raw materials were ground in a ball mill to obtain homogenized mixtures.

TABLE 2. Batches composition (wt. %).

Sample code	Kaolin	Talc	$Al(OH)_3$
C1	37.19	29.75	33.06
C2	36.66	29.33	34.01
C3	36.15	28.91	34.94
C4	35.65	28.52	35.83
C5	35.17	28.13	36.70

The powders were pressed uniaxially in a universal hydraulic press (MEGA KSC-10A) at 120 MPa pressure in a stainless steel die. The prepared specimens were heated at different temperatures of 1250, 1300 and 1350^{o}C with soaking time of 1 hr with constant heating rate of 10^{o}C /min. The kiln was allowed to cool by itself in air.

Techniques

An X-ray diffraction investigation of samples preheated in air at 1250, 1300 and 1350 ^{o}C was conducted using Bruker D8 diffractometer with Cu-kα radiation (λ=1.54) at 40 kV and 40 mA. Scanning electron microscopy (SEM) was taken on (Philips XL30); the samples were first sputtered with a thin layer of gold to avoid charging.

Results and Discussion

X-ray diffraction analyses

The phases formed for different batch compositions C1- C5 after sintering at different temperatures ranging from 1250 ^{o}C to 1350 ^{o}C were identified by XRD and are shown in Fig. 1-5, where the signals are associated to the cordierite substrate identified with α symbol. Diffractograms show the presence of the phases mullite and cristobalite for C1 composition at 1250°C (Fig. 1). When the temperature is raised upto 1300°C the cordierite phase appeared but the intensity of the lines is still low due to low crystallinity indicating that, the solid state reaction is started. At 1350°C a considerable increase in crystallinity is observed, with a large number of patterns corresponding to cordierite.

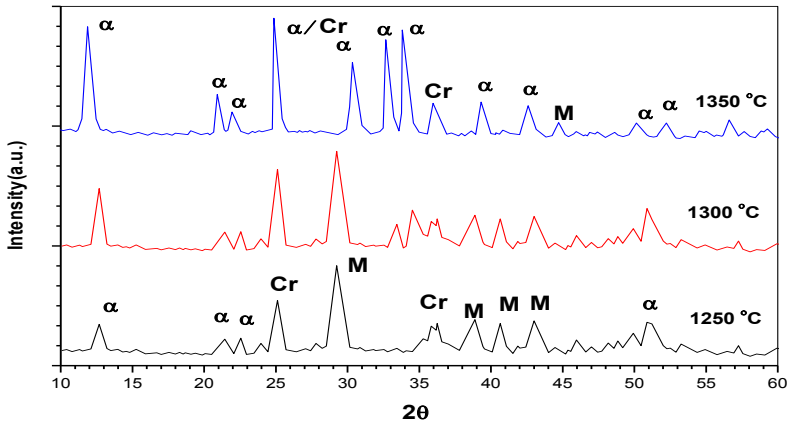

Fig. 1. XRD pattern of C1 composition after calcinations between 1250^{O}C and 1350^{O}C for 1 hr (α = Cordierite, Cr = Cristobalite and M = Mullite).

Diffractograms of C2 composition (Fig. 2) at 1250°C show the appearance of cristobalite and mullite patterns. The increase of temperature up to 1300°C the pattern of cordierite appeared but with low intensity. At 1350°C pattern of cordierite was detected but still with low intensity and the phases of cristobalite and mullite are still existed. X-ray diffraction patterns of C3 composition (Fig. 3), show that sample preheated at 1250 ^{o}C gives cordierite with patterns of low intensities. Further increase of temperature upto 1350^{o}C a large number of patterns related to cordierite was detected, while the lines corresponding to quartz, cristobalite and mullite greatly diminished. From X-ray diffractograms of C4 and C5 compositions (Fig. 4, 5), it can be seen that cristobalite, mullite and cordierite phases appeared at all calcination temperature.

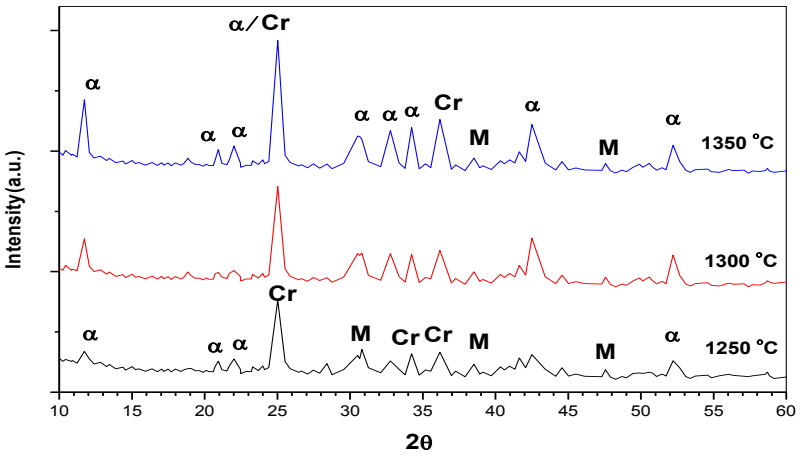

Fig. 2. XRD pattern of C2 composition after calcinations between 1250^{O}C and 1350^{O}C for 1 hr (α = Cordierite, Cr = Cristobalite and M = Mullite).

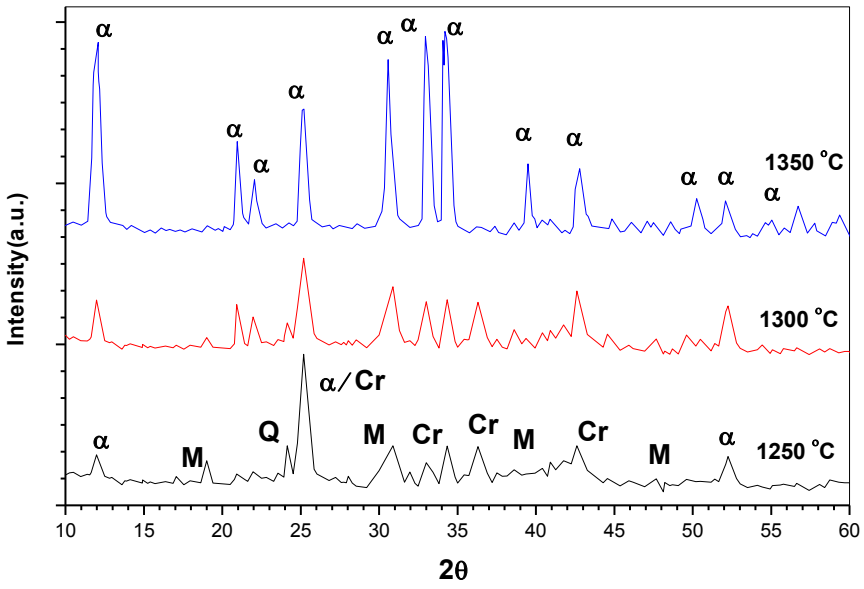

Fig. 3. XRD pattern of C3 composition after calcinations between 1250^{O}C and 1350^{O}C for 1 hr (α = Cordierite, Q = Quartz, Cr = Cristobalite and M = Mullite).

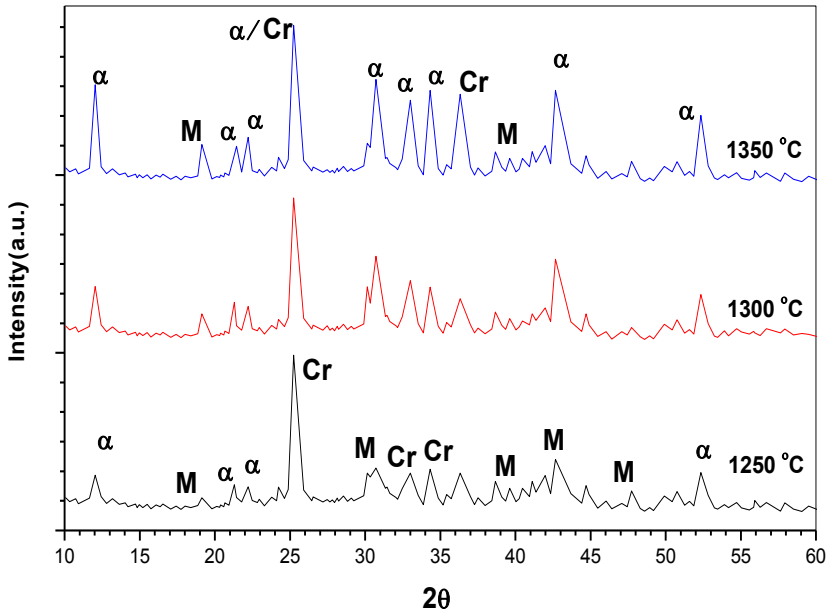

Fig. 4. XRD pattern of C4 composition after calcinations between 1250^{O}C and 1350^{O}C for 1hr (α = Cordierite, Cr = Cristobalite and M = Mullite).

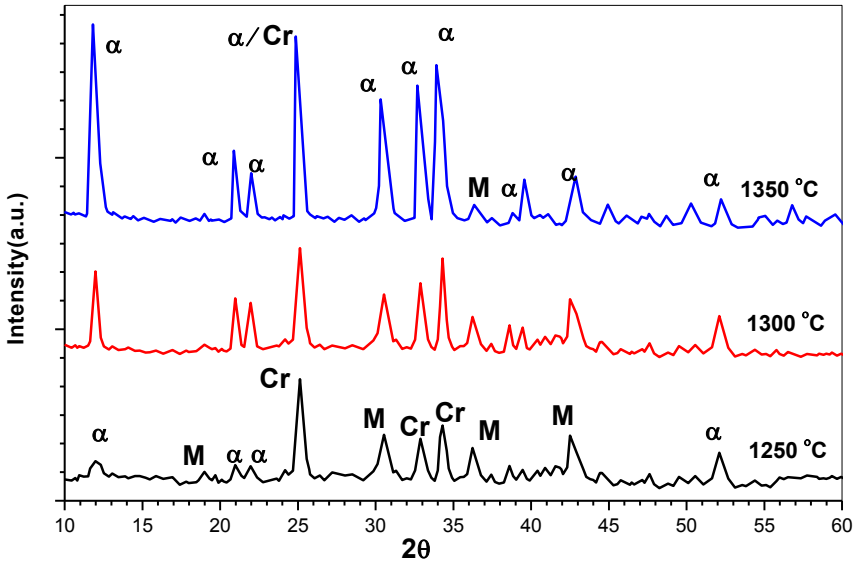

Fig. 5. XRD pattern of C 5 composition after calcinations between 1250°C and 1350°C for 1hr (α = Cordierite, Q = Quartz, Cr = Cristobalite and M = Mullite).

Based on these results, it can be found that, at 1250°C and 1300 °C all compositions produce more than one phase and at 1350 °C the only composition produces pure cordierite was C3, which exhibit well crystalline phase and has an excellent degree of crystallinity. Table 3 summarized the result of X- ray patterns of five compositions.

TABLE 3. Summarized results of x- ray analysis of five compositions preheated at different temperatures.

Sample Code	Phase Formed*		
	1250°C	**1300**°C	**1350**°C
C1	α , Cr , M	α , Cr , M	α , Cr , M
C2	α , Cr , M ,Q	α , Cr , M	α , Cr , M
C3	α , Cr , M ,Q	α , Cr , M ,Q	α
C4	α , Cr , M ,Q	α , Cr , M	α , Cr , M
C5	α , Cr , M ,Q	α , Cr , M	α , Cr , M

* α = Cordierite, Q = Quartz, Cr = Cristobalite and M = Mullite.

Density and porosity evaluation

Cordierite sample C3 was subjected to physical property evaluation as ASTM standards [16]. The density and porosity of the synthesized cordierite mixtures were determined using Archimedes' technique. It gives low apparent porosity values (10.25%), due to that the closed porosity increases with sintering temperature, because some closed pores formed in the liquid phase of low viscosity at higher temperatures. The bulk density was (2.2 g/cm^3)

Scanning electron microscope (SEM)

In order to investigate the morphology of cordierite, SEM analysis was performed. The SEM of samples shown in Fig. 6 corresponds to C3 sample. The surface of cordierite appeared to be smooth, homogenous and non porous due to calcination at high temperature 1350^{o}C, which matches with the porosity measurements using Archimedes' technique.

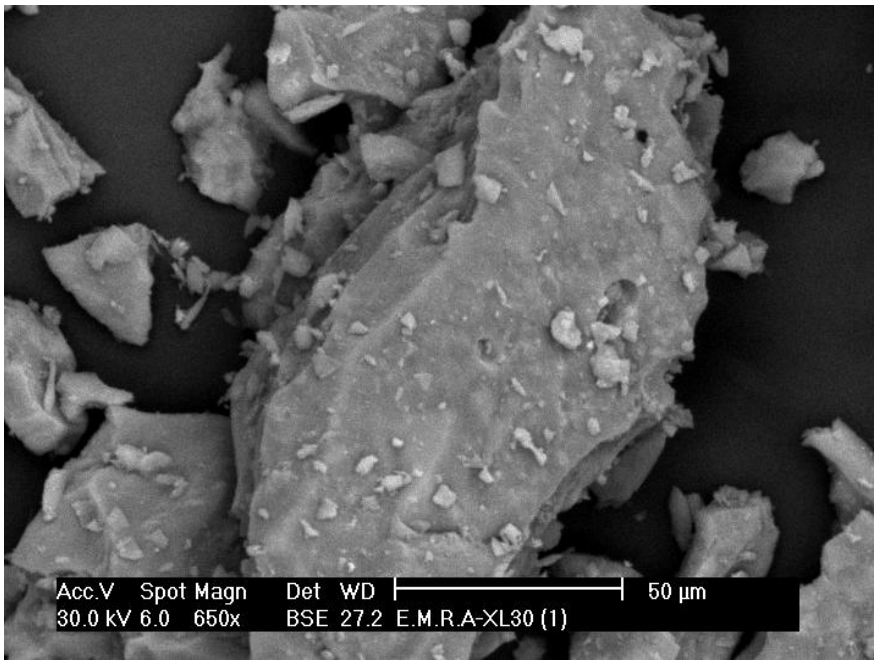

Fig. 6-a. SEM images of cordierite ceramics (Sample C3) sintered at 1350°C for 1 hr soaking time at magnification 650x.

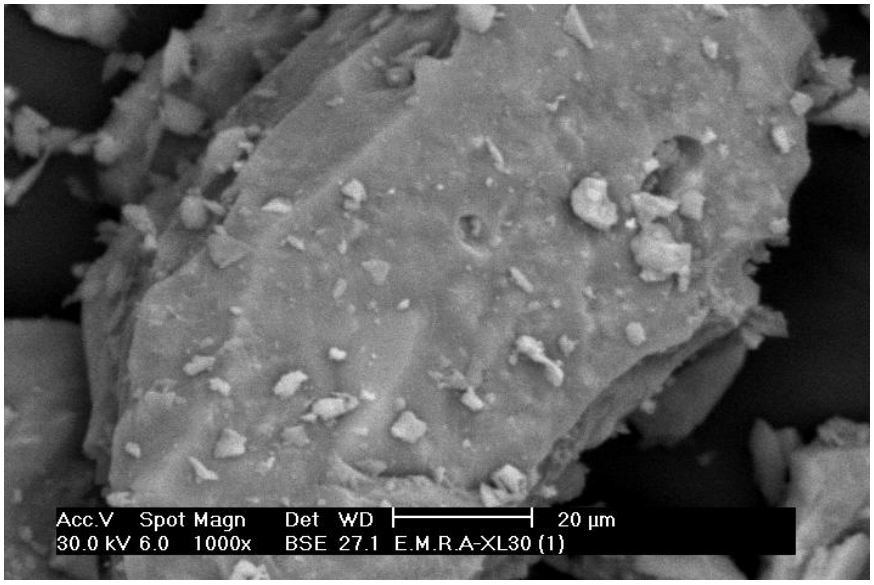

Fig. 6-b. SEM images of cordierite ceramics (Sample C3) sintered at 1350°C for 1 hr soaking time at magnification 1000x.

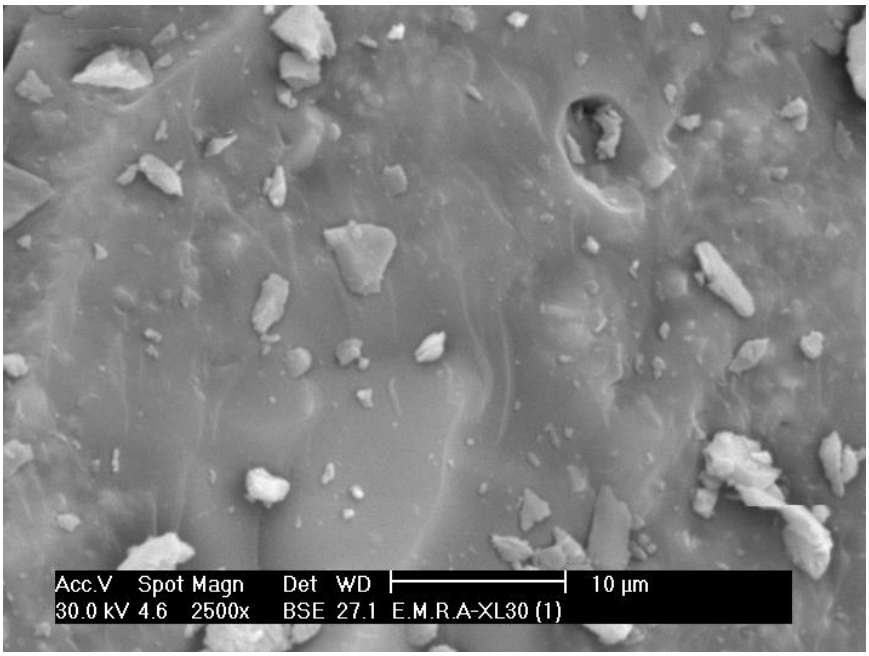

Fig. 6-c. SEM images of cordierite ceramics (Sample C3) sintered at 1350°C for 1 hr soaking time at magnification 2500x.

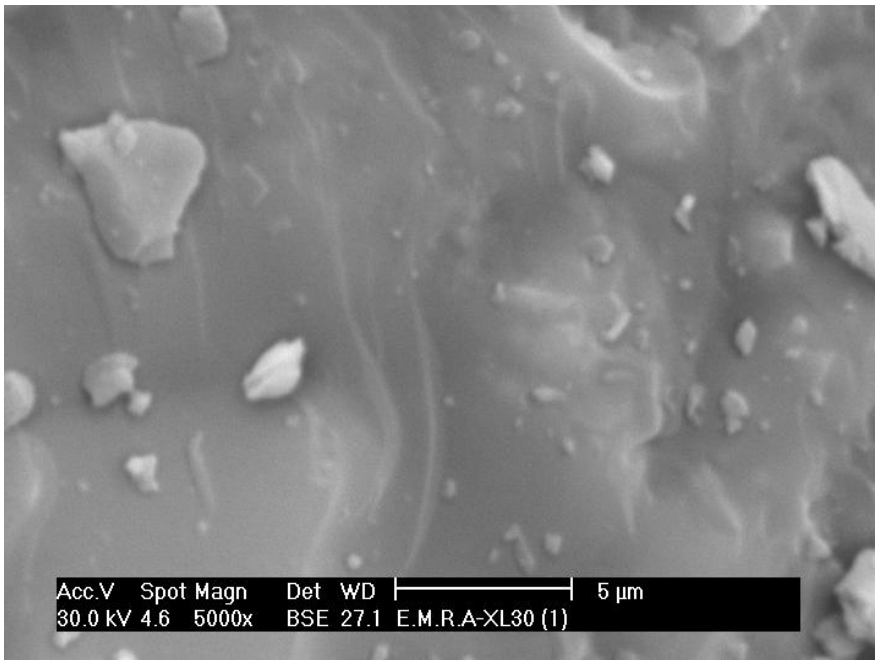

Fig. 6-d. SEM images of cordierite ceramics (Sample C3) sintered at 1350°C for 1 hr soaking time at magnification 5000 x.

Conclusion

In this work, cordierite ceramic was prepared by solid state reaction between Egyptian kaolin, talc and aluminum hydroxide, where different batch composition examined. X-ray diffraction method proved pure cordierite phase in sample C3 preheated at 1350^{o}C for 1 hr soaking time and the surface was homogenous and non porous as shown by SEM.

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توصيف الكورديوريت المحضر من الكاولين المصرى والتلك

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تم دراسة تحضير الكورديوريت من الكاولين المصري و التلك و هيدروكسيد الالمونيوم عن طريق تفاعل الحالة الصلبة في درجات حرارة مختلفة ١٢٥٠ - ١٣٥٠ درجة مئوية . تم توصيف العينات المحضرة المختلفة من الكورديوريت باستخدام تقنية حيود الأشعة السينية (XRD) ، ومجهر المسح الإلكتروني (SEM). وأظهرت نتائج حيود الاشعة السينية وجود الكورديوريت المحضر فى صورة نقية عند درجة حرارة ١٣٥٠ درجة مئوية . وجرى أيضا تحديد الكثافة والمسامية للكورديوريت.