Journal of Ovonic Research Vol. 18, No. 4, July - August 2022, p. 609 - 615

Synthesis and characterization of cordierite and cordierite-zirconia by conventional approach

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A wide recognition for cordierite ceramic due to its superior mechanical and electrical properties made cordierite a candidate for study. A simple approach of synthesis of cordierite and cordierite zirconia (5-15 wt %) was tried at the stoichiometric composition with standard raw material of high purity. The binder, lubricant and flux such as polyvinyl alcohol, ethylene glycol and sodium hydroxide respectively were added with 1 wt%. The powders were mixed, wet milled, dried at 100°C for 12 h, compacted and sintered at different temperatures between 600-1400°C for 3 h. The studies *on* XRD, FTIR and TG/DTA were carried out and the results from the studies confirmed the presence of cordierite phase alone for pure cordierite and cordierite along with zircon for cordierite-zirconia compositions.

(Received April 21, 2022; Accepted August 12, 2022)

Keywords: Cordierite, Zirconia, Sintering, Ethylene glycol, Powder metallurgy

1. Introduction

Rising concern on environmental pollution in IC engines is due to its increasing population has caused a serious impact on imposing very strict emission regulation. This has led to the development of new technologies in engine; modifications in fuel, alternative fuel technology and exhaust treatment applications. The exhaust gases from the vehicles are CO_2, H_2O, Hydrocarbon, NOx and particulate matter pollutants. There have been many studies on the pollutants due to unburnt gases to reduce and oxidize the gases. This has led to the development of after treatment technologies like two-way, three-way catalysts and diesel particulate filters to convert them in CO_2 and H_2O vapor. The catalyst pt, pd and Rh were used for chemical treatment, as the catalysts used were in small quantities, they require a support material. The support materials developed by researchers were silicon carbide, alumina, cordierite etc [1–2]. cordierite remains a prominent material because of its excellent resistance to thermal shock and mechanical properties [3]. The cordierite acceptance as support material is mainly due to its low operating costs; very high durability and reliability, less operating temperatures[4]. Apart from this the cordierite has been in use of many industrial applications such as in refractory, foundries, tools, structural, and biomaterials microelectronic applications, etc [5–6]. This has provided it with a leadership role. The synthesis of cordierite has been carried out through techniques like conventional and non-conventional process. The synthesis of pure cordierite and cordierite efficient solvent with raw materials magnesium nitrate hexa-hydrate, aluminium nitrate nonahydrated, zirconium oxychloride octahydrate and tetra-ethoxysilane [7]. The other paper

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https://doi.org/10.15251/JOR.2022.184.609

reported the use of magnesium aluminium hydroxide, magnesium chloride and sodium silicate samples with high density and small grains through precipitation and yet another research paper studies the colloidal processing technique various zirconia compositions up to 30 wt % using boehmite sol as a source for alumina, silica sol, and magnesium nitrate sold and zirconia sol [8]. The cordierite characterization through conventional method for casting application was prepared from naturally available materials kaolin and talc to from cordierite ceramics and a difference procedure of precipitation method and mechanical mixing from clay based materials, exhibited the presence of mullite phase in mechanical mixing preparation method [9]. Though all the methods for synthesis of cordierite and cordierite –zirconia were found to be successful, it explains the need for controlled composition will result with decrease in sintering temperatures [10–11]. However the practical limitation is the complex fabrication process for non-conventional techniques and uses of non-standard materials have resulted only for specific applications [12]. Even though there happens to be no such single processing methods adopted for fabricating a ceramic component from starting powder, this has resulted in development of variety of synthesis techniques.

This paper reports the synthesis of pure cordierite and cordierite-zirconia composites (5 to 15 wt %) prepared from standard raw materials with high purity using conventional route. The powders were sintered at different sintering temperatures between 800 to 1400°C for 3 h. The evolution of phases was carried out at the powder samples by studies on XRD, TE/DTA and FT-IR.

2. Experimental procedure

The preparation of cordierite through conventional processing technique was carried out considering the cordierite stoichiometric composition ($2MgO.2Al_2O_3.5SiO_2$). The ceramic powders were mixed in the ratio of 14; 35; 51 for the oxides of magnesium, aluminium, silica and zirconia (5-15 wt%). The dried samples powders added with binder flux and lubricant were added with 1 wt% respectively each [13–14]. The samples were compacted to a diameter of 10 mm at 200 MPa. The green body compacts were sintered at temperatures between 800 to 1400°C for 3 hours at a heating rate of 10°C/ min and allowed to cool in the furnace itself. The XRD patterns were obtained with diffractometer using CuKα radiation, FT-IR spectra using KBr medium with spectrometer (BRUKER) and TG/DTA analysis curve from thermal analyzer (NETZSCH) at a heating rate of 10^0C min to characterize the samples [15–16].

3. Results and discussions

The results revealed the importance of the sintering temperature and time is a criterion for synthesis of cordierite and cordierite/ZrO_2 samples. This has an influence on the zirconia's oxidation characteristics making it a promising candidate for the study. Figure 1 shows the XRD patters of the pure cordierite sample (M1) for various sintering temperatures. The patterns at 600 and 800^0C indicate the amorphous nature and above this temperature, formation of µ- cordierite (metastable) and cristobalite up to 1200°C. The solid state reaction synthesis at 1400°C for 3 h indicated the presence of stable α- cordierite, which is evident from the previous research the α-cordierite formation occurs within 1h to 12 h [17]. XRD patterns for various zirconia compositions (5-15 wt%) are shown in figures 1and 2 for various sintering temperatures. The phase transformation characteristics for both the samples at various temperatures and compositions, which exhibited the characteristics for both the samples at various temperatures and compositions, which exhibited the characteristic importance, have been shown in Table 1. The addition of zirconia with varying compositions confirmed the presence of α- cordierite, Zircon and impurity phases of zirconia in small percentage at 1400°C for 3 h. The zirconia reacts with the silica to form zircon, which is present mainly in the grain boundaries [18–19]. The addition of zirconia has a significant importance as reported in the previous papers increase the mechanical properties as well as an interface to oxidize the soot at lower temperatures; this makes it a potential candidate for the field of study in this application [20].

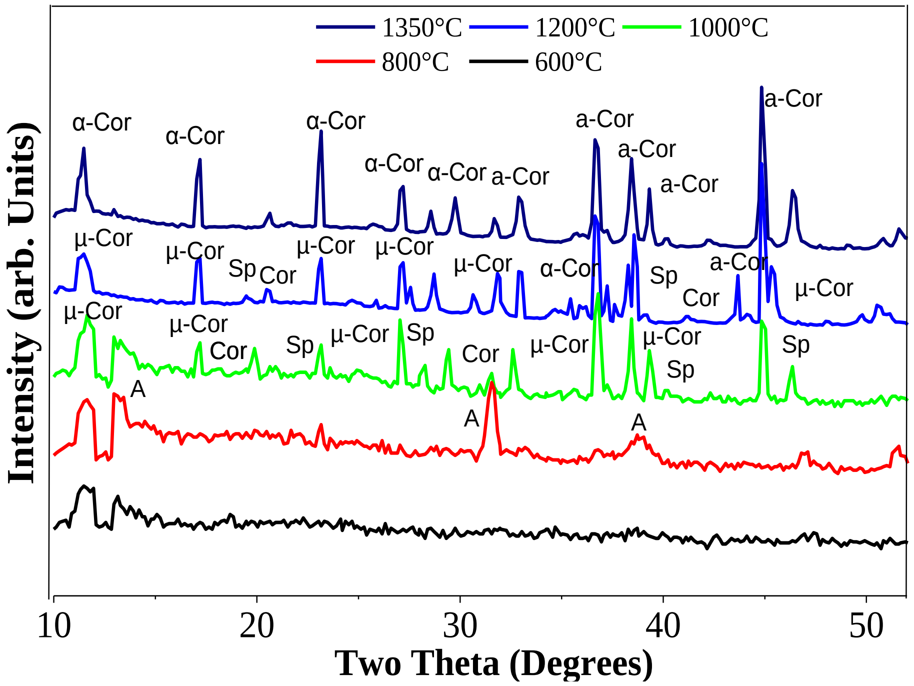

Fig. 1. XRD pattern for pure cordierite.

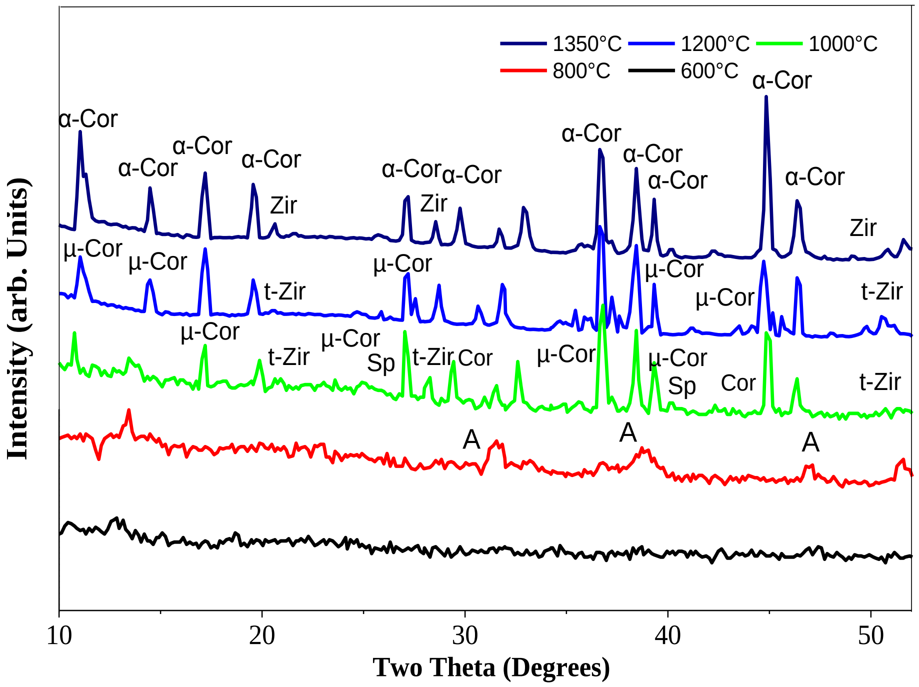

Fig. 2. XRD pattern for cordierite-zirconia (15 wt%).

Table 1. Phase identification of samples various sintering temperatures.

Samples	Phase Identification				
	600°C	**800°C**	**1000°C**	**1200°C**	**1400°C**
Cord	A	A	μ-Cor	μ-Cor	α-Cor
			Sp	α-Cor	
			Cor	Sp	
				Cor	
Cord-Zirconia (5wt %)	A	A	μ-Cor	μ-Cor	α-Cor
			Sp	α-C	Zir
			Cor	Sp	t-Zir
			t-Zir	Cor	
				t-Zir	
Cord-Zirconia (10wt %)	A	A	μ-Cor	μ-Cor	α-Cor
			Sp	α-Cor	Zir
			Cor	Sp	t-Zir
			t-Zir	Cor	
				t-Zir	
Cord-Zirconia (15wt %)	A	A	μ-Cor	μ-Cor	α-Cor
			Sp	α-Cor	t-Zir
			Cor	Sp	
			t-Zir	Cor	
				t-Zir	
				M-Zir	

A-Amorphous; μ-Cor – Cordierite (metastable); Sp- Spinel; C-Cristobalite; Zir- Zircon; t-Zir – Tetragonal zirconia; m-Zir- Monoclinic Zirconia; α-Cor – cordierite (Stable)

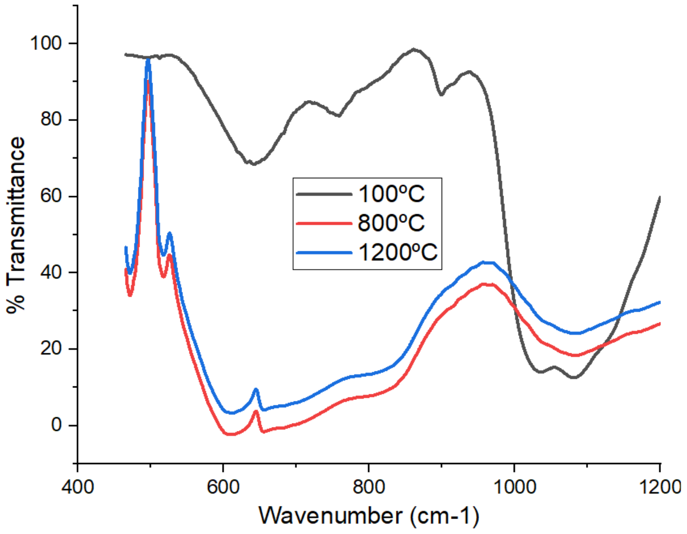

Fig. 3. FTIR spectra for pure cordierite.

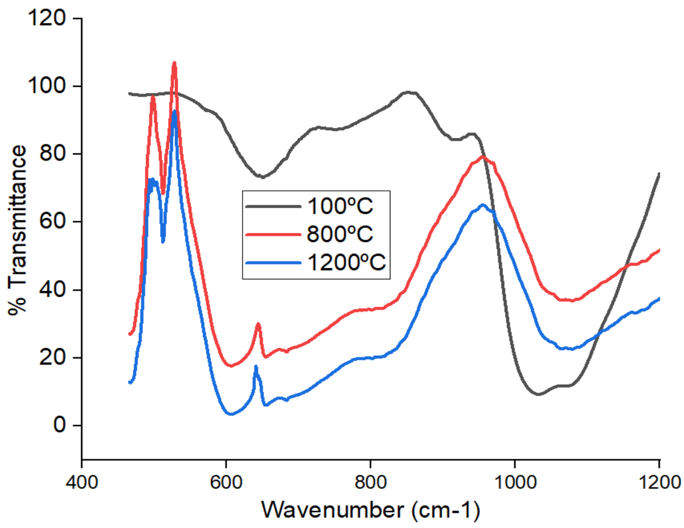

Fig. 4. FTIR spectra for cordierite-zirconia (15 wt%)

The FT-IR spectra given in Figure 5 for pure cordierite powder heat treated at various temperatures. The spectrum curve at 100°C shows the absorption band diminishes with increase in temperature at 845 cm^{-1} SiOH and /or AlOH groups, indicates the condensation rate proceeds during heating. At 1064 cm^{-1}, the band of Si-O-Al shift to a lower wave number. During the green body heating, AlO_6 transforms into AlO_4 and aluminium ions get incorporated in SiO_4 tetrahedral unit to form a Si-O-Al network structure [21]. The typical adsorption silica bonds and μ cordierite for the cordierite powder treated at 1200°C shows peaks at 1100 cm^{-1}, 923 cm^{-1} and 430 cm^{-1}. The bonds stretching motion of a Si atom against its tetrahedral oxygen cage is observed. Bands near 1100 cm^{-1} is due to the symmetric motion of bridged oxygen in the plane bisecting the Si-O-Si bond is observed near 430 cm cm$^{-1.}$

The band at 923 cm^{-1} is due to the vibration modes of non-bridging oxygen’s in alumino-silicate at Mg content. The absorption bands at 760cm^{-1} corresponds to stretching vibrational modes of Al-O and Mg-O bonds. The bands 650 cm^{-1} and 530 cm^{-1} show the presence of spinel peak. The absorption bands at 1100 cm^{-1} and 805cm^{-1} are due to the occurrence of β-quartz and μ-cordierite at high temperature in hexagonal form. The presence of μ and α cordierite is confirmed in the spectrum at 1200^{0}C shows evidence of complete transformation of μ-cordierite to α-cordierite above this sintering temperature. FT-IR spectrum given in Figures 3 and 4 for the cordierite-zirconia samples at different temperatures show the characteristics bands of cordierite, zircon and zirconia at 605 cm^{-1}.

TG \ DTA analysis for the green body samples of cordierite and cordierite-zirconia (15 wt%) shown in Figs.5 and 10 dried at 100°C for 12 h exhibited the following results. The endothermic peak was observed in two regions in both the samples. The first one at 500^{0}C indicates dehydration of solid-state reaction confirms the phase transformation. It shows a broad exothermic in the region beyond 1000°C. The samples revealed a weight loss of 12% due to dehydration at the amorphous phase.

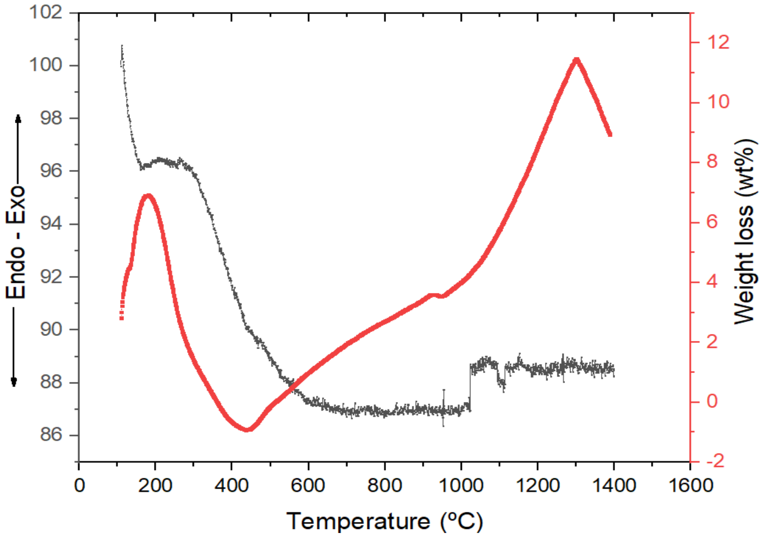

Fig. 5. TG/DTA curve for cordierite-zirconia (15 wt%).

The results on interpretation of FT-IR spectra and TG/TDA curves are found to be in consistent with the XRD results confirming the ceramic as a suitable substrate material.

4. Conclusion

The synthesis of pure cordierite and cordierite-zirconia under controlled composition for standard application as support material on catalytic converters was successfully synthesized. However, the results obtained below showed the sintering time and temperature an important parameter. Cordierite presence was observed at temperature above 1300°C. Cordierite-zirconia (5-15 wt %) samples confirmed the presence of cordierite, zircon along with impurity phases in small amounts. The zirconia addition decreased the optimum sintering temperature at 1400°C.

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