

Influence of particle size distribution of precursor oxides on the synthesis of cordierite by solid-state reaction

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Abstract

The influence of the particle size distribution of the precursor oxides on the synthesis of cordierite by solid-state reaction has been investigated. Expectedly, it is found that reducing the particle size of the raw materials results in a decrease in the temperature at which the synthesis process starts. Likewise, small particle sizes promote the sintering when subjecting the monoliths to high temperatures (1400 °C) for long periods of time (500 min), with almost no destruction of the highly crystalline honeycomb structure, which is mainly comprised of a pseudo-cordierite.

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1. Introduction

Cordierite, $2\text{MgO} \cdot 3\text{Al}_2\text{O}_3 \cdot 5\text{SiO}_2$, is a material having a low thermal expansion coefficient, a low dielectric constant, and a high chemical and mechanical stability. These characteristics make it an interesting candidate for many industrial applications [1]. Taking into account that the natural mineral is not abundant or sufficiently pure, cordierite has to be synthesised. Some typical processes to synthesise cordierite are the sol–gel technique [2], sintering and crystallisation from the vitreous state [3], and the solid-state reaction [4] or consolidation of the structure with starch and subsequent calcination to obtain a porous cordierite with a compatibility with washcoat and catalysts [5].

One of the most significant applications of cordierite is as honeycomb catalytic support. This structure is increasingly used for many reactor applications, such as petrochemical industry [6], selective reduction of nitrogen oxides [7], selective hydrogenation of alcohols [8], automobile emis-

sion control [9] and control of volatile organic compounds [10].

The application of cordierite as monolith honeycomb carrier requires the synthesis by solid-state reaction from a mixture of various precursor oxides. This technique allows conformation of the monolithic structure with the sufficient refractoriness and thermal shock resistance to be used as catalytic support. Nevertheless, the formation of cordierite from its constituent oxides by solid-state reaction presents as a significant disadvantage due to the close calcination temperature range to the different eutectic points [11]. In addition, the polymorphic character of cordierite can complicate the microstructural evolution during the thermal process, which predicts a faster crystallisation for the most stable polymorph [12].

Three different forms of cordierite have been recognised. α -Cordierite is the form usually found in nature and ceramic bodies; the other two forms, namely μ and β cordierite, are less common and can be prepared only under special conditions. Only α -cordierite can be obtained by solid-state reaction from the precursor oxides [13]. The stable forms are that of hexagonal symmetry (α -cordierite or indialite), which is obtained at high temperatures, and that of

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orthorhombic symmetry (β -cordierite or just cordierite), which is obtained at low temperatures [14]. Apart from polymorphism, it is worth noting that commercial cordierites generally exhibit a pseudohexagonal lattice. Hence, analysis performed with synthetic cordierites allows comparison of their characteristics with those of natural minerals, showing intermediate behaviour between orthorhombic and hexagonal symmetries in most cases [15].

Our previous study [16] demonstrated that the distribution of the particle size of the precursor oxide mixture is a key factor not only in the evolution of the solid state reaction, but also in the formation of different polymorphs mainly at high temperatures and long periods of reaction.

The objective of this work was to determine the particle size distributions of the precursor oxides that facilitates synthesis without altering the monolithic structure and favours the synthesis of a crystalline phase with an intermediate symmetry between cordierite and indialite polymorphs.

2. Experimental

The raw materials used were kaolin, talc, and silica provided by Lorda and Roig and alumina supplied by Vicar. Table 1 shows the compositions determined by XRF measurements.

Based on the SiO_2 – MgO – Al_2O_3 diagram [17] the raw materials were mixed in the following proportion: 20.6 wt.% alumina, 34.5 wt.% kaolin, 40.4 wt.% talc, and 4.5 wt.% silica, along with a synthetic methylcellulose polymer, which acts as a binder for the extrusion process. The raw materials are milled (with zirconia balls in a Retsch mill) and sieved (in an Endecotts Octagon 2000 Sieve). The particle size distribution was determined by laser scattering in a Malvern Mastersizer equipment.

The dry kneading was carried out in a laboratory 350-E Brabender mixer at room temperature with a speed of 40 rpm for 30 min. Once the solid mixture was homogeneous, water was added and a cohesive ceramic paste was obtained by kneading for an additional 45 min. The resultant paste was ready to be manipulated in a 19/20DN Brabender extruder with a circular geometry die. This process was conducted at a speed of 40 rpm and at 35 °C. As a result, a perfectly conformed ceramic honeycomb monolith, which has a cell density of 122 cells per square inch, was obtained with a suitable consistency to be handled in subsequent steps. The samples were dried by controlling precisely both temperature and wetness.

Table 1
Composition of the raw materials (wt.%)

	Al_2O_3	SiO_2	MgO	CaO	Na_2O	K_2O	TiO_2	Fe_2O_3
Kaolin	34.69	50.64	0.47	0.30	0.32	2.59	0.12	1.08
Talc	0.31	62.88	31.33	0.15	–	–	0.014	0.49
Silica	0.39	99.13	0.20	0.15	–	0.07	0.02	0.04
Alumina	99.44	0.21	0.03	–	0.03	–	–	0.02

The determination of the compositions was carried out using a Philips PW 1480 wavelength dispersive X-ray fluorescence (XRF) spectrometer. All the chemical elements were analysed using a side window Sc-Mo target X-ray tube and under a vacuum path.

The identification and the evolution of the crystalline phases during the cordierite honeycomb synthesis process were carried out by X-ray diffraction analysis (Phillips PW1710, $\text{CuK}\alpha$ radiation, 2θ range=5–60°, step scan=0.015) of samples calcined at different time and temperature.

3. Results and discussion

Three particle size distributions of precursor oxides, named as D1, D2 and D3, were prepared (Fig. 1). The particle size for 90% of the overall number of particles was around 30, 20 and 12 μm for the D1, D2 and D3 distributions, respectively. Decreasing the particle size, or moving a distribution of particle sizes to a smaller range, can have at least two advantages. It will (i) increase the rate of sintering for a fired product and (ii) decrease the tendency of phase separation during extrusion. A disadvantage of decreasing the particle size is an increased tendency of the powder to agglomerate prior to extrusion [18].

Previous differential thermal analysis (DTA) results allowed the optimal temperature range for the solid-state reaction to be established, between 1250 and 1400 °C [16]. In order to determine the actual optimal temperature to obtain the desired phase, it is necessary to study the evolution of the different crystalline phases for each particle size distribution in this temperature range. Likewise, it is required to estimate the treatment time that completes the solid-state reaction.

Fig. 2 shows the diffractograms obtained for monoliths extruded with the D1 distribution and submitted to various temperature treatments for 10 min. At 1250 °C the peaks corresponding to indialite, a polymorph of cordierite, can be observed. The intensity of these lines is still low due to reduced crystallinity, but this indicates that the solid-state reaction has started with the development of only a fraction of the characteristic peaks of this phase. The more abundant crystalline species at this temperature is corundum, despite the fact that other species such as cristobalite and spinel are present in reduced amounts and similar to that of indialite. The least predominant material is quartz that exhibits intense peaks associated with its more important crystallographic planes.

At 1300 °C indialite is the major species indicating that the reaction has occurred to a considerable extent. However, other raw materials that have to react to conclude the process are still present, since peaks corresponding to protoenstatite, corundum and even quartz appear in noticeable amounts. From 1300 to 1325 °C, quartz is almost consumed from the mixture but the crystallinity of the present phases is low. This lack of crystallinity, as measured

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