



Some studies on ceria–zirconia reinforced solvothermally synthesized cordierite nano-composites



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ABSTRACT

Nanostructured cordierite and 12 mol% ceria stabilized zirconia were chemically prepared separately. Pure cordierite was synthesized solvothermally, whereas [(Zr)_{0.88}(Ce)_{0.12}O₂] termed as CeSZ was processed with gelation and co-precipitation method. The evolution of crystalline phases and the microstructures have been studied using X-ray diffractometer, and FE-SEM with EDX. Varying contents (0–20 weight%) of CeSZ were then mixed with cordierite to form sample blocks of desired shapes. These nano-composite blocks were fired at 1100–1400 °C for a soaking time of 3 h to observe their thermo-mechanical, micro-structural and physical properties. Results indicate that ceria not only acts as a stabilizer in zirconia ceramics but it also acts as a sintering aid in cordierite formation. CeO₂ also improves compressibility and structural properties of nano-composites. Sintering temperature also plays an important role in the behavior of nano-composite blocks. All properties show a major improvement than previous reported data's.

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1. Introduction

In recent years, nano-crystalline ceramics and the application of nano particles to improve the ceramic properties have attracted considerable interest, as the mechanical, optical, electrical and magnetic properties are crystallite size sensitive. Natural cordierite (Mg₂Al₄Si₅O₁₈) is a rare magnesium–alumino–silicate mineral occurring in nature [1–3]. The cordierite ceramics are known due to their low thermal expansion coefficient, high chemical durability, low dielectric constants, high refractoriness and excellent resistance to thermal shock. However, cordierite ceramics are susceptible to low fracture toughness, which hinders their structural applications. Unfortunately processing of synthetic cordierite is difficult as it is always accompanied by a spinel (MgAl₂O₄) phase, which impairs the thermal shock resistance of end product due to its high thermal expansion coefficient. It is difficult to produce dense cordierite ceramics by the solid-state reaction without sintering aids because of the narrow sintering range near the incongruent melting point of cordierite [4,5].

Ceria stabilized tetragonal zirconia (CeSZ) polycrystalline ceramics possess distinct advantages over other conventional structural ceramic materials because of their better thermal stability in moist environments, wider range of solid solubility in tetragonal region [6]. Consequently, ceria stabilized zirconia ceramics

have been investigated extensively for structural applications. The grain size and microstructure of these materials are difficult to control in conventional technique. Gelation and co-precipitation process is a promising candidate for production of CeSZ particles giving a better control over size and shape of the synthesized nano-powders [7,8].

There is some data available on the cordierite synthesis by the novel chemical processes but not for structural properties [9]. The aim of this study was the synthesis of nanostructured cordierite by solvothermal technique and investigation of the phase transformation during heat treatment of precipitate. Addition of ZrO₂ (mostly in tetragonal form) in cordierite has been extensively studied but very little is known about its cubic polymorph reinforcement with effect of mineralizer additions on both the sinterability and the mechanical properties of cordierite bodies fabricated by wet chemical routes. The effect of CeO₂ as mineralizer has been studied in the present work with the addition of cubic ZrO₂ in nanostructured cordierite matrix. To do this CeSZ was incorporated in thus prepared cordierite matrix and blocks were formed to investigate their micro-structural and mechanical properties.

2. Materials and methods

2.1. Synthesis of nanostructured cordierite by solvothermal technique

A solvothermal process [10–12] was used to synthesize the cordierite nano-powders. All reagents were analytical grade, purchased from LobaChem (Mumbai, India) and used as received. First appropriate metal–nitrate precursors; Mg(NO₃)₂·6H₂O, Al(NO₃)₃·9H₂O and orthosilicic acid (H₄SiO₄) were dissolved in deionized

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water–ethanol (1:1 volume ratio) under rigorous stirring to form 0.5 molar solution. The precursors were taken in a ratio to give a stoichiometric composition of cordierite. To this solution 100 g of urea and 1 mmol sodium dodecyl sulfate (SDS) was subsequently added. The solution was sealed in a tightly closed stainless steel autoclave and heated at 120 °C for 12 h. Upon completion of the reaction, the product was centrifuged at 10,000 rpm for 5 min and washed with deionized water and ethanol. The resulting precipitate was dried at 100 °C for 24 h and then calcined at varying range of temperature (1100–1300) °C for a soaking period of 2 h to obtain cordierite nano-powders.

2.2. Synthesis of $[(Zr)_{0.88}(Ce)_{0.12}O_2]$ by gelation and co-precipitation method

AR grade $ZrOCl_2 \cdot 8H_2O$ and $Ce(SO_4)_2 \cdot 4H_2O$ were dissolved in deionized water with vigorous stirring and mild heating to give 12 mol% ceria stabilized zirconia $[(Zr)_{0.88}(Ce)_{0.12}O_2]$. For preparation of gel, ammonia solution was added drop wise and pH of the solution was maintained over 9. A point was reached when the gel became fully viscous and the process of stirring was stopped due to the viscous nature of the gel. This gel was oven dried at 100 °C for 24 h and finally calcined at 750 °C for 5 h to give super fine CeSZ [13–15].

2.3. Preparation of nano-composites

Batches were formed employing above processed nano-powders of cordierite and CeSZ in the varying ratio. Nanostructured cordierite formed at 1300 °C was selected for the formulation of nano-composites as by the X-ray diffraction analysis it showed the lowest content of spinel phase and highest content of α -cordierite. A zero weight% CeSZ containing cordierite mix was named CZ00. Similarly 5, 10, 15 and 20 weight% CeSZ containing cordierite mixes were named CZ05, CZ10, CZ15 and CZ20 respectively. These batches were pulverized in a high energy planetary ball mill. The jar and grinding media were of titanium-coated stainless steel material. At one time 250 g of a batch was taken in a jar and milled for 45 min at 600 rpm. Similarly, it was processed to complete the grinding of all batches. These nano-powdered batches were then cold pressed into desired blocks as per ASTM standards by a uniaxial hydraulic press machine under a constant 10 tones pressure load. Several blocks of each composition were made and sintered in a temperature range of 1100–1400 °C for a soaking period of 3 h. Sintered and dense composite blocks were investigated for their thermo-mechanical and micro-structural properties [16,17].

2.4. Characterization of the nano-composites

Powder X-ray diffraction patterns were recorded using a Rigaku high resolution powder X-ray diffractometer employing $Cu K\alpha_1$ radiation and Ni-filter. Data were collected in the 2θ range from 0° to 90°.

Synthesized and sintered blocks of formulated composites were examined using a FEI Quanta 200F Field Emission Scanning Electron Microscope (FE-SEM). The samples were metalized by gold sputtering for better image definition.

Apparent Porosity and Bulk Density of sintered nano-composite were investigated according to ASTM C20-00.

The Cold Crushing Strength (CCS) is the capacity of a material to withstand axially directed pushing forces. Cubic test specimens of 51 mm size were prepared and the value of maximum uniaxial load (in N) was noted when the sample block failed completely. Finally CCS value was calculated using the method stated in ASTM C133-97.

Cold modulus of rupture (CMOR) measurements was carried out under three-point bending tests (ASTM C133-97) using 152 mm \times 25 mm \times 25 mm samples.

Sample briquettes were measured for their mechanical strength in terms of cold crushing strength (MPa) and modulus of rupture (MPa) according to ASTM C133-97.

3. Results and discussions

Fig. 1 shows the XRD plot of as prepared cordierite nano-powders. Two primary factors directly determine the crystalline size of the samples, namely, nucleation and crystal growth processes. At lower temperature 1100 °C the μ -cordierite starts to form but still the major phase is recognized as a magnesium aluminate spinel ($MgAl_2O_4$). Spinel occurs as an intermediate phase which ultimately reacts with μ -cordierite at high temperatures to produce α -cordierite; α -cordierite itself is formed at temperatures >1200 °C. Here inset figure illustrates the complete formation of cordierite focusing on a specific range which clearly depicts intense and sharp peaks corresponding to α -cordierite formation {110} plane at $2\theta = 10.5^\circ$. In sample calcined at 1200 °C {241} plane at $2\theta = 29.5^\circ$ portray improvement in crystallinity of cordierite due to rise in synthesis temperature. Cordierite peaks were matched

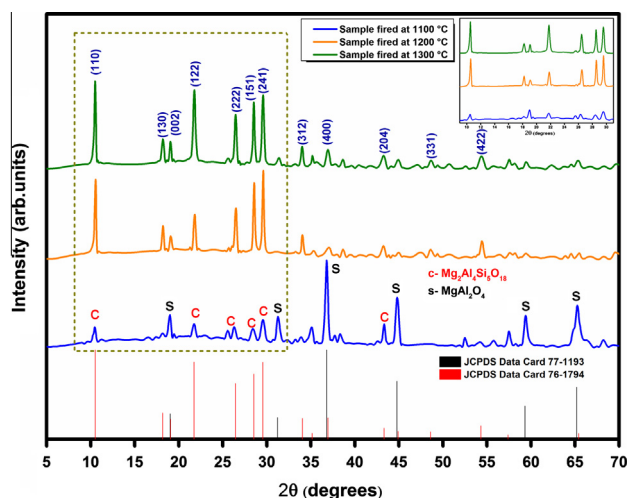


Fig. 1. XRD plot of solvothermally prepared cordierite.

with JCPDS data card 76-1794 and spinel peaks were identified with JCPDS data card 77-1193.

Presence of broad peaks in XRD patterns of calcined powders show that particle size is small. Crystalline size, d of calcined powder was calculated from X-ray line broadening analysis using Scherrer's formula.

$$d = 0.9\lambda/\beta\cos\theta$$

where β is the full width at half maximum (FWHM) intensity of a Bragg reflection excluding instrumental broadening, λ is the wavelength of the X-ray radiation and θ is the Bragg angle. β is taken for the strongest Bragg's peak corresponding to 2θ . All the three cordierite powders calcined have an average crystallite size in the range 20–35 nm.

Fig. 2 reveals the formation of gelation and co-precipitation processed 12 mol% ceria stabilized zirconia. No evidence of crystalline impurities is found in the pattern. Cubic ZrO_2 peaks matched exactly with JCPDS data card 65-0461. These results indicate that the cubic ZrO_2 with high phase-purity can be obtained in the process reported. In addition, according to the Scherrer's formula, the average crystallite size of the sample is evaluated to be approximately 18 nm from the half-peak width of the {111} diffraction peak at $2\theta = 30.13^\circ$.

Particle size distribution of as prepared Cordierite and CeSZ is shown in Fig. 3. J1.46 tool was used to identify and count the number of particles as well as their mean diameters. Maximum

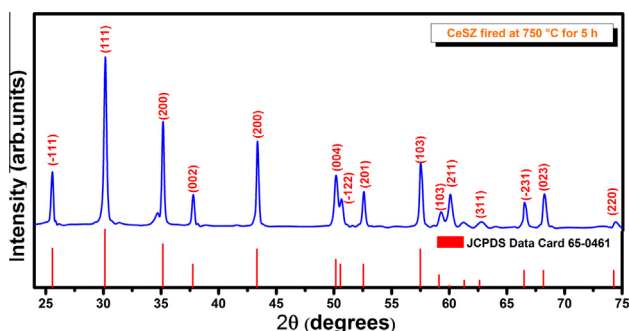


Fig. 2. XRD plot of gelation and co-precipitation processed 12 mol% ceria stabilized zirconia.

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