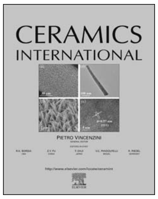
ARTICLE IN PRESS

Ceramics International xxx (xxxx) xxx–xxx

Contents lists available at ScienceDirect

Ceramics International

journal homepage: www.elsevier.com/locate/ceramint



Processing and properties of cordierite-silica bonded porous SiC ceramics

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ARTICLE INFO

Keywords:
Sol-gel processes
Cordierite
Porosity
Corrosion
Mechanical properties

ABSTRACT

Cordierite-silica bonded porous SiC ceramics were fabricated by infiltrating a porous powder compact of SiC with cordierite sol followed by sintering at 1300–1400 °C in air. The porosity, average pore diameter and flexural strength of the ceramics varied 30–36 vol%, ~ 4–22 µm and ~ 13–38 MPa respectively with variation of sintering temperature and SiC particle sizes. In the final ceramics SiC particles were bonded by the oxidation-derived SiO_2 and sol-gel derived cordierite. The corrosion behaviour of sintered SiC ceramics was studied in acidic and alkaline medium. The porous SiC ceramics were observed to exhibit better corrosion resistance in acid solution.

1. Introduction

Hot gas filtration from industrial processes offers various advantages in terms of improvement of process efficiencies, heat recovery and protection of plant. The use of porous SiC ceramics to control emissions of particulate matter in incineration, industrial wastes, metal smelting, manufacturing of cement, glass, etc., and hot gas filtration process especially for pressurized fluidized bed combustion (PFBC) and integrated gasification combined cycle (IGCC) is increasing to protect the environment [1–6]. For hot gas filtration process, the ceramic filters must be able not only to resist the chemical attack at high temperature (600–900 °C) and high pressure (6–10 atm) by variety of gases such as O_2, HCl, H_2S, Cl_2, SO_2, NO and H_2O, but also to withstand the mechanical stress or thermal shock in the pulse cleaning process [7]. SiC is especially recommended as useful material for fabrication of hot gas filters because of their unique combination of properties such as high strength, high hardness, high thermal conductivity, low thermal expansion coefficient, superb mechanical and chemical stabilities at high temperatures and hostile atmospheres. The literature survey indicates that processing of suitable SiC material for such application is still challenging. Oxide bonding is widely used technique that has been used for synthesis of porous SiC ceramics at low temperatures (1300–1550 °C) under an ambient atmosphere using commercial-grade raw powders. This technique is simple and inexpensive as it does not require any sophisticated equipment and delicate instrumentation. Clay, silica and mullite are often used as oxide bonds for SiC [8–12]. Cordierite ($2MgO{\cdot}2Al_2O_3{\cdot}5SiO_2$) is also considered another important bond as it has a low thermal expansion coefficient, good thermal and chemical stability and can be processed at a low temperature [13].

The oxide bonding technique was found to have several disadvantages as oxidation of SiC is limited in the depth of the zone hence limited for large scale production and mechanical failure of the final ceramics occurred due to inhomogeneous mixing of fine sintering aids. To avoid those problems our group prepared mullite bonded porous SiC ceramics following incorporation of sol-gel bond phase precursor by infiltration technique to ensure a homogeneous distribution of the bond phases [14]. The applicability of ceramic filter as hot gas filter depends on the filtration performance, durability, etc. in actual hot gas filtration condition. Most of the previous studies on off-gas cleaning in advanced power generation process were carried out using commercial SiC ceramic candle filters [4–6,15–17]. Recently many authors reported the permeability and collection efficiency behaviour of porous ceramics using the laboratory test setup to filter nano aerosol particles [18–20]. Very few studies are reported in literature on evaluation of filtration performance of the ceramic filter using laboratory made test setup to filter coal fly ash particles in actual power generation conditions [21]. The objective of the present work was to prepare rectangular and disc types SiC ceramic filter by incorporation of cordierite as secondary bond phase following infiltration technique. The effect of starting particle size of SiC and sintering temperatures on the formation of oxide bond phases, porosity, microstructure and mechanical properties were examined. In addition, corrosion resistance properties of final porous SiC ceramics were studied in acid and alkaline salt medium at 90 °C with respect to time.

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http://dx.doi.org/10.1016/j.ceramint.2017.07.196
Received 21 June 2017; Received in revised form 26 July 2017; Accepted 27 July 2017

Please cite this article as: Baitalik, S., Ceramics International (2017), http://dx.doi.org/10.1016/j.ceramint.2017.07.196

2. Experimental

Commercial α-SiC powder (Grindwell Norton, India) of three different grit sizes 400 (d_{50} = 22.4 μm), 220 (d_{50} = 51.7 μm) and 150 (d_{50} = 99.2 μm) were used in this study. The powders individual grit sizes were mixed with 15 wt% solution of polyvinyl alcohol binder (Loba Chemie, India) and pressed in a hydraulic press to produce rectangular green bars (50 × 20 × 16 mm^3). The bars were dried at 100 °C and subsequently fired in air at 1100 °C for 4 h to obtain oxidation derived silica bonded porous SiC network with sufficient handling strength. The rectangular bars were evacuated and infiltrated with a liquid precursor of cordierite ($5SiO_2{\cdot}2MgO{\cdot}2Al_2O_3$). The process was repeated until the weight gain became constant. Infiltration kinetic analysis of SiC compacts of three different particle sizes were carried out by suspending the well-dried and evacuated small piece of SiC bar sample into cordierite sol which was attached to an electric balance by a wire and its weight change was recorded as a function of time. For the preparation of cordierite sol [22] at first step, magnesium nitrate hexahydrate ($Mg(NO_3)_2{\cdot}6H_2O$), and aluminium nitrate nonahydrate ($Al(NO_3)_3{\cdot}9H_2O$) were dissolved in ethanol in 2:4 M ratio and this solution was kept as precursor (A). In the second step, TEOS (Tetraethyl orthosilicate) was taken in ethanol in 1:3 M ratio and was kept as precursor B. Precursor A was added drop wise into B at room temperature with continuous stirring for 6–7 h to form transparent sol in which stoichiometric molar ratio 2:4:5 of Mg:Al:Si was strictly maintained. The sol viscosity was measured in rotational viscometer (MCR-102, Anton Paar GmbH, Graz, Austria, Europe) with cup and stirrer geometry (ST-24-2D/2 V/2V-30). The particle size of the sol was measured using Zetasizer (ZEN 3690, Malvern Instrument Ltd. Worcestershire, UK). The thermogravimetric analysis (TGA) of cordierite gel was carried out at a heating rate of 20 °C/min up to 1400 °C in dry air medium (STA 490 C, Netzcsh-Geratebau, GmbH, Germany).

The infiltrated bar samples were finally sintered in air at 1300–1400 °C for 3 h with a hold at 750 °C for 2 h to remove possible fugitives from the infiltrated compacts in an electrically heated ceramic tube furnace (Model No.-TE-3499-2, M/S Therelek Engineers (P) Ltd., Bangalore, India). The samples were characterized by measurement of density and porosity by water immersion method, identification of crystalline phases by XRD analysis (PW1710, Philips, Holland) using Cu- Kα radiation of wavelength α = 1.5418 Å and determination of pore size distribution (PSD) by Hg intrusion porosimeter (Poremaster, Quanta chrome Instruments Inc., Florida, USA). The quantitative phase analysis was done by the Rietveld technique using the High Score Plus software (version 3.0e, PAN analytical B.V.). The room temperature flexural strength was determined in three-point mode (with a span of 40 mm, speed of 0.5 mm/min, sample cross-section of 4.75 by 2.25 mm^2; samples were ground and polished up to 10 μm finish and tensile surfaces were chamfered) using an Universal Testing Machine (UTM) (Model 1123, Instron, Canton, MA, USA). The deflection was monitored through LVDT with a resolution of 0.05% of full scale and from the load-deflection data Young's modulus was determined using standard software (Instron Bluehill-2, UK). The average of the five readings was reported for the mechanical property data. The microstructure was examined by the scanning electron microscopic (SEM) technique (SE-440, Leo-Cambridge, Cambridge, UK). To study the effect of acid-base corrosion on the SiC ceramics, small cut piece (40 × 4.75 × 3.5 mm^3) of cordierite bonded SiC samples sintered at 1400 °C were kept into solutions of 20 wt% HCl (pH ~ 1) and in 20 wt% $NaHCO_3$ (pH ~ 11.5) respectively at 90 °C for 10 days and mass loss was recorded with time. For further characterization, the corroded samples were washed several times with boiling water to ensure complete removal of leached constituents from the surface and bulk. The three point bending strength of the corroded specimens was measured. The changes in surface morphologies were investigated and oxide bond phase compositions were determined to observe the effect of acid-base corrosion on the SiC specimens.

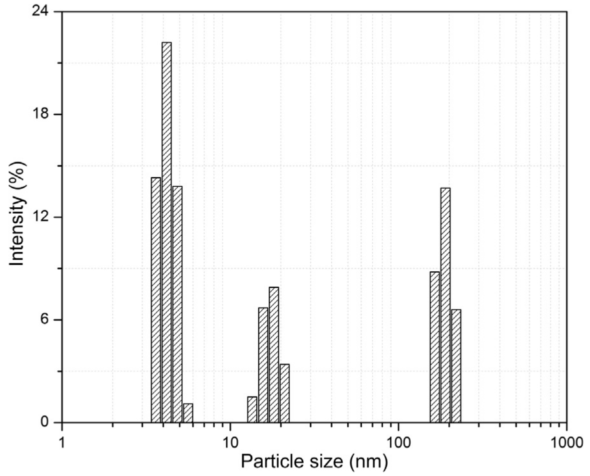

Fig. 1. Particle size distribution of cordierite sol.

3. Results and discussion

3.1. *Characteristics of cordierite sol*

The broad particle size distribution pattern of the sol is shown in Fig. 1. The cordierite sol particles exhibited multimodal size distribution with three major peaks with an average particle diameter 4.2 nm, 17.6 nm and 190 nm respectively. The wide variation was due to common tendency of sol agglomeration, still the average diameter of the sol particles were significantly smaller than the pore diameter of the pressed green bars. The viscosity of sol was found to be 14.2 mPa s at 100 1/s shear rate. Thermal analysis result of cordierite gel is presented in Fig. 2. The endothermic peak appeared below 200 °C was corresponding to ~ 16% weight loss which can be attributed due to desorption of water part from the cordierite gel. The first exothermic hump appeared at ~ 390 °C, resulted ~ 43% of weight loss due to the release of weak bonding organic groups and oxidation of residual organic group. Rapid reduction of weight was observed up to ~ 400 °C and above which a slow reduction of weight was noticed and the weight became constant (~ 53 wt% mass retention) at the temperature higher than 700 °C. In DTA analysis of cordierite gel, Janković-Častvan et al. [23] reported exotherm at ~ 967 °C for crystallization of μ-cordierite phase from amorphous phases. Two exotherms at 987 and 1305 °C and three endotherms at 850, 1225 and 1350 °C are reported [24] in TG-DTA analysis of cordierite gel prepared from nitrate precursors. The authors explained the first exothermic peak for the formation of μ-

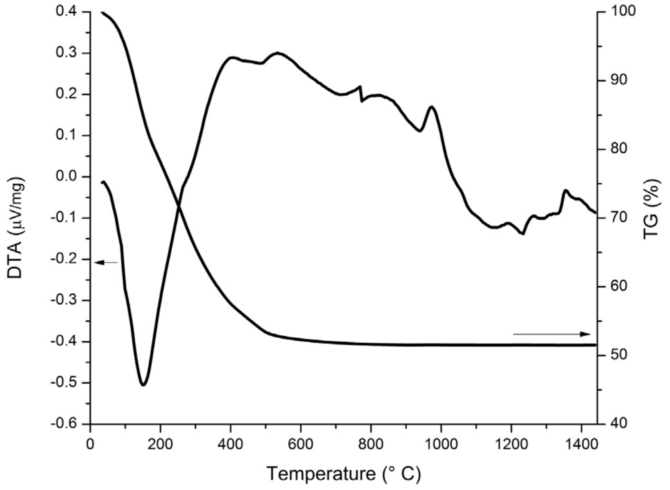

Fig. 2. TG – DTA results of dried cordierite gel during heat treatment in air up to 1450 °C.