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Investigation of thermal oxidation of Al_2O_3-coated SiC powder

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ABSTRACT

SiC powder (β-form) of sub-micrometre size (d_{50} of 0.65 μm) was coated with a thin layer (~12 nm) of boehmite by sol–gel technique. The final powder having ~24 wt% boehmite was characterized by particle size and surface area determination, chemical analysis, surface potential measurement and Fourier transform infrared (FTIR) spectroscopic analysis. The thermal oxidation behaviour of the coated powder was examined using the simultaneous TG/DTA technique in dry air. The results indicated crystallization of mullite phase and the activation energy of crystallization was determined to be 368.9 ± 15.1 kJ mol^{-1}. Synthesis of porous SiC ceramics of fine pore structure (with average porosity and pore size ranging from 47% to 54% and 170–260 nm, respectively) using thermal oxidation of coated powder was also demonstrated.

1. Introduction

SiC ceramics have low bulk density, excellent mechanical and chemical stability, high thermal conductivity and thermal shock resistance, and they can also withstand high temperatures and hostile atmospheres. Processing routes are found to retain or create porosity without sacrificing these special properties. One of the important methods of producing porous SiC ceramics of pore size in micrometre scale involves heating of a porous compact of SiC powders in air when oxidation derived silica bridges the gap between the SiC particles at the contacting regions forming a porous body [1–3]. Al_2O_3 is added as a secondary phase in the starting mixture to produce mullite for improvement of mechanical properties, corrosion and thermal shock resistance of the porous ceramics [4–6]. Porous SiC ceramics produced by the oxidation bonding technique are promising filter materials and they can be used for removal of dust and fine particles in the off-gas cleaning systems of many important industrial processes (coal combustion and gasification processes for power generation, thermal remediation of contaminated soils, incineration of hospital and industrial wastes, metal smelting, manufacturing of cements, carbon blacks and glasses, etc. [7–10]). Efforts are also made to bring the emissions of ultrafine particles – particles finer than 0.1 μm – from industrial processes under the stringent regulatory control [11]. Porous media composed of agglomerates or granules of ceramic nanoparticles are successfully used for filtration of submicron solid or liquid aerosol particles [12]. In this respect development of oxidation bonded porous SiC ceramics of very fine pore structure using SiC–Al_2O_3 powders of sub-micrometre sizes assumes importance. For this purpose complete understanding of oxidation behaviour of ultrafine SiC + Al_2O_3 powder systems becomes necessary. Al_2O_3 coating on SiC particles may serve as a way to distribute the secondary phase more uniformly in the system of SiC–Al_2O_3 powder of submicron sizes and makes possible the use of coated powder in oxidation bonding of porous SiC of fine pore sizes. In this paper we present the results of an investigation on Al_2O_3 coating of SiC powder of sub-micron size and the kinetic study of oxidation of the coated powder.

2. Experimental procedure

β-SiC powder (Grade 059N/200; Lot # 050710C10; SiC: Min. 98% (w/w), free C: Max. 2.0% (w/w), oxygen: Max. 1.1% (w/w), nitrogen: Max. 0.25% (w/w), metallic Si: Max. 0.03% (w/w); Superior Graphite, Chicago, IL, USA) with an average particle size (d_{50}) of 0.65 μm and surface area of 11–16 m^2 g^{-1} was used. Boehmite coating of SiC powder was carried out following the method described by Yang and Shih [13]. Aluminium tri-sec-butoxide (Product No. 201073; Mol. Formula: $C_{12}H_{27}AlO_3$; Purity: 97%; Sigma Aldrich Co., MO, USA) was used as the precursor for boehmite. As-received SiC powder was dispersed in de-ionized water with ultrasonication (Sonaprob PR-1000MP, Oscar Ultrasonic Sakinaka, Mumbai, India) to disintegrate the agglomerates. The suspension was heated to 90 °C under constant magnetic stirring. 97% aluminium-tri-sec-butoxide was then added and the molar ratio of alkoxide to water

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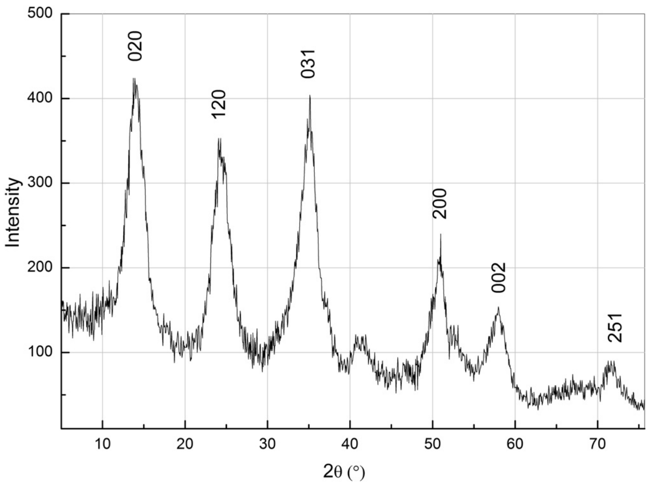

Fig. 1. XRD pattern of the precipitate from the aluminium secondary butoxide solution after drying at 100 °C.

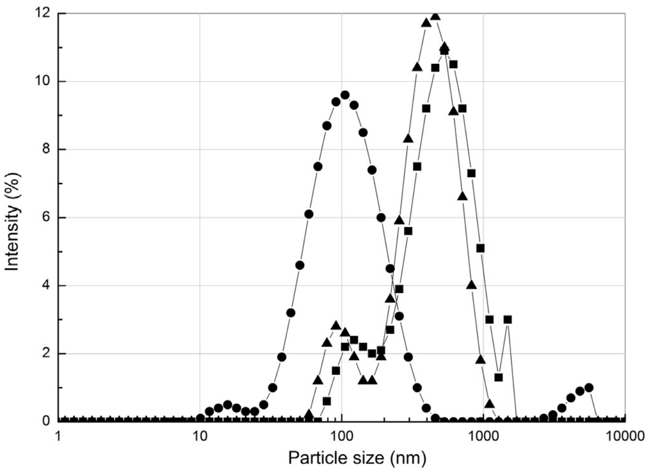

Fig. 2. Particle size distribution of different particles: (●) boehmite coating material; (▲) SiC powder; (■) SiC powder coated with ~24 wt% boehmite.

was kept at 1:150 in order to ensure complete hydrolysis. After the alkoxide addition, HCl was continually added to maintain the suspension pH at 3. After stirring for about half an hour the suspension was centrifuged (REMI-PR-24, Remi Electronics Ltd., Vasai, India) and the coated powder was obtained by repeatedly rinsing with water followed by drying and grinding. In the present work the final powder with ~24% boehmite coating was prepared using 12 g of aluminium tri-sec-butoxide and 8.8 g of SiC powder. The coated powder was characterized by measuring particle size (ZEN 3690, Malvern Instrument Ltd., Worcestershire, UK) and, BET surface area (NOVA 4000e, Quantachrome Instrument, FL, USA). Zeta potential measurement (ZEN 3690, Malvern Instrument Ltd., Worcestershire, UK) of the coated and uncoated SiC powders was done and their Fourier transform infrared transmission spectra (Nicolet 5700, Thermo Scientific, MA, USA; wave number range of 400–4000 cm^{-1}) were also recorded. The amount of coating in the final powder was determined by wet chemical analysis method (complexometric determination of aluminium). The oxidation experiment was carried out using a simultaneous thermal analyser (STA 449 F3 Jupiter, NETZSCH Geratbau GmbH, Selb, Germany). DTA tests under non-isothermal conditions were performed keeping a constant flow of dry air (XL Grade, BOC India Ltd., Kolkata, India; flow rate of 60 ml min^{-1} under atmospheric pressure) at different linear heating rate (20 K min^{-1}; 25 K min^{-1}; 30 K min^{-1} and 40 K min^{-1}).

3. Results and discussion

3.1. Preparation of boehmite coated SiC powder

The coating material was produced from the aluminium secondary butoxide solution following the same process but without the presence of SiC. The particles so produced were boehmite as was evident from the XRD pattern of the precipitate from the aluminium secondary butoxide solution after drying at 100 °C (Fig. 1). Formation of boehmite was also reported in the experimental conditions similar to those of present study [13]. The average particle size of SiC powder before and after coating was measured to be 400 and 479 nm, respectively, based on the number average and 517 and 689 nm, respectively, based on the volume average (Fig. 2). The results of particle size measurement tests indicated presence of very few free boehmite particles in the suspension of coated particles. The mean particle size of boehmite was reported to be around 80 nm [13]. In the present study an average particle size of ~90 nm has been obtained (Fig. 2). None of the peaks centred on this average particle size was found in case of coated powder. The average particle size of SiC increased after the coating process. The individual boehmite particles possibly aggregated on SiC particles increasing the average particle size after coating. Fig. 3 shows the zeta potential versus pH plots for the uncoated and coated SiC particles. The SiC particles exhibited the isoelectric potential (IEP) at pH 4.6. With boehmite coating the IEP shifted to a higher value at pH 9.4. The boehmite powders were reported to have an IEP at pH 9.8 [14]. The surface of boehmite has neutral surface sites and can react with either H^+ or OH^- ions in acidic or basic solutions to produce dominant positive or negative surface sites below or above the IEP, respectively. After coating SiC particles with boehmite the surface was terminated with neutral surface sites and the behaviour of the coated SiC suspension became similar to that of boehmite suspension. The zeta potential measurement test further confirmed that the coating was successful as the surface potential of SiC changed to that of boehmite after coating. The FTIR transmission spectrum of the uncoated powder had the characteristic peak of SiC at 866 cm^{-1} (Fig. 4). For coated powder peaks appeared at 487, 534, 649, 717 and 877 cm^{-1}. The band position at 717 cm^{-1} indicated formation of the Al–O–Si linkage [15]. The peaks at 487, 534 and 649 cm^{-1} were for boehmite [16]. The peak at 877 cm^{-1} was similar to the characteristic peak of SiC; Lee and Kim examined simultaneous hydrolysis and coating of $Al(OH)_3$ on SiC particles in water/aluminium

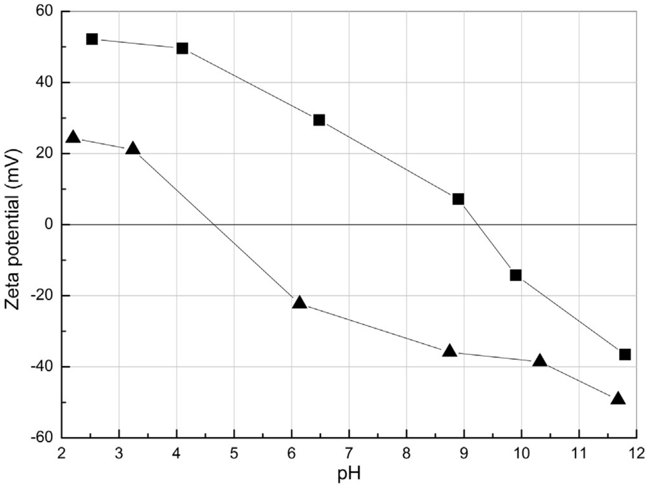

Fig. 3. Zeta potential versus pH: (▲) SiC powder; (■) SiC powder coated with ~24 wt% boehmite.