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Preparation of silicon carbide ceramics using chemical treated powder by DCC via dispersant reaction and liquid phase sintering

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ABSTRACT

Dense silicon carbide ceramics using chemical treated powder by DCC via dispersant reaction method and liquid phase sintering was reported. Ammonium peroxydisulfate ($(NH_4)_2S_2O_8$) and ammonium carbonate ($(NH_4)_2CO_3$) were used as acid and base solutions to treat the silicon carbide powder, respectively. Influence of silicon carbide powder with chemical treatment on the preparation of silicon carbide suspension was studied. It was indicated that 50 vol% and 52 vol% silicon carbide suspensions with viscosities of 0.71 Pa s and 0.80 Pa s could be prepared using acid and base treated powders. Influence of silicon carbide powder with chemical treatment on the coagulation process and properties of green bodies and sintered ceramics were studied. It was indicated that silicon carbide green bodies with compressive strength of 1.13 MPa could be prepared using base treated powder. Dense silicon carbide ceramics with relative density above 99.3% and flexural strength of 697 ± 30 MPa had been prepared by DCC via dispersant reaction and liquid phase sintering using Al_2O_3 and Y_2O_3 as additives at 1950 °C for 2 h.

1. Introduction

Silicon carbide ceramics as a kind of promising candidate material for high performance ceramics has many excellent properties, such as high hardness, high mechanical strength, high resistance to corrosion and so on [1,2]. Silicon carbide ceramics have been widely applied in petroleum, chemical, micro electronics, automotive, aerospace, aviation, laser, mining and atomic energy and other industrial fields [3–5]. However, due to the brittle nature and high hardness, post machining will be difficult and significantly increase the cost of SiC product especially in the fabrication of parts with large size and complex shape [6,7].

Direct coagulation casting (DCC) via dispersant reaction is a near–net–shape forming method. The coagulation mechanism is that dispersant reacts with the coagulation agent and desorbs from the surface of particles to reduce the zeta potential of the suspension. This method does not need to adjust the pH value with the advantages of simple experimental procedure, less additive and short coagulation time etc. [8]. Though the preparation of high solid loading and low viscosity suspension is highly demanded in DCC process, the preparation and dispersion of high solid loading silicon carbide suspension is often hindered. The surface oxidation and ion content of silicon carbide powder are the main factors that hinder the dispersion of silicon carbide powder in the dispersion medium [9]. Surface chemical treatment can effectively reduce the degree of surface oxidation and ion content on the powder. The influence of silicon carbide powder with chemical treatment on rheological properties of silicon carbide suspension had been characterized in previous reports [9–11]. Yet, the influence of silicon carbide powder with chemical treatment on the coagulation process and properties of sintered ceramics has been rarely reported.

In the present work, the oxidation degree and ion content of silicon carbide powder are effectively reduced by chemical treatment. Well dispersed silicon carbide suspension was prepared using chemical treated powder. Influence of chemical treatment on preparation of suspension, coagulation process, properties of green bodies and sintered ceramic were studied. Dense silicon carbide ceramic with excellent mechanical properties was prepared by DCC via dispersant reaction method and liquid phase sintering.

2. Experimental

2.1. Materials

Commercially available α–SiC powder (Qinhuangdao Eno High–Tech Material Development Co., Ltd. China) with average par-

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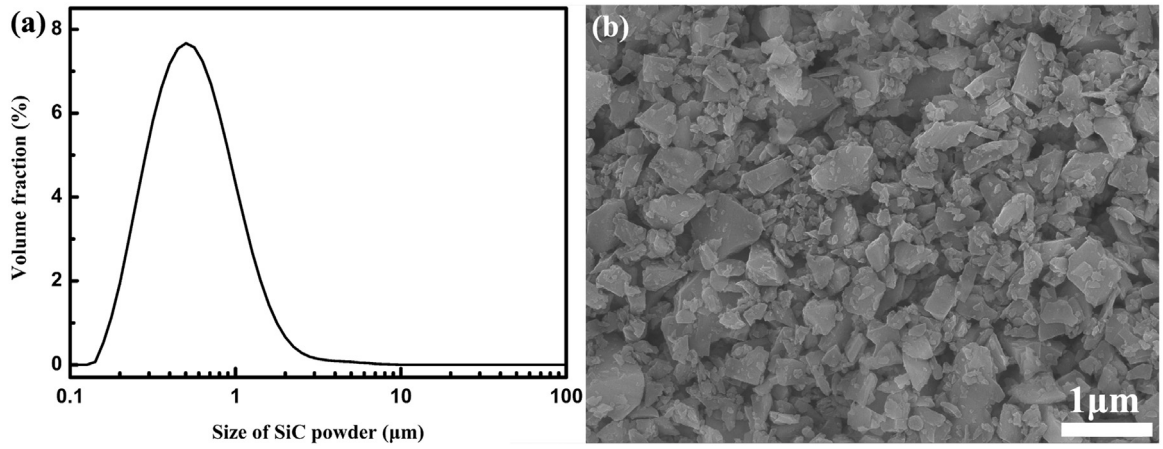

Fig. 1. (a) Particle distribution and (b) microstructure of as–received SiC powder.

ticle diameter of 0.5 μm and specific surface area of 9.62 m^2/g was used. Fig. 1 shows the (a) particle distribution and (b) microstructure of SiC powder. For liquid phase sintering, CT3000SG alumina powder (Almatis, Ludwigshafen, Germany) with average particle diameter of 0.33 μm, yttrium oxide powder (Shanghai Junyu Ceramic–molded Product Co., Ltd., China) with average particle diameter of 0.39 μm were used as sintering additives. Ammonium peroxydisulfate ($(NH_4)_2S_2O_8$) and ammonium carbonate ($(NH_4)_2CO_3$) were produced from Sinopharm Chemical Reagent Co., Ltd., China which were used to leach the silicon carbide powder. Tetramethyl ammonium hydroxide (TMAOH) aqueous solution with concentration of 10 wt% was used to disperse the suspension. Glycerol diacetate (GDA) was used as coagulation agent. Both were purchased from Hengye Zhongyuan Chemical Co., Ltd., Beijing, China. Analytical purity ammonia was used to tailor the pH value of the suspension. Deionized water was used in all preparation processes.

2.2. Powder treatment and preparation of suspension

In DCC process, high solid loading and low viscosity suspension was needed. Ammonium peroxydisulfate and ammonium carbonate were used to soak and leach the as–received silicon carbide powder by a vacuum machine till pH = 2.5–3 and pH = 10–10.5, respectively. Then, wet powder was washed and leached by deionized water till pH = 7.5–8. The treated powder was dried in a drying closet at 80 °C for 24 h. The as–received silicon carbide powder is abbreviated as **AR**, the acid and base leached silicon carbide powders are abbreviated as **AL** and **BL** in all figures and tables of this paper, respectively. Silicon carbide suspensions with different solid loadings were prepared by tumbling the treated silicon carbide powder, sintering additives, water, dispersant, ammonia, and grinding media in polyethylene container for 24 h. Tetramethyl ammonium hydroxide was used as dispersant with 0.1–0.5 wt% based on silicon carbide powder. For liquid phase sintered, 4 wt% Al_2O_3 and 6 wt% Y_2O_3 based on silicon carbide powder were used as sintering additives. Agate balls with diameter of 5–10 mm were used as grinding media. The mass ratio between grinding media and silicon carbide powder was 1:2. Ammonia was used to adjust the pH value of the suspension to ca. 10.5–11.

2.3. DCC and sintering

Suspensions with different solid loadings prepared using as–received and treated powder were degassed under vacuum condition for 20 min. 2 vol% glycerol diacetate based on silicon carbide suspension were added and mixed thoroughly by continuing the tumbling process for another 20 min to increase the homogeneity of the suspension. Then, the suspension was cast into a plastic mold. The samples were placed in a water bath with a heating rate of 5 °C/min and treated at different temperatures for a period of time and then demolded. The green bodies were dried at 80 °C for 24 h. For the liquid phase pressureless sintering process, the dried samples were sintered at 1950 °C for 2 h at a heating rate of 5 °C/min, under argon atmosphere (0.1 MPa).

2.4. Characterization

The phases of silicon carbide powder and ceramic were analyzed by X–ray diffraction (XRD) method using Cu Kα radiation (D8ADVANCE, Bruker, Karlsruhe, Germany). Wave length–dispersive sequential X–ray fluorescence spectrometer (XRF–1800, Shimadze, Tokyo, Japan) with a Rhodium target was used to analyze the chemical composition of silicon carbide powder. X–ray photoelectron spectroscopy (XPS) microprobe using monochromated micro–focused Al K–Alpha radiographic source with spot size of 200–900 μm (ESCALAB 250Xi, Thermo Fisher Scientific, USA) was used to analyze the degree of surface oxidation of silicon carbide powder. Zeta potential was measured by a Zeta Potential Analyzer (CD–7020, Colloidal Dynamics Co., Ltd., Ponte Vedre Beach, FL, USA) via the electroacoustic measurement technique with a stirring speed set at 300 r/min. In zeta potential measurement, 10 vol% silicon carbide suspension was prepared. 1 mol/l HCl and NaOH solutions were used to adjust the pH value. The rheological properties of the suspension were measured using a rheometer (KINEXUS PRO, Malvern Instruments, Worcestershire, UK) attached with C25 R0634 SS spindle and PC25 C0138 AL cylinder. The pH value of silicon carbide suspension was measured by pH meter (LE438, Mettler, Toledo, Switzerland). For wet compressive strength measurements, cylindrical bodies with 25.5 mm in diameter and a height between 25 and 30 mm were cast. The samples were demolded after coagulating, the wet green strength was measured immediately in a mechanical testing machine (AG–IC20KN, Shimadzu, Tokyo, Japan) with a crosshead speed of 0.5 mm/min. Using the same mechanical testing machine, the flexural strength of the specimens with dimension of 3 mm × 4 mm × 36 mm was measured via the three–point bending test. The compression faces of samples were polished before measurement. The density of the sintered samples were measured using the water displacement technique. The microstructure of the green bodies and plasma–etched (by a CF_4 gas) and fracture surface of sin-