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ScienceDirect

Ceramics International 41 (2015) 6478–6487

CERAMICS INTERNATIONAL

www.elsevier.com/locate/ceramint

A low cost preparation of C/SiC composites by infiltrating molten Si into gelcasted pure porous carbon preform

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Received 26 November 2014; received in revised form 8 January 2015; accepted 19 January 2015
Available online 22 January 2015

Abstract

C/SiC composites were prepared by infiltrating molten Si into gelcasted pure porous carbon preforms derived from mesocarbon microbeads (MCMBs). First large size flaw-free carbon preforms with smooth surfaces were produced by gelcasting method. The porosity and pore size of the carbonized preforms can be adjusted by changing the solid loading (MCMBs) ranging from 40 to 65 wt%. Subsequently, C/SiC composites were fabricated by liquid silicon infiltration of the porous carbon preforms. The results show that the preforms with solid loading from 40 to 50 wt% were completely infiltrated with Si and formed a C/SiC with homogeneous microstructure. While "SiC shell—black carbon core" structures were observed for the C/SiC composites with solid loading above 50 wt%, indicating that the Si infiltrating process was stopped in the surface of the preform. Reaction and infiltration mechanism analysis reveals that the siliconization process of porous carbon preform and the microstructures of the C/SiC samples were mainly affected by the reaction activity of carbon materials and pore structures of carbon preforms.

Keywords: C/SiC composites; Gelcasting; Mesocarbon microbeads; Liquid silicon infiltration; Microstructure

1. Introduction

C/SiC composite products are attractive due to their excellent properties, such as high thermal resistance, strength, wear resistance, hardness, good electrical conductivity, and self-lubrication property [1–4]. Therefore they are widely used in plain bearing bush, mechanical sealing, high-temperature refractory, aerospace products, water lubrication thrust bearing, and so on [5–7]. Usually two steps are needed for fabricating this kind of carbon (or graphite) and silicon carbide contained composites: preparation of porous carbon based preforms and infiltration of molten Si into the preforms [2,8–10]. Liquid silicon infiltration (LSI) process is a cost effective method for fabricating C/SiC composites due to its lower processing temperature, shorter reaction time and near-net shape fabrication capability.

The main source of the carbon preforms generally includes graphite powder, soot, carbon black, carbon fiber, etc. However, some binders such as resin and pitch are necessary during the green body forming process using above carbon sources [11–12]. Mesocarbon microbeads (MCMB) have been reported to be a superior precursor for high performance carbon/graphite material because it has good self-sintering behavior, high yield of carbon and homogeneous shrinkage [13–17]. MCMB derived from liquid pitch or organic compounds [13], consists of aromatic oligomers where aromatic layers stack approximately parallel to each other in the same direction. It is generally believed that the β-resin on the surface of MCMB acts as a binder. Therefore no additive is necessary in the forming and sintering processing when one use MCMB as carbon source.

The traditional carbon based preforms are formed by mould pressing. However the size and shape of the preforms are limited by the mould configuration. Moreover, the density and porosity at different part of the carbon preform is not uniform

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http://dx.doi.org/10.1016/j.ceramint.2015.01.087
0272-8842/

induced by the inhomogeneous of mould pressure. It always resulted in higher density and lower porosity in the surface contacted with the mould, which is unfavorable to the liquid silicon infiltration. Fortunately, gelcasting, a colloidal forming process, can overcome the problems of mould pressing due to the liquidity of the suspension [14,18]. Many complex shape and large size green bodies with homogeneous distribution are produced using gelcasting method. Additionally gelcasting is suitable for various mould material including metal, glass and polymer. Recently gelcasting has been widely used to fabricate homogeneous porous ceramics [8,18–22]. The typical process is dispersing ceramic powder in pre-mixed organic monomer solution to prepare suspensions with high solid loading and low viscosity, and then solidifying in a mould. The pore characteristics of the formed ceramic are mainly affected by the powder size and its solid loading [14,23,24].

However, no previous studies have been found using MCMBs as carbon powders for gelcasting a porous carbon preform, which could be further used to fabricate C/SiC composites. In this paper, using MCMB as the carbon source, a pure porous carbon preform was produced by gelcasting processing. The porosity of the carbon preform is adjusted by varying the solid loading. Finally C/SiC composites are fabricated by infiltrating molten Si into the pure porous carbon preform. The influence of reactivity and pore structures of the carbon preform to microstructure of the C/SiC composites is investigated. The carbonization of the carbon preform and the reaction mechanism of Si/C are also analyzed.

2. Experiment and analysis

2.1. Materials

The MCMB, a mean particle size of 21 μm, was supplied by Tianjin Tiecheng battery materials Co. Ltd., China. For gelcasting, acrylamide (AM) and *N′N′*-methylenebisacrylamide (MBAM) were used as monomer and coupling agent, respectively. The catalyst was *N′N′N′N′*-tetramethylethylene diamine (TEMED) and the initiator was ammonium persulphate (5 wt% water solution). The dispersant used for stabilizing MCMB suspension was polyvinylpyrrolidone (PVP).

2.2. Preparation of suspensions, carbon preforms and the C/SiC composites

The flowchart of the whole process is described in Fig. 1.

In order to form a stable suspension, the deionized water, AM, MBAM was at an optimal mass ratio of 100:18:0.9. The dispersant was 1 wt% based on the MCMB powder weight. First, AM and MBAM were added into deionized water forming a monomer solution. Second, the dispersant and MCMB were added into the pre-mixed solution to form a stable suspension. Then the suspension was ball milled for 16 h and de-aired by a vacuum pump with a 0.095 MPa vacuum degree for 1 h. Third, the catalyst and initiator was added into the suspension, respectively. Finally, the suspension was poured into a Teflon mould and then moved to a 70 °C water bath for about 0.5 h forming a wet body. After being demoulded, the wet body became green body slowly when dried at room temperature in a controlled humidity chamber. Some green bodies with complex shape, large size, smooth surfaces and flaw free were prepared by gelcasting in this study, such as a rectangle block with size 100 mm × 45 mm × 15 mm, a circle plate with size *Φ*90 × 15 mm and a tube with a *Φ*25 internal diameter, *Φ*35 outer diameter and 25 thickness. Then the green body was carbonized to carbon preform in a tubular furnace in nitrogen atmosphere at 900 °C. The carbon preforms with different solid loading (SL) including 40, 45, 50, 55, 60, 65 wt% are prepared and SL40, SL45, SL50, SL55, SL60, SL65 are used for short in the following discussion. Here, the solid loading is the ratio of the mass of MCMB powders to the total mass of deionized water and MCMB powders. Finally the carbon preforms after carbonization were infiltrated by molten Si in a vacuum furnace at temperature of 1550 °C for 0.5 h; then the C/SiC composites were prepared.

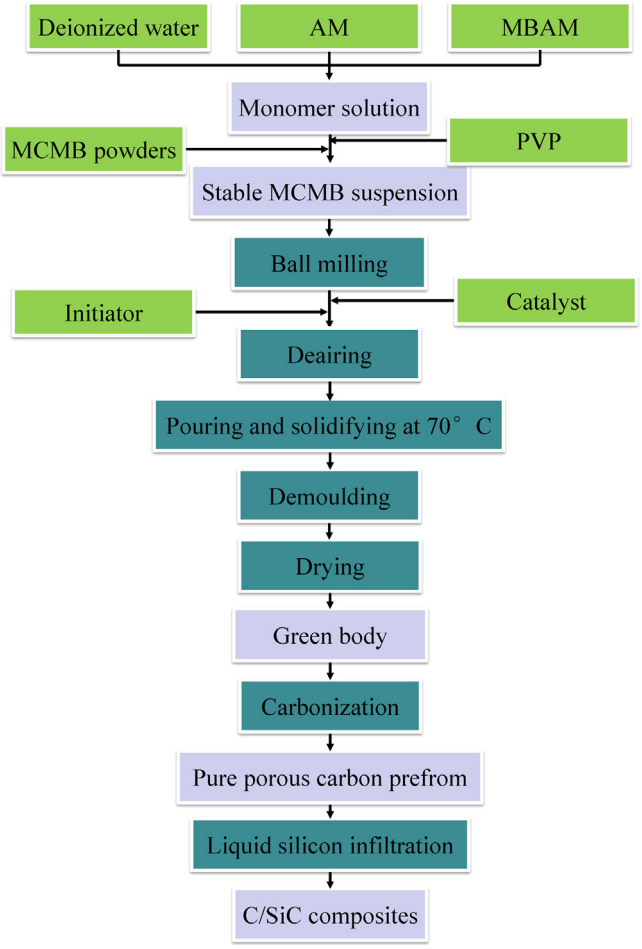

Fig. 1. The flowchart of the process for fabricating C/SiC composites.

2.3. Characterization

Differential thermal analysis (DTA) and thermogravimetric (TG) analysis of MCMB powders and gelcasted green body were carried with a TGA2950 (TG, USA) analyzer, at a rate of 10 °C/min under N_2 atmosphere air. After carbonization, the