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Properties of Si/SiC ceramic composite subjected to chemical vapour infiltration

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Abstract

Si/SiC ceramic composite was prepared by infiltration of liquid silicon into carbon preforms that was made from cotton fabric and phenolic resin. This composite was subjected to the chemical vapour infiltration (CVI), using methyltrichlorosilane as a precursor gas. The effect of infiltration time on densification and mechanical properties was studied. Results show a significant improvement in density by pore closure. Flexural strength increases with increasing infiltration time. However, beyond 60 h of infiltration, the strength improvement was insignificant. The high temperature oxidation resistance of the above ceramics was also studied. The CVI treated samples show considerable resistance to oxidation compared to untreated samples. Thermogravimetric analysis also confirmed the better oxidation resistance of the CVI treated samples.

Keywords: SiC; Cotton fabric; Chemical vapour infiltration; Porous ceramic; Carbothermal reaction

1. Introduction

Silicon carbide has long been recognized as an ideal material for applications where superior hardness and stiffness, strength and oxidation resistance at elevated temperatures, high thermal conductivity, low coefficient of thermal expansion, and resistance to wear and abrasion are of primary importance [1–5]. Silicon carbide has been made using various methods such as hot pressing, reaction bonding, etc. In the last decade, making silicon carbide ceramics from natural materials has evoked particular interest [6–9]. In this approach, the carbon preform is prepared by pyrolysing the natural wood under N_2 atmosphere in a furnace. The resulting porous carbon preform is infiltrated with liquid silicon. The main advantages of this approach are cheap and renewable raw material and capability to create near net shape. The significance of wood based SiC is that the complex hierarchical cellular structure is preserved in the final product [10–13]. Similarly, one could produce silicon carbide ceramics from any carbon yielding materials. A few researchers attempted to make silicon carbide ceramics from cellulose fiber based on cotton [14,15]. The main drawback, however, is the inhomogeneous pore distribution and pore size. The strength of this ceramic is controlled by pores, free silicon and free carbon. The reported open porosity values vary from 2% to 20%.

Chemical vapour deposition remains an important technique to synthesize ceramics in structural and electronic industries [16–18]. A range of precursors have been used for the deposition process, under widely varying conditions of input gas composition, temperature and pressure [19]. In chemical vapour infiltration (CVI) processing, the substrate is heated up to the desired temperature in an inert atmosphere and then the reactant gases are introduced into the open channels. This is a promising method for obtaining highly dense silicon carbide ceramic components [20–22], but it is a time consuming and expensive process.

The present work was undertaken to combine the advantages of liquid silicon infiltration (LSI) and CVI to obtain better properties in the final ceramic product. Commercially available cotton fabric has been used to make a laminate with phenolic resin as a binder. This was pyrolysed to get a carbon preform, which was used for obtaining silicon carbide ceramics by

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silicon infiltration. A low density (2.05 g/cm^3) cotton fabric based silicon carbide ceramic produced from LSI was taken for further densification through chemical vapour infiltration. The physical and mechanical properties of the CVI treated and untreated ceramics were compared. Thermal stability of the above ceramics was also studied.

2. Experimental procedure

Phenolic resin powder was purchased from Chemisol Polymer India limited. Commercially available cotton fabric was used. In the initial step, 20% phenolic resin solution was made by dissolving in isopropyl alcohol. Cotton fabric was cut into the dimensions of 200 mm × 200 mm and treated with dilute sodium hydroxide solution to remove the starch coating on the cloth and then dried. Cotton fabric laminate was made by hand lay-up method using phenolic resin as the binder. Cotton fabric was positioned manually in an open mould, resin was poured, and brushed over the fabric. Entrapped air was removed manually with squeeze rollers. This process was repeated for several times until the desired thickness (7 mm) is reached. Subsequently the laminate was kept in an oven at 130 °C for curing. To avoid any delamination, the laminate was submerged again in phenolic resin solution and cured at 130 °C. Carbon preforms were prepared by pyrolysing the cured cotton fabric laminates under N_2 atmosphere in a furnace. A heating rate of 2 °C/min was used up to 600 °C and a heating rate of 5 °C/min was used up to 1100 °C. The specimens were held at this temperature for 2 h. The resulting porous carbon preform was infiltrated with liquid silicon in a vacuum graphite furnace at 1600 °C under argon atmosphere. The specimens were kept at this temperature for 2 h to allow complete reaction of silicon with carbon to form SiC.

The density of the Si/SiC ceramics varied from 2.05 to 2.58 g/cm^3. The low density (2.05 g/cm^3) ceramic was used for isothermal CVI treatment because of the ease of reactant gas infiltration. Four-point bend test specimens were prepared according to the ASTM C 1161 standard. These specimens were subjected to the CVI treatment for 40, 60 and 80 h. A minimum of three samples was used for every CVI treatment. The SiC ceramic samples were placed in an ICVI reactor (M/s Archer Technicoat Ltd., UK) chamber and SiC infiltration was carried out using the methyltrichlorosilane (MTS, 99%, M/s. Spectrochem, India) and H_2 (99.995%, M/s. Inox air products Ltd., India) precursor system with the fixed pressure of 1.65 mbar. In order to avoid the formation of excess free silicon and carbon, the MTS/H_2 molar flow rate ratio was maintained at 1:16 (MTS = 700 g/h, H_2 = 28 SLM). The reactor was evacuated and heated at slow rate (3– 4 °C/min) with Ar purging. Once the temperature reached the set value (950–1100 °C), it was stabilised for 10–20 min. MTS precursor gas was passed through a liquid mass flow meter (M/s. Bronkhorst, Netherlands), vapourised in the evaporator and mixed with H_2 in the mixing chamber. The MTS/H_2 mixture entered the reactor and formed SiC in and around the samples.

3. Characterisation

The X-ray diffraction (XRD) patterns of CVI treated and untreated SiC ceramics and heat treated samples were recorded using X-ray diffractometer (Bruker, Discover D8) with nickel filtered Cu Kα radiation. The microstructures were observed with scanning electron microscope (SEM) (FEI, QUANTA 200) operated at 30 kV and 20 mA and with an optical microscope. Density was determined by the Archimedes method. The thermal characteristics of CVI treated and untreated cotton fabric based SiC ceramics were studied by thermogravimetric analysis (TGA) at a heating rate of 10 °C/min from room temperature to 1200 °C under atmospheric conditions (Netzsch, STA 409C).

The flexural strength of infiltrated and non-infiltrated SiC ceramics was measured by four-point bend test according to ASTM C 1161 standard. Four-point bend testing was carried out using an Instron 4301 universal testing machine with a span of 50 mm and a crosshead speed of 0.5 mm min^{-1}.

Hardness was measured with the help of Vickers hardness tester (Matsuzawa co. ltd, Japan, MMT-7). Vickers hardness of the CVI treated and untreated samples was determined according to the ASTM C 1327-03 standard.

To determine the thermal stability of the above CVI treated (80 h) and untreated ceramics, the flexural specimens (prepared according to the ASTM C 1161) were exposed to 600, 1200 and 1400 °C for 2 h under atmospheric condition. After thermal exposure, these samples were tested for flexural strength. A minimum of three samples has been treated at each temperature and then the flexural strength was determined.

4. Results and discussion

CVI treatment leads to significant improvement in density. Fig. 1 shows the variation of density with infiltration time. The density increases with increasing infiltration time. However there is very little improvement in density after 60 h. During CVI treatment, the deposition of SiC mainly depends on the diffusion of the gaseous reactant species into the pores. The diffusion rate is decreased when the pore size is reduced. As a result, it is very difficult to completely infiltrate the pores. For the large pores, the bottleneck effect hampers the densification. Therefore, beyond 60 h, the infiltration of reactant gases may

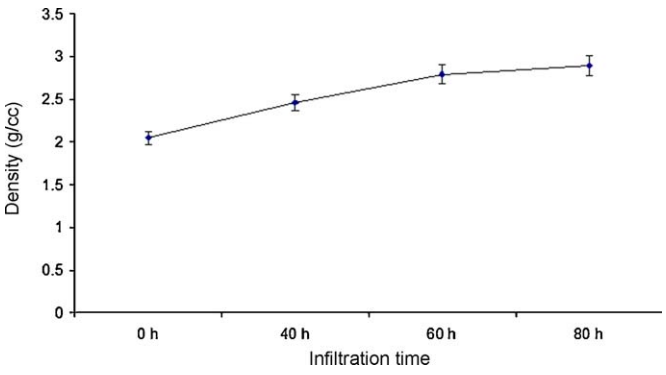

Fig. 1. Variation of density with infiltration time.