



Physical, thermal and ablative performance of CVI densified urethane-mimetic SiC preforms containing in situ grown Si₃N₄ whiskers



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ABSTRACT

A two-step process has been developed for silicon carbide (SiC) coated polyurethane mimetic SiC preform containing silicon nitride (Si₃N₄) whiskers. SiC/Si₃N₄ preforms were prepared by pyrolysis/siliconization treatment at 1600 °C, of powder compacts containing rigid polyurethane, novolac and Si, forming a porous body with in situ grown Si₃N₄ whiskers. The properties were controlled by varying Si/C mole ratios such as 1–2.5. After densification using a chemical vapour infiltration, the resulting SiC/Si₃N₄/SiC composites showed excellent oxidation resistance, thermal conductivity of 4.32–6.62 Wm⁻¹ K⁻¹, ablation rate of 2.38 × 10⁻³–3.24 × 10⁻³ g cm⁻² s and a flexural strength 43.12–55.33 MPa for a final density of 1.39–1.62 gcm⁻³. The presence of a Si₃N₄ phase reduced the thermal expansion mismatch resulting in relatively small cracks and well-bonded layers even after ablation testing. This innovative two-step processing can provide opportunities for expanded design for using SiC/Si₃N₄/SiC composites being lightweight, inexpensive, homogeneous and isotropic for various high temperature applications.

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1. Introduction

Silicon carbide (SiC) based composites have become a prime candidate material for thermal protection systems and high-temperature heat exchange applications, primarily because of their thermal stability above 1000 °C, high specific strengths, sufficient thermal conductivity, ablation resistances, corrosion and oxidation resistances at high temperatures [1–3]. Since the invention of Acheson method, many processes have been developed including hot isostatic pressing, reaction bonding/reaction forming, polymer pyrolysis/chemical vapour deposition and bio-templating technique etc. [4–6]. As compared to conventional methods, innovative bio-templating SiC is cheap, lightweight, porous, and strong in compression and flexural loading [7]. They constitute a new class of non-oxide ceramics synthesised in two-steps; reactive infiltration of molten Si into a carbon preform obtained from pyrolysis of wood, bamboo, cotton fabric, lotus root, hulls or fibreboards etc. [8–12]. However, anisotropic properties, non-uniform density distribution, existence of flaws, and uncontrollable mechanical

properties are the associated features of this method [13]. Medium density fibreboard and paper have better control of pore structure and properties but they are two-directional in nature [14,15]. Using a simple method to prepare isotropically homogeneous SiC composite ceramics was the main motive for this investigation. The current work has been divided into two steps. The first step is to present a new polyurethane-mimetic process for the preparation of SiC preforms in only one-step with the advantages of industrially manufactured products being isotropic and reproducible with reference to the desired composition, density, porosity or strength etc. It is supposed that the excellent properties are also related with the novolac resin used as a carbonaceous binder [16,17]. Furthermore, the carbon sources and gradual porosity creation by the pyrolysis of polyurethane and novolac resin contributed to the formation of expected one-dimensional nano phases, such as Si₃N₄ whiskers (in this case) and SiC nanowires in previous studies by our group [18–20]. The results showed that different porosities could be obtained using this method by varying the amount of Si. After ensuring isotropic compressive strength depending on density and the amount of Si₃N₄ whiskers, the preforms were further densified with additional SiC matrix in a second step. During this step, a chemical vapour infiltration (CVI) process was used to coat fine gain SiC matrix in the pores and around Si₃N₄ whiskers. This step was undertaken to combine the advantages of in situ grown

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Table 1
Processing parameters of polyurethane-mimetic SiC/Si₃N₄ preforms.

ID	PUMT (g)	Si (g)	Density (g/cm ³)		
			Curing (p _{230 °C})	Siliconization (p _{1600 °C})	Etching (p _{24h})
SiC/Si ₃ N ₄ –1.0	10.00	6.50	0.78	0.89	0.89
SiC/Si ₃ N ₄ –1.5	10.00	9.75	0.94	1.10	1.10
SiC/Si ₃ N ₄ –2.0	10.00	13.00	1.17	1.36	1.35
SiC/Si ₃ N ₄ –2.5	10.00	16.25	1.39	1.48	1.46

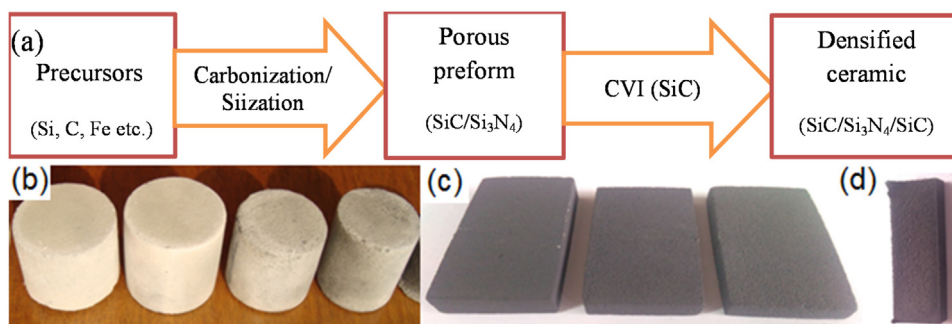


Fig. 1. A two-step processing of SiC/Si₃N₄/SiC ceramics; (a) process flow diagram, (b) SiC/Si₃N₄ preforms, (c) SiC/Si₃N₄/SiC ceramics and (d) cross section of SiC/Si₃N₄/SiC-2.5 ceramic with a gradient coating.

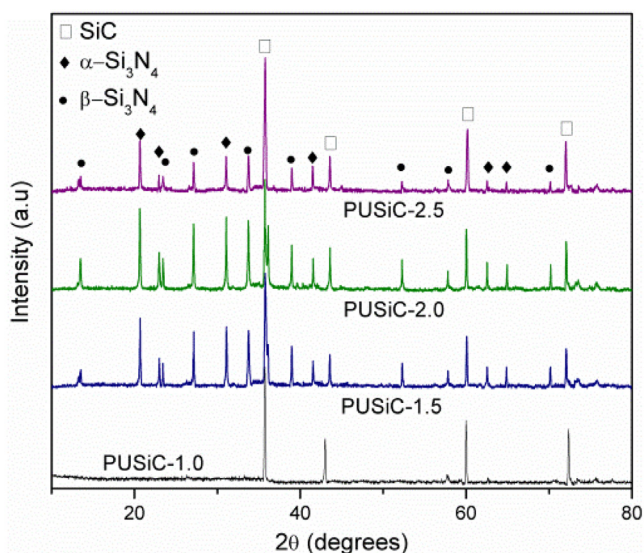


Fig. 2. XRD profile of SiC/Si₃N₄ preforms containing Si₃N₄ whiskers.

Si₃N₄ whiskers in the preform to be densified, forming adjacent carbide/nitride/carbide phases to obtain better control of properties in the final ceramics. It has already been established that the SiC–Si₃N₄ ceramics possess the highest oxidation resistance among all the non-oxide ceramics due to the formation of the lowest permeable SiO₂ adherent film [21,22]. In our case the pores of SiC/Si₃N₄ preform were homogeneously covered with in situ grown Si₃N₄ whiskers and after coating with SiC using a CVI process, the resulting composites were designated as silicon carbide/silicon nitride/silicon carbide (SiC/Si₃N₄/SiC). In this paper, these two steps have been investigated with a special focus on the effect of processing parameters on the resulting microstructures, physical, thermal and ablative properties. This method is attractive in comparison with other processes where nano reinforcements are added in additional processing steps with the associated problems of inhomogeneous microstructure and dispersion.

2. Experimental

2.1. Step-one: preparation of SiC/Si₃N₄ preforms

The first step in the processing of SiC/Si₃N₄/SiC composites was to prepare porous SiC/Si₃N₄ preforms reinforced with short carbon fibres (SCFs) and in situ grown Si₃N₄ whiskers. Rigid-polyurethane (RPU) foam, novolac resin, SCFs, Si powder, and FeCl₃ were purchased from Sinopharm Chemicals, Beijing China. The bulk density of RPU was $4.3 \times 10^{-1} \text{ g cm}^{-3}$, with the monomers of methylene diphenyl diisocyanate and polyol of ~300 molecular weight. According to the manufacturer specifications, the elemental composition was 58.9 wt.% C, 6.9 wt.% H, 8.1 wt.% N and 26.1 wt.% others including oxygen. The RPU was cut into smaller pieces and ground by a grinding machine until the powder passed a 150-mesh screen (particle sizes of RPU $\leq 105 \mu\text{m}$). A batch of mixture containing 60 wt.% RPU, 30 wt.% novolac (150-mesh, and particle size $\leq 105 \mu\text{m}$), 5 wt.% SCFs and FeCl₃ each was prepared and ball milled in a tungsten carbide jar for about 4 h using a QM-1SP2 planetary-type ball mill. The objective was to further reduce the particle sizes and make an intimate contact of all powdered constituents thereafter called polyurethane-mimetic template (PUMT). After ball milling, screening was not performed as most of the particles of RPU, novolac and FeCl₃ were embedded in each other. Carbon yield of PUMT as determined by TGA experiment came out to be ~28 wt.% (Fig. S1). To produce SiC/Si₃N₄ preforms with varying density, stoichiometric ($\text{Si}_{(28.0855 \text{ g/mol})} + \text{C}_{(12.0107 \text{ g/mol})} \rightarrow \text{SiC}_{(40.09 \text{ g/mol})}$) and excess amount of Si was added to PUMT as given in Table 1. The amount of Si relative to the expected yield of C from PUMT was varied in the range of 1–2.5 of Si: C mole ratio, i.e., from stoichiometric to over-stoichiometric. Excess Si was used to obtain porous SiC ceramic with a higher density. After ensuring complete mixing of four batches, the powders were put in a graphite mold fitted with plungers, pressed using a press (3.5 MPa) and cured using a curing cycle of 120 °C/2 h + 150 °C/2 h + 180 °C/2 h + 110 °C/2 h + 230 °C/4 h. For pyrolysis/siliconization treatment, a tubular alumina reactor heated in a resistance furnace was used. After demolding, the green compacts were first heated under N₂ flow with a slow heating ramp of 2 °C/min up to 600 °C and kept constant for 2 h. After ensuring evaluation of maximum volatiles, the samples were heated with

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