Effect of pyrocarbon content in C/C preforms on microstructure and mechanical properties of the C/C–SiC composites

Yonghui Zhang*, Zhichao Xiao, Jiping Wang, Jianfeng Yang, Zhihao Jin

State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, No. 28 Xianning West Road, Xi'an Shaanxi 710049, People's Republic of China

ARTICLE INFO

ABSTRACT

Carbon fiber reinforced silicon carbide matrix (C/C–SiC) composites were prepared by liquid silicon infiltration (LSI) process, using four kinds of quasi-three-dimensional C/C preforms with the same fiber volume fraction (32%) but different pyrocarbon content (from ~20% to ~50%, in volume) filled by chemical vapor infiltration (CVI). The microstructure and mechanical properties of C/C–SiC composites have been investigated. X-ray diffractmeter (XRD) analysis results show the composites are composed of three phases of β-SiC, C and Si. With the increase of the pyrocarbon content in the C/C preforms, the content of formed silicon carbide in the C/C–SiC composites decreases but the residual pyrocarbon increases. Mechanical tests were performed to assess the role of the matrix and phase composition in the C/C–SiC composites. The results demonstrate that the flexural strength and shear strength of the composites increase gradually as the increase of the content of residual pyrocarbon. The flexural and shear strength of the C/C–SiC composite produced of the C/C preform with pyrocarbon content of ~42 vol.% reached the highest value.

1. Introduction

Carbon fiber reinforced silicon carbide matrix (C/C–SiC) composites have been attracted an increasing number of applications in the aerospace industry as well as many other fields, due to their superior mechanical properties, oxidation resistance and abrasive properties [1–4]. Different processing techniques are currently in use for the manufacture of complex shaped ceramic matrix composites (CMCs) components. A novel technology developed by the German Aerospace Centre (DLR) to produce CMCs structures with lower costs and shorter manufacturing times is the liquid silicon infiltration (LSI) method [5,6], which includes a two-step process starting with the fabrication of the porous carbon/carbon (C/C) preform and concluding with LSI to form final C/C–SiC composites.

It is well known that the properties of CMCs are determined not only by the fibers and matrix themselves, but also mainly by the microstructure of the matrix and the properties of the fiber–matrix interface [7–9]. Thus, the manufacturing method has a great influence on the composite properties. As the porous C/C composite preform is key intermediate material for preparing the C/C–SiC composites by LSI process, the internal or external factors of the C/C composite preform such as the microstructural evolution or the matrix porosity have great effects on the performance of the C/C–SiC composites, while few publications are available which try to elucidate the effect of these factors of C/C preforms on the C/C–SiC composites [10].

Generally, the preparation of C/C composites involves the liquid polymer impregnation (LPI) method and chemical vapor infiltration (CVI) method. The C/C composites prepared by LPI process usually result in matrix-cracking, consequently the deteriorated performance of the final C/C–SiC composites. The C/C composites prepared by CVI process have a good matrix quality and controlled microstructure, so it is possible to obtain the C/C–SiC composites with high performance. Therefore, the aim of the present investigation was to evaluate the effects of carbon content and matrix porosity of C/C preforms prepared by CVI on the microstructure and mechanical properties of C/C–SiC composites fabricated by the LSI process. Correlations of matrix volume fraction, microstructure, and mechanical behaviors are discussed.

2. Experimental procedures

2.1. Materials

Polyacrylonitrile (PAN)-based fibers were used as the reinforcement for fabrication of the C/C preforms. Propylene was applied as the matrix precursor for the C/C preforms. Industrial silicon powder with a diameter range of 0.5–3 mm was used as the silicon source during LSI process.

* Corresponding author. Tel.: +86 29 8266 7942; fax: +86 29 8266 5443.
  E-mail address: huihui0922@gmail.com (Y. Zhang).

Table 1
The density and porosity of the C/C performs and C/C–SiC composites.

Sample	C/C preforms		Sample	C/C–SiC composites	
	Density (g/cm³)	Porosity (%)		Density (g/cm³)	Porosity (%)
M1	1.02	45.9	S1	2.42	2.3
M2	1.32	32.4	S2	2.30	2.8
M3	1.46	25.3	S3	2.15	4.0
M4	1.62	13.5	S4	1.98	4.8

2.2. Preparation of samples

At first, quasi-three-dimensional fiber preform was prepared by needling method. The volume fraction of carbon fiber was 32%. The porous C/C preforms with different pyrocarbon content (20–50 vol.%) were fabricated by CVI process for different infiltration time of 100–500 h; then the C/C–SiC composites were fabricated by the LSI (infiltration of the liquid silicon into the porous C/C preform) process, which was conducted at 1650 °C under vacuum.

2.3. Measurement

Mechanical properties of the C/C–SiC composites were investigated under compressive, bending and shear loading. The samples for measurement were cut from the original C/C–SiC bulk materials. The sample sizes were 10 mm × 10 mm × 10 mm for compressive test, 10 mm × 6 mm × 50 mm for bending test, 30 mm × 2.5 mm × 3.5 mm for inter-laminar shear test and Φ50 mm/Φ10.5 mm × 4 mm for in-plane shear test. All the measurements were performed on an Instron 1195 test machine.

The open porosity and bulk density of the samples were measured by the Archimedes method. The X-ray diffractmeter (XRD) patterns of the C/C–SiC composites were recorded using XRD (model D/max-rA, Rigaku, Japan) with nickel filtered Cu Kα radiation produced at 35 kV and 20 mA. The phase composition of the composites was calculated taking into account the bulk density of the C/C preform and of the C/C–SiC composite as well as the volume fraction of free silicon in the composites determined by image analysis. The microstructure morphology and fracture surfaces were observed by an optical microscope and a scanning electron microscope (SEM, model S-2700, Hitachi, Japan) operated at 20 kV and 20 mA.
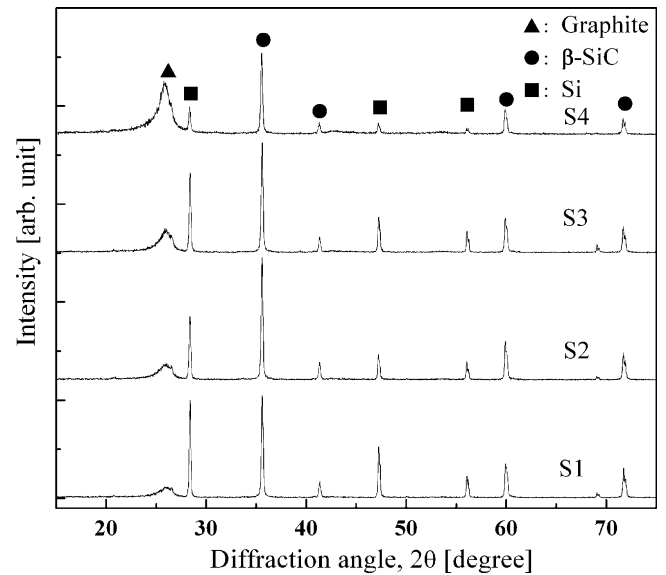
3. Results and discussion

3.1. Density and porosity of the C/C preforms and C/C–SiC composites

Table 1 lists the bulk density and open porosity of the C/C preforms and C/C–SiC composites. Samples M and S represent the C/C preforms and the C/C–SiC composites, respectively (the same as following). It can be seen that with increasing the density of the C/C preforms, the density of the C/C–SiC composites decreases and its open porosity increases. It shows that the melt silicon into composites was retarded by the increase of the matrix carbon, and the

Fig. 1. XRD patterns of the C/C–SiC composites.

infiltration process of the composites became increasingly difficult by the increasing density of C/C preforms in the densification process.

3.2. Phases in matrix of C/C–SiC composites analysis

The XRD phase analysis result of the C/C–SiC composites is shown in Fig. 1. It reveals that there are silicon carbide, graphitic carbon and some unreacted silicon in the C/C–SiC composites. The broad carbon-peak is associated to the carbon fibers and unreacted pyrocarbon. The main phase of the silicon carbide is a face-centered cubic (fcc; β) type SiC.

Fig. 2 shows the typical optical micrographs of the C/C–SiC composites. The phase composition of the composites is predominantly determined by the bulk density, structure and infiltration behavior of the C/C preforms [11]. In the region of the sparse fibers, local C/C composites were formed by CVI carbon matrix around the carbon fibers, and wrapped by SiC (the light grey region in Fig. 2(c) and (d)), which was generated by siliconizing. The macrospores were filled with residual silicon (the white region in Fig. 2(c) and (d)). However, composite S1 as shown in Fig. 2(a) exhibited more regions with silicon and silicon carbide than that of composite S2 (Fig. 2(b)). Table 2 shows that the residual silicon content in the C/C–SiC composites decreases from 24% to 7% and the formed silicon carbide content decreases from 40% to 8% with increasing the bulk density of C/C preforms, while the residual carbon content in the C/C–SiC composites increases from 3% to 50% with the increase of the pyrocarbon content in the C/C preforms.

It is known that the morphology, size and content of the pores in the composites have significant influences on densification behavior during the LSI process. Since the C/C performs in this study are different with the bulk density and the matrix porosity. The LSI

Table 2
Phase contents of the C/C preforms and C/C–SiC composites.

Sample	C/C preforms		Sample	C/C–SiC composites		
	Carbon fiber (vol.%)	Pyrocarbon (vol.%)		Silicon carbide (vol.%)	Residual pyrocarbon (vol.%)	Residual silicon (vol.%)
M1	32	22	S1	40	3	24
M2	32	36	S2	30	21	15
M3	32	43	S3	18	34	13
M4	32	55	S4	8	50	7