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Effect of carbon nanotubes electrophoretically-deposited on reinforcing carbon fibers on the strength and toughness of C/SiC composites

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ABSTRACT

The present work reports on the strength and toughness enhancement due to carbon nanotube (CNT) presence in silicon carbide–matrix composites reinforced by CNT-coated carbon fibers (CNT–C/SiC). The composites were prepared via a chemical vapor infiltration/electrophoretic deposition methodology which enables CNT-coating of reinforcing fibers at varying load fractions. Both the tensile strength and toughness were found to first increase and then decrease with CNT electrophoretic deposition time, hence also CNT loading. The optimized deposition time turned out to be 10 min where excellent mechanical properties were obtained and for instance, tensile strength and work of fracture reached their maximum of 392 MPa and 1869.4 kJ/m^3, respectively. Scanning electron microscopy observations of CNT–C/SiC fracture surfaces showed that the magnitude of the fiber pull-out phenomenon increased significantly compared with pristine C/SiC. Abundant CNT pull-out within the CNT-rich layers around the fibers acted complementary to carbon fiber pull-out and was identified as the main toughening mechanism.

1. Introduction

The use of high performance fiber reinforced composites, such as the carbon fiber/silicon carbide (C/SiC) family, in aerospace thermal protection and advanced friction applications has expanded and still expands substantially due to the excellent thermomechanical behavior of these materials and the ease of customizing fiber preform design to meet application requirements [1,2]. Such materials are used today in gas turbines, engines, space shuttles and nuclear reactors [3], brake materials for shipboard aircraft [4–7] and will be used in the future in thermal protection systems in reusable launch vehicles materials [8]. Although C/SiC composites are less brittle and more reliable than single-phase ceramics [9], their relatively low compressive and torsional performance still hampers the expansion of their applicability.

Carbon nanotube (CNT) is a relatively new type of nano-structured material [10] which has attracted much attention as potential reinforcement elements for structural composites due to their exotic mechanical, thermal, and electrical properties [10–12]. For example, CNTs have been used to reinforce alumina composites fabricated by hot-pressing [13] as well as aluminum matrix composites [14] and the enhancement in mechanical properties was investigated. Recently, multi-walled carbon nanotubes (MWCNTs) reinforced C/SiC composites were prepared via a polymer infiltration and pyrolysis (PIP) process; the addition of 1.5 wt% MWCNTs in C/SiC composites led to a 29.7% increase in the flexural strength, and a 27.9% increase in the fracture toughness [15]. The main problem of CNT usage in composites is associated with their natural tendency to agglomerate considerably as a result of their large specific surface areas and aspect ratios, a problem that renders their uniform dispersion in the matrix a difficult task which must be overcome in order to achieve improved material performance [16]. One method for effectively and uniformly dispersing CNTs in aqueous solutions as a step for preparing composites is electrophoretic deposition (EPD). A relevant review which gives an overview of the EPD methodology for the fabrication of CNT-reinforced ceramic matrix composites, including SiO_2/CNT, TiO_2/CNT, MnO_2/CNT, and Fe_3O_4/CNT was presented in [17]. In another review [18], the EPD mechanism is analyzed along with discussions on the role of CNT coatings and CNT/nanoparticle composite films. Firm and homogenous CNT-coatings on SiC fibers were recently reported for CNT–SiC-fiber-reinforced SiC matrix composites (CNT–SiC/SiC) fabricated by the EPD process [19]. The process of preparing C/SiC composites by CVI has been quite mature, but the EPD of CNTs to the surface of carbon fiber and then SiC matrix was infiltrated via CVI to prepare CNT–C/SiC has not yet been fully investigated.

In this study, an EPD process is used to promote CNT deposition on the carbon fibers. CNT–C/SiC composites were prepared by CVI

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of SiC into preforms of CNT-coated carbon fibers and tested in monotonic tension. Composite strength and work of fracture were obtained for composites with varying amounts of CNT coatings and compared to their counterparts from pure C/SiC composite tests.

2. Experimental

The T-300™ carbon fiber used in the current experiment was provided by the Toray Company, Japan. A commercial MWCNTs (hereinafter referred to as CNTs) aqueous solution (5 wt%) was provided from Chengdu Institute of Organic Chemistry, Chinese Academy of Sciences. The surfactant used for CNTs aqueous solution was TNWDIS, which was actually aromatic modified polyethyleneglycol ether. And the quantity of the TNWDIS was 20% the weight of CNTs. The CNT diameter is *ca.* 8–15 nm and the length is *ca.* 50 μm. Uniformly-dispersed aqueous CNT solutions were obtained by diluting the as-received solution with deionized water to 0.05 wt% and subsequent ultrasonication of the diluted solution for 30 min at 1500 Watt in an ultrasonic emulsifying dispersion machine (BILON-1500, BILON Biological Technology Co., Ltd., Shanghai, China). Prior to the EPD process, a *ca.* 100 nm thick layer of pyrolytic carbon layer (PyC) was deposited on the fiber surface via chemical vapor deposition with propylene as gas source.

The EPD process involved fixing the PyC-coated carbon fiber on the graphite frame as positive electrode ($120 \times 80 \times 4\ mm^3$), then immersing the electrode in the 0.05 wt% CNT aqueous solution. The voltage between the electrode plates was set to 15 V DC and electrode spacing was *ca.* 3 cm. Four different deposition times were used: 5, 8, 10, and 15 min, which produced reinforcing fibers with increasing loading of CNTs on their surface. Laser confocal Raman microscopy (RMS, Renishaw, UK) equipped with a He–Ne laser ($\lambda = 514.5$ nm) was employed to characterize the graphitization degree of CNT-coated carbon fibers. Following CNT deposition, CNT–C reinforcing preforms were prepared and CVI was used to infiltrate the SiC matrix into the preforms to obtain the CNT–C/SiC mini-composites, which have a *ca.* 80 mm length with a radius of 0.5–0.6 mm. The area of C/SiC composites was obtained by weight measuring method [20], not use the formula ($\pi \times R \times R$) to calculate directly, because the C/SiC composites were not dense.

The composites were subjected to monotonic tensile testing to failure and the strength and toughness of C/SiC and CNT–C/SiC composites with varying CNT loading were compared in order to identify improvements related to CNT presence. Testing was performed on a universal testing machine (Instron 3345, Instron Ltd., High Wycombe, England) equipped with a 1 kN load cell. The gauge length of specimens for tensile testing was 50 mm. Corresponding to each EPD time a group of 10 composite specimens were tested. The diagrammatic sketch of tensile test is shown in Fig. 1. All tests were performed under crosshead displacement control at a rate of 0.2 mm/min with a ±0.5% precision. With the gauge length of specimens 50 mm, the displacement is *ca.* 0.3–0.4 mm (gauge length multiplied the strain). Then effective loading time which is defined as displacement divided by crosshead speed (0.2 mm/min) is calculated to be *ca.* 120 s. In the current work, work of fracture was obtained by integration of stress–strain curve data for all specimens (value is equal to the area surrounded by

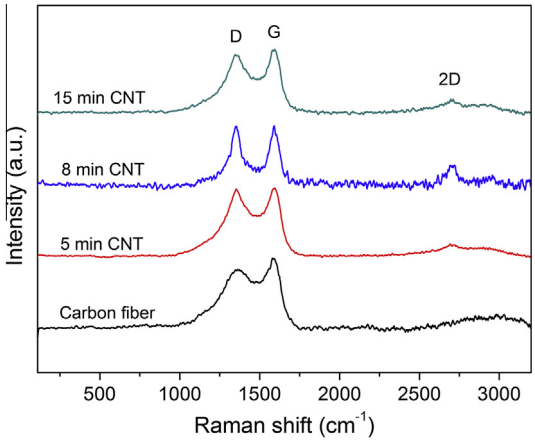

Fig. 2. Raman spectra of T300 carbon fiber with the EPD times of 0, 5, 8, and 15 min.

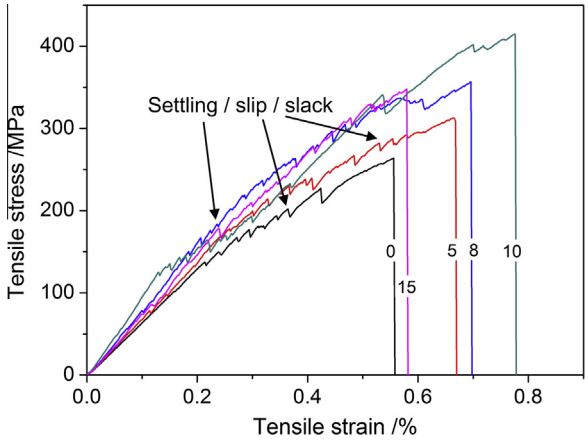

Fig. 3. The typical stress–strain curves of C/SiC mini-composites with different CNT EPD time (0, 5, 8, 10, and 15 min).

Table 1
Statistical results of tensile stress, failure strain, Young's modulus, and work of fracture of C/SiC with different EPD times of CNTs.

Deposition time (min)	Tensile stress (MPa)	Failure strain (%)	Young's modulus (GPa)	Work of fracture (kJ/m^3)
0	270 ± 40	0.60 ± 0.11	55.8 ± 8.8	846.8 ± 150.7
5	299 ± 20	0.66 ± 0.13	68.7 ± 7.5	1265.0 ± 100.5
8	376 ± 30	0.73 ± 0.10	81.2 ± 10.9	1546.7 ± 160.2
10	392 ± 32	0.79 ± 0.14	95.2 ± 6.8	1869.4 ± 156.6
15	348 ± 36	0.61 ± 0.09	75.5 ± 6.3	1120.3 ± 121.8

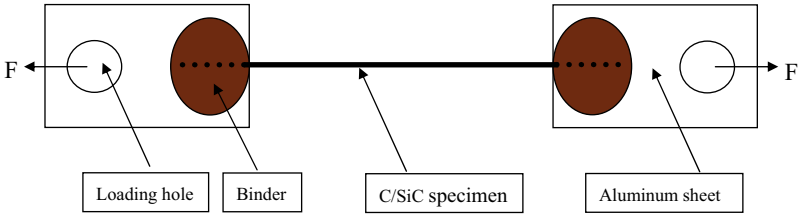

Fig. 1. Diagrammatic sketch of tensile test of mini C/SiC composites.