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Highly conductive and high-strength carbon nanotube film reinforced silicon carbide composites

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ABSTRACT

Film formed by carbon nanotubes is usually called carbon nanotube film (CNT_f). In the present study, CNT_f fabricated by floating catalyst method was used to prepare CNT_f/SiC ceramic matrix composites by chemical vapor infiltration (CVI). Mechanical and electrical properties of the resulting CNT_f/SiC composites with different CVI cycles were investigated and discussed, and the results revealed that the CNT_f has a good adaptability to CVI method. Tensile test demonstrated an excellent mechanical performance of the composites with highest tensile strength of 646 MPa after 2 CVI cycles, and the strength has a decline after 3 CVI cycles for an excessively dense matrix. While, the elastic modulus of the composite increased with the CVI cycles and reached 301 GPa after 3 CVI cycles. Tensile fracture morphologies of the composites were analyzed by scanning electron microscope to study the performance change laws with the CVI cycles. With SiC ceramic matrix infiltrated into the CNT_f, enhanced electrical conductivity of the CNT_f/SiC composite compared to pure CNT_f was also obtained, from 368 S/cm to 588 S/cm. Conductivity of the SiC matrix with free carbon forming in the CVI process was considered as the reason.

1. Introduction

One-dimensional nanoreinforcements, such as carbon nanotubes (CNT) [1–3] and silicon carbide nanofibres or nanowires [4,5], with outstanding thermal, electrical and mechanical properties are currently considered as high-potential nanoreinforcements for ceramic matrixes to prepare novel ceramic matrix nanocomposites with satisfied mechanical performance and tailored thermal and electrical properties. Despite many theoretical simulations predicting superior performances of CNT reinforced ceramic composites, there are few studies reporting significant properties improvements by introducing CNT into ceramic matrixes, and the main reason is the restriction of its preparation methods. Two-step method is primary method to prepare CNT reinforced ceramic composite, it requires dispersing CNT with ceramic particles and subsequent sintering, and it has been used to prepare composite with varying ceramic matrixes, such as Al_2O_3, ZrO_2, and Si_3N_4 [6–8]. With high surface area and high aspect ratio, CNT tend to spontaneously agglomerate, bundle together and entangle [9], which pose a big challenge to two-step method for that it is very hard to dispersing CNT throughout ceramic matrix uniformly. Typical CNT dispersion techniques in ceramics include powder mixing, colloidal processing, sol–gel processing and in-situ growth of CNT on ceramic grains [10–13]. While, with these dispersion techniques, it is also very difficult to prepare ceramic composites with high CNT content. In addition, the subsequent sintering process with high temperature and necessary pressure will damage the CNT in the ceramic matrix [14], which lead to a degradation of the properties of the composites.

With the above-mentioned problems, infiltrating CNT assemblies, such as CNT arrays, CNT fibers, CNT films (CNT_f), CNT buckypaper [15–20], with ceramic matrices is the preparation-tendency of CNT reinforced composites. The challenging problem of dispersing CNT in the matrices uniformly can be avoided with this method, so it is suitable to fabricate nanocomposites with uniform CNT dispersion and high loadings. CNT_f is a two dimensional network composing of continuously and uniformly interweaved CNTs, and the volume fraction of CNTs is very high, which resulted in good thermal, electrical and mechanical properties of the CNT_f [17,19]. With excellent properties, CNT_f is commonly used to prepare CNT reinforced polymer or ceramic matrix composites [20,21]. Polymer derived ceramic (PDC) method was used to prepare CNT film reinforced SiCN composite [21], the resultant nanocomposites contained a high volume fraction of CNTs (60 vol%), and reached an electrical conductivity of up to 2.2×10^5 S/m. SiC matrix was also introduced into CNT_f by PDC method [22], and the composite showed well thermal resistant, mechanical and electrical properties. Chemical vapor infiltration (CVI) method was also commonly used to introduce ceramic matrices into CNT assemblies, such as

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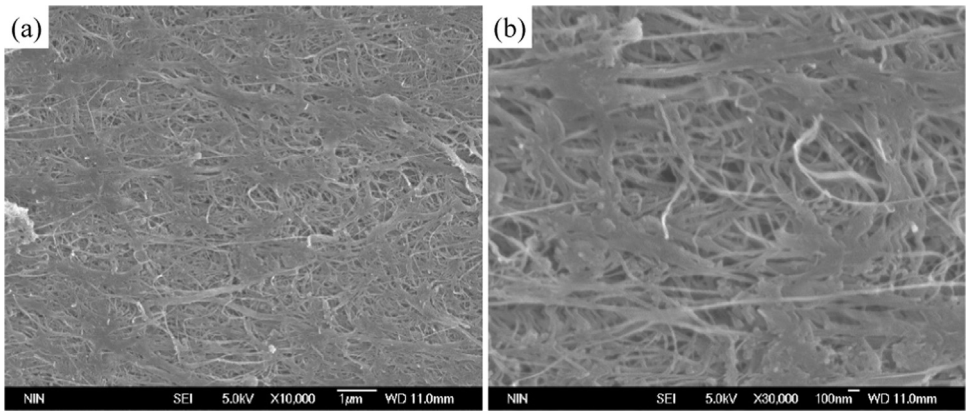

Fig. 1. Microstructure of the CNT_f prepared by floating catalyst method.

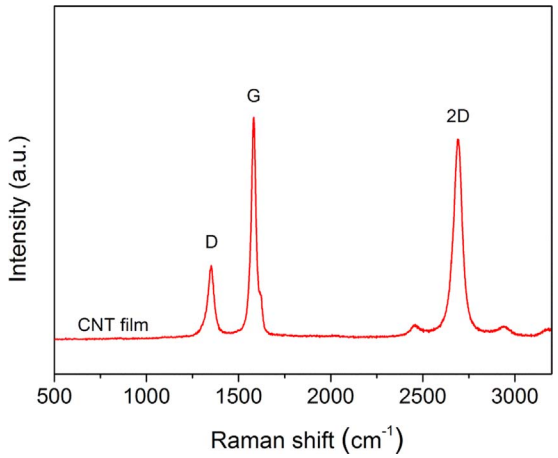

Fig. 2. Raman spectrum of the CNT_f prepared by floating catalyst method.

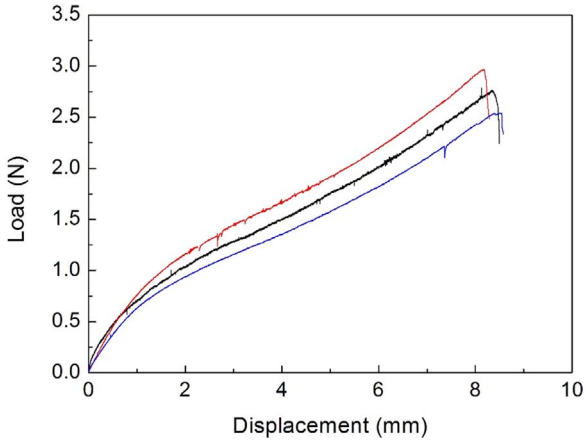

Fig. 3. Load-displacement curves of pure CNT_f.

CNT array [23], while, with micro architectures of the obtained nanocomposite, it is difficult to test its macro-mechanical and electrical performances. CNT film combined with CVI method is potential to prepare large-scale and high-performance composites, however, it was not found in available literature.

In the present paper, CNT_f fabricated by floating catalyst method were used to prepare CNT_f/SiC composite cooperated with CVI. Compared to the conventional CNT-reinforced ceramic composites, CNT_f/SiC composites can reach impressively high CNT loadings and significantly improved CNT dispersions. Meanwhile, CVI method is ideal for not damaging the CNTs structure. Content of SiC matrix in the composites was controlled by CVI cycles, and the effect of the CVI cycles on fracture morphology, mechanical and electrical performances were investigated and discussed in the text.

Table 1
Statistical results in tensile strength, elastic modulus and electrical conductivity of CNT_f/SiC with different CVI cycles.

CVI cycles	Tensile strength (MPa)	Elastic modulus (GPa)	Electrical conductivity (S/cm)
0	65.2 ± 4.6	0.394 ± 0.016	368 ± 21
1	315 ± 28.4	148 ± 12.3	576 ± 25
2	646 ± 36.4	290 ± 14.3	585 ± 34
3	578 ± 31.2	301 ± 16.5	588 ± 36

2. Experimental

Pristine CNT_f used in this work was purchased from Jiangsu Jiedi Nano Technology Co., Ltd., China. It was fabricated by floating catalyst method, and its thickness and area density is about 10 μm and 5.2×10^{-6} g/mm^2 with about 10 wt% Fe catalyst. The CNTs in the film are multiwall CNTs, with the diameter of 10–20 nm and length of about 1 mm. Strips of approximate width of 3.5 mm were cut from the as-received CNT_f, they were twined on the graphite frame (120 mm×60 mm), and then were placed into the CVI furnace subsequent infiltration of SiC matrix at ca. 1000 °C via the isothermal process described in [24]. One CVI cycle last for 70 h.

The obtained CNT_f/SiC composites were subjected to monotonic tensile testing to failure and the strength and elastic modulus of the composites with varying CVI cycles were compared. Testing was performed on a universal testing machine (Instron 3345, Instron Ltd., High Wycombe, England) equipped with a 1 kN load cell, and all tests were performed under crosshead displacement control at a rate of 0.02 mm/min with a ± 0.5% precision. The gauge length of specimens for tensile testing was 40 mm. Corresponding to each CVI times, a group of 5 composite specimens were tested. Volume electrical conductivity of the specimens were measured by two-wire method using a Keithley 6220 DC current source (Heithley, Cleveland, USA). Specimens were fixed onto a circuit board and their two ends were connected to the circuit board by copper wires fastened with silver paste. The circuit board was then plugged into the slot of the current source for electrical conductivity measurement with an input current of 1 mA.

Scanning electron microscopy (SEM, Hitachi S-4700, Tokyo, Japan) was employed for the micro-structural characterization of the failed specimens, raman spectra (RMS, Renishaw, UK) equipped with a He-Ne laser (λ=532 nm) was employed to characterize the graphitization degree of the as-received CNT_f, and the phase composition of the composite was examined by X-ray diffraction (XRD, D8-Advance, Bruker, Germany).