J Mater Sci (2007) 42:9498–9500
DOI 10.1007/s10853-007-2049-0

LETTER

Carbon nanotube: carbon composites with matrix derived from oxidized mesophase pitch

Yongzhong Song · Gengtai Zhai · Jinli Shi · Quangui Guo · Lang Liu

Received: 16 July 2007 / Accepted: 23 July 2007 / Published online: 21 August 2007

Due to their nanometer-scale dimensions, high mechanical strength, and very high thermal and electrical conductivity, carbon nanotubes (CNTs) have attracted much attention to be recognized as an excellent material choice for nanocomposites, in which even a very small amount of CNTs can induce significant changes in the material's properties [1–4].

Since carbon and carbon–carbon composites have many advantages, such as excellent mechanical behavior at high temperatures, low reactivity, high heat capacity, and so on, modern advanced technologies make wide use of these materials in different fields, including nuclear reactor walls, rocket nozzles, battery electrodes, and seal and friction materials [5–7]. However, the conventional techniques to prepare carbon and carbon–carbon composites are complicated and costly due to the time-consuming process, such as liquid phase impregnation and chemical vapor deposition which are indispensable to obtain high-performance carbon and carbon–carbon composites. Much effort has been taken to develop new methods to simplify the processing and shorten the time of preparing carbon–carbon composite. Most significantly, Naphthalene-derived synthetic mesophase pitch (MP) have been recognized as an excellent precursor to the high-performance carbon and graphite due to their high carbon yield [8–11].

Y. Song · G. Zhai · J. Shi · Q. Guo · L. Liu (✉)
Key Laboratory of Carbon Materials, Institute of Coal Chemistry, Chinese Academy of Sciences,
Taiyuan 030001, China
e-mail: liulang@public.ty.sx.cn

Y. Song
Graduate School of Chinese Academy of Sciences,
Beijing 100039, China

However, few researchers try to fabricate the MP based composites reinforced with CNTs because of the difficulties in homogeneously distributing CNTs in a MP matrix by traditional methods. In this communication, CNTs reinforced oxidized MP based composites were prepared by one-step self-sintering. The influence of CNTs on the mechanical and physical properties on obtained CNTs/carbon derived from oxidized MP composites.

The CNTs (multi-walled carbon nanotubes, purity 99 wt.%, diameter 20–40 nm) synthesized by chemical vapor deposition were supplied from Wuhan University of Science and Technology, China. MP was obtained by Mitsubishi Gas Chemical Co., Japan. MWNTs were sonicated in a bath containing toluene, and then the milled MP was added to the suspension and hot-mixed at 333 K in the other bath containing toluene under moderate stirring for 2 h. Subsequently, the system was moved to a plate and dried at 373 K. The dried compound was ground by a ball-mill into grains of particle size in the range of 2.8–8.2 μm. Then the grains were heated to 433 K at a rate of 3 K/min, and then heated to 473 K at the rate of 0.5 K/min, finally the temperature was kept for 60 min in air. The oxidized powders were pressed under 100 MPa in quadrate dies for the measurement of physical properties. The molded tapes were carbonized at different temperature from 1,200 °C for 1 h with a heating rate of 1 °C/min in a nitrogen atmosphere. Further the graphitization treatment was performed in argon at temperature up to 2,800 °C with a heating rate of 10–20 °C/min. In order to make comparisons, oxidized MP powders were also molded into tapes for the measurement of physical properties.

Figure 1 presents a scanning electron microscopy (SEM) image of the composites with 5 wt.% CNTs (MP-C05), where no aggregation of CNTs can be seen. The excellent dispersion is achieved by using sonication and

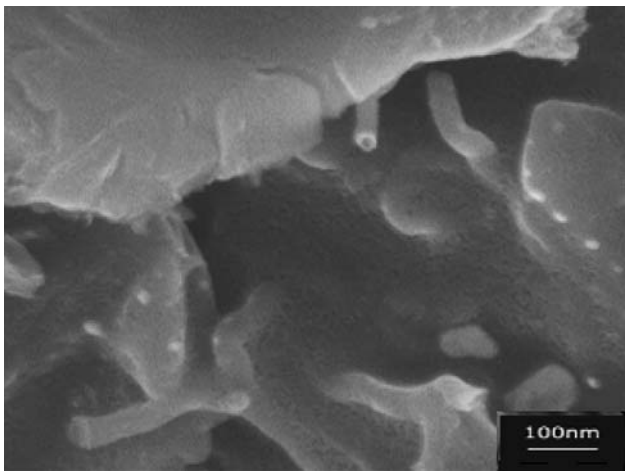

Fig. 1 Scanning electron microscopy (SEM) image of a composite with 5 wt.% carbon nanotubes (CNTs)

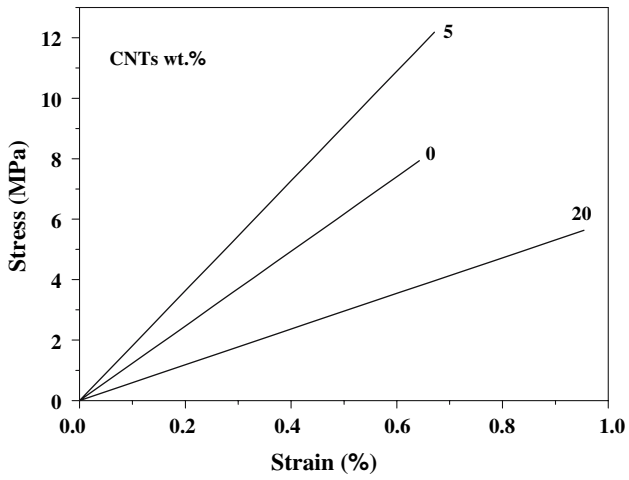

Fig. 3 Stress–strain profiles of mesophase pitch (MP) and its composites at different carbon nanotubes (CNTs) concentrations

hot-mixing method for composite preparation. The homogeneous distribution of CNTs in the composite can significantly decrease the CNT loading and increase the bonding force, which increases the mechanical strength and modulus by avoiding intertube sliding [12]. However, aggregation of CNTs can be observed in the composites with 20 wt.% CNTs (MP-C20) (as shown in Fig. 2), which indicates that excessive CNTs amount effects the dispersion.

Bending properties are measured and presented in Fig. 3. The data show that the neat MP-based graphite has a bending strength value of 60.5 MPa and a bending modulus value of 6.4 GPa. The inclusion of 5 wt.% CNTs significantly increases the bending strength value. The values reach 78.6 MPa. However, the bending modulus value is dramatically decreased to 4.3 GPa. Decrease in bending strength and increase in bending modulus

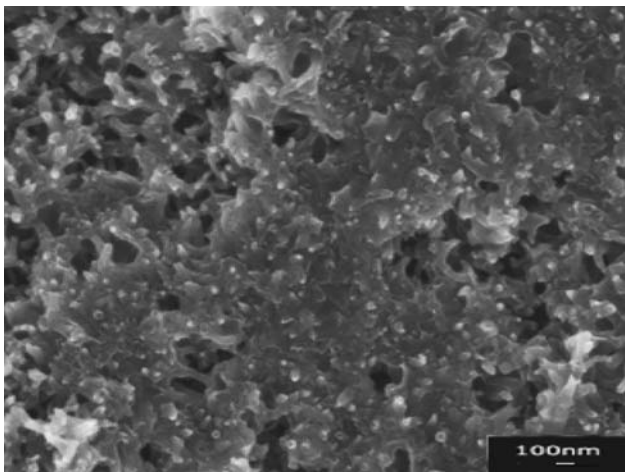

Fig. 2 Scanning electron microscopy (SEM) image of a composite with 20 wt.% carbon nanotubes (CNTs)

composite with 20 wt.% CNTs loading shows that excessive addition of CNTs make a disadvantageous effect to mechanical properties.

Figure 4 shows XRD profiles of MP and its composites graphitized at 2,800 °C, and their crystalline parameters are summarized in Table 1. The acute peaks seen in these scans indicate that the crystallite sizes in these carbons are quite great. With addition of CNTs of 5 wt.%, the value of d_{002} decrease from 0.3371 to 0.3366 nm and the value of La(110) increase from 558.1 to 791.0 nm. However, excessive addition of CNTs into composites increases the value of d_{002} to 0.3377 nm and decreases the value of La(110) to 413.0 nm. It indicates that suitable addition of CNTs promotes graphitization degree of MP due to carbon

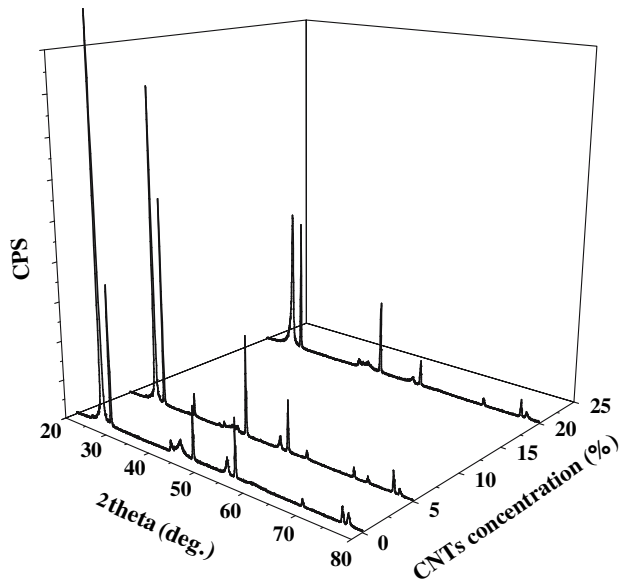

Fig. 4 XRD patterns of mesophase pitch (MP) and its composites powders graphitized at 2,800 °C

Table 1 Crystalline parameters and thermal/electrical conductivity of mesophase pitch (MP) and its composites

	d_{002} (nm)	Lc(002) (nm)	La(110) (nm)	Electrical conductivity (S cm^{-1})	Thermal conductivity (W m^{-1} K^{-1})
MP	0.3371	102.10	558.8	854	53.6
MP-C05	0.3366	75.11	791.7	1,175	118.1
MP-C20	0.3377	28.91	413.0	554	14.7

atoms may orderly grow along CNTs. Yet more CNTs may also obstruct the carbon atoms into graphite crystal lattice, which leads to disordering of carbon atoms in the composites with 20 wt.% CNTs. Increase in ordering of carbon atoms leads to the increases of thermal/electrical conductivity of composites (Table 1).

In summary, well-dispersed MP/carbon nanotube composites were fabricated. After graphitization at 2,800 °C, the crystallinity was increased by 8% for a composite with 5 wt.% CNTs. Due to the combined excellent mechanical and thermal/electrical properties of CNTs, the bending strength and thermal/electrical conductivity of the composites were simultaneously improved by 30% and 120/38%, respectively, in composites with 5 wt.% CNTs.

References

1. Iijima S (1991) Nature 354:56
2. Ajayan PM (1999) Chem Rev 99:1787
3. Sreekumar TV, Liu T, Min BG, Guo H, Kumar S, Hauge RH, Smalley RE (2004) Adv Mater 16:58
4. Shaffer MSP, Windle AH (1999) Adv Mater 11:937
5. Schmidt J, Moergenthaler KD, Brehler KP, Arndt J (1998) Carbon 36:1079
6. Chen MH, Wu GT, Zhu GM, You JK, Lin ZG (2002) J Solid State Electrochem 6:420
7. Blanco C, Santamaria R, Bermejo J, Menendez R (2000) Carbon 38:1043
8. Mochida I (1995) Carbon 33:265
9. Dumont M, Chollon G, Dourges MA, Pailler R, Bourrat X, Naslain R et al (2002) Carbon 40:1475
10. Drbohlav J, Stevenson WTK (1995) Carbon 33:713
11. Hoffmann WR, Hüttinger KJ (1993) Carbon 31:259
12. Suhr J, Koratkar N, Keblinski P, Ajayan P (2005) Nat Mater 4:134