Indian Journal of Engineering & Materials Sciences
Vol. 21, August 2014, pp. 477-481

Continuous carbon nanotube-polycarbonate composite fibers through melt spinning

Soma Das[a], Mitali Saha[a]* & Sabyasachi Sarkar[b]

[a]Department of Chemistry, National Institute of Agartala, Tripura 799 046, India
[b]Department of Chemistry, Indian Institute of Technology, Kanpur 208 016, India

Received 22 August 2013; accepted 23 May 2014

Among the many potential applications of carbon nanotubes (CNT), its usage to strengthen polymers has been paid considerable attention due to the exceptional stiffness, excellent strength and the low density. This has provided numerous opportunities for the invention of new material systems for applications requiring high strength and high modulus. In this paper, composite fibers of polycarbonate (PC) and multiwall carbon nanotubes (MWNTs) are prepared in dimethyl formamide (DMF) using melt mixing-coagulation technique followed by melt spinning at 220°C. The spinning of the coagulated PC-MWNTs is done at various draw ratios and the resultant fibers are studied by optical microscope, SEM and AFM. The mechanical properties of the composite fiber are determined and it is found that the addition of 1-1.5 wt% of MWNT increased the tensile strength of the polymer fiber from 900 MPa to 1890 MPa and Young's modulus from 1372 MPa to 2060 MPa. The results show that the mixing of MWNT with the polycarbonate matrix, above its melting point followed by coagulation in water might have developed a strong interfacial bonding between functionalized MWNTs and PC.

Keywords: MWNTs, Polycarbonate, Composite fiber, Mechanical strength

The exceptional mechanical properties of carbon nanotubes (CNT) have made it as the super-strength fiber material for the present generation. The researchers are trying to build the worlds' strongest cables, fibers and fabrics from these nano-sized carbon structures. In spite of vast developments in the preparation of high-strength CNT-based fibers[1-7], many fibers still do not exceed strengths that have been achieved for hydrocarbon-based materials such as Dyneema and Kevlar. So far, various composite materials have been prepared by incorporating SWNTs or MWNTs into a polymer matrix, as polymers can be easily processed and fabricated into intricately shaped components without damaging CNTs during processing[8-11]. A variety of techniques to align CNTs have been documented in the literature, including methods based on stretching[12], magnetic[13], slicing[14], liquid crystals[15], shear flow[16], and surface acoustic waves[17]. Fibers reinforced with aligned CNTs have also been produced using wet spinning[18], melt spinning[19], and direct spinning[20]. Despite the significant amount of progress made towards producing high-performance fibers from polymer materials, the mechanical properties still remain only a fraction of the expected theoretical values for these materials. Besides, one major hang-up has also been the cost of these materials and a potential route towards reducing the cost of the CNT-based composites is either through using small quantities of CNT to reinforce the polymer or to modify the existing methods for high-performance applications.

In view of these observations, we have made some modifications in the existing processes and have applied melt mixing-coagulation method to achieve good nanotube dispersion throughout the polymer matrix. The resultant composite material was then subjected to spinning using a low cost home-made melt spinning equipment to get the CNT-polymer composite fibers.

Experimental Procedure

Preparation of multi-walled carbon nanotubes (MWNTs)

MWNTs were synthesized by burning mustard oil under insufficient flow of oxygen and functionalized following our own indigenous process[21]. Polycarbonate (PC) and other reagents were purchased from Sigma-Aldrich.

Preparation of PC-MWNT composite fibers

A simple technique was adopted for the preparation of PC-MWNTs composites. MWNT was first

*Corresponding author (E-mail: mitalisah@gmail.com)

functionalized by refluxing in dilute nitric acid. Commercially grade PC was first dissolved in minimum amount of dimethyl formamide (DMF) and then mechanically mixed with the MWNTs at a temperature above its melting point. The black liquid obtained, was then poured in excess amount of water to get a grey-black composite material which was washed with water and finally dried under vacuum.

Melt spinning of PC-MWNTs composite fibers and determination of mechanical properties

The PC-MWNT composite material was subjected to spinning using a self-designed, low-cost home-made melt spinning equipment. They were spun into fibers with a single holed spinneret of 1 mm diameter. The spinning was carried out at 220°C with a ram speed of 1.4 mm/min. The composite fibers were characterized by optical microscope, SEM and AFM. The tensile strength and modulus of drawn fibers were tested using Instron Universal Testing machine. The gauge length for testing a single fiber was kept at 20 mm with a testing rate of 20 mm/min.

Results and Discussion

Figure 1 showed the optical micrographs of fiber having different percentages of MWNTs in PC matrix. As, no agglomerations of the MWNTs were observed in the figure, it indicated that the nanotubes were uniformly distributed within the polymer matrix on a micrometer scale.

Figure 2 presented the SEM images of PC-1.5% MWNT fibers which showed that MWNTs were uniformly distributed throughout the polymer matrix and also properly aligned within the polymer matrix along the fiber axis. It may be attributed to the fact that

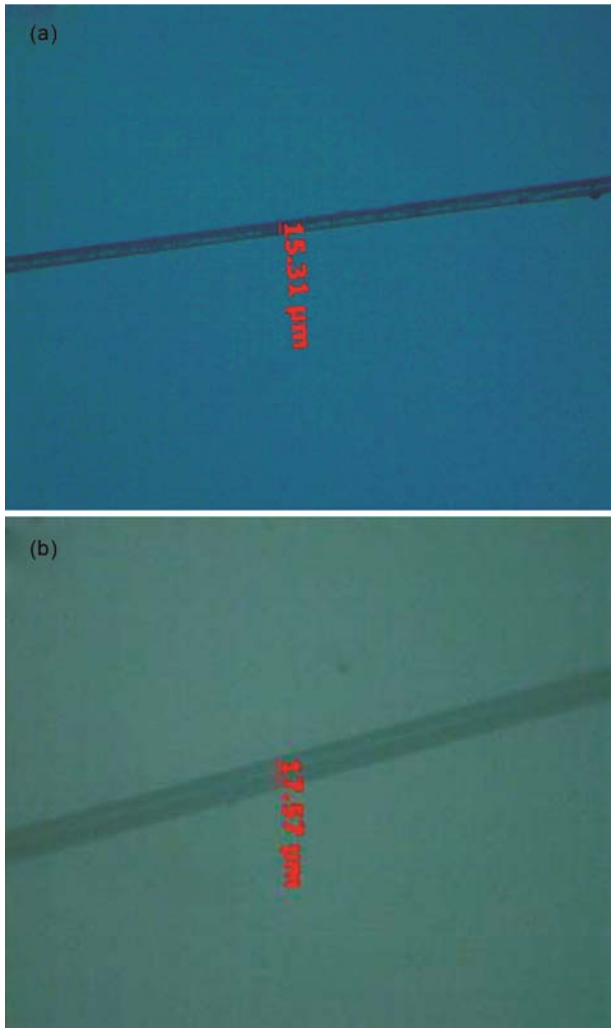

Fig. 1(a,b)—Optical micrographs of PC-1% MWNT fiber

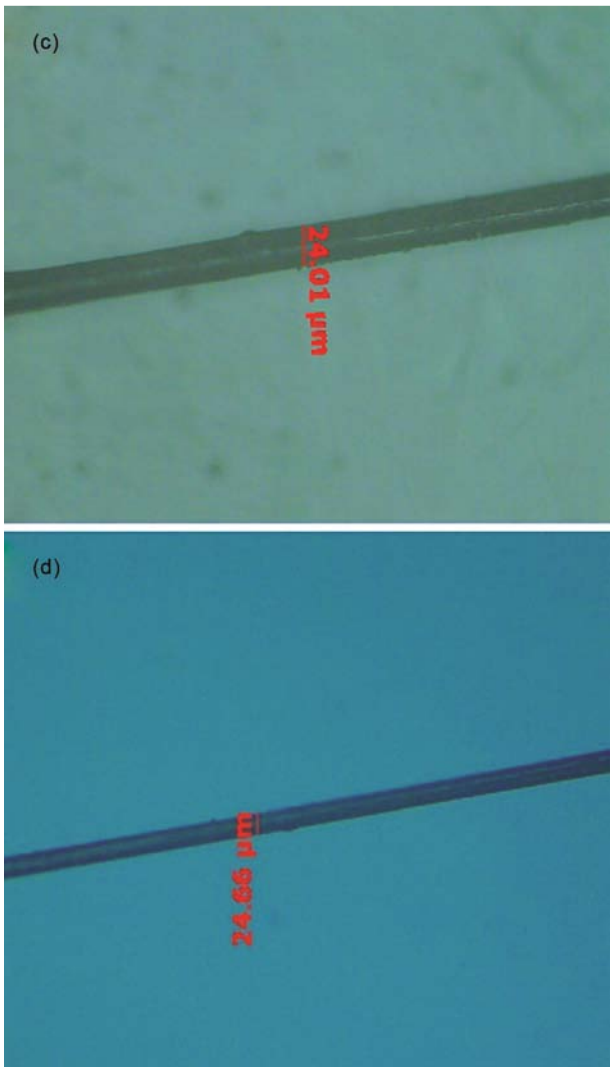

Fig. 1(c,d)—Optical micrographs of PC-1.5% MWNT fiber

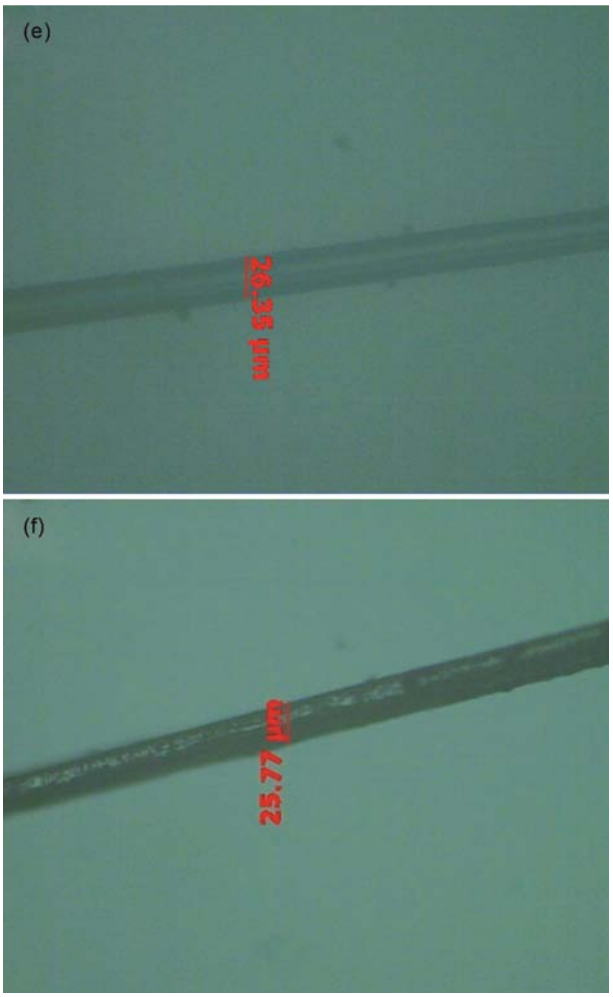

Fig. 1(e,f)—Optical micrographs of PC-2% MWNT fiber

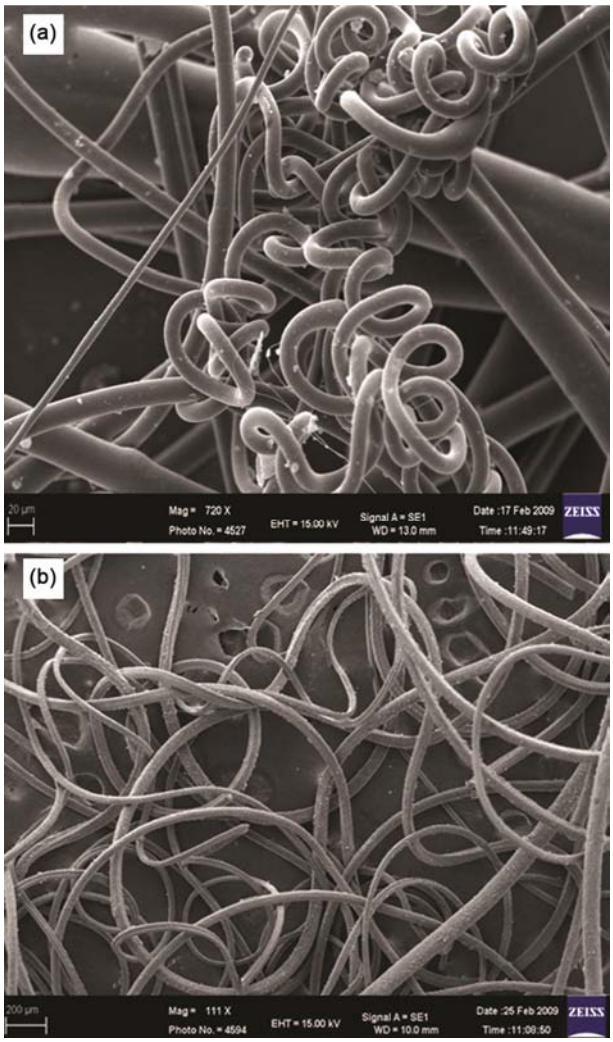

Fig. 2(a,b)—SEM images of PC-1.5% MWNT fibers

during the process of melt mixing along with coagulation, the polymer chains might have entrapped the surrounding nanotube bundles and the carbon nanotubes were not able to agglomerate as the composite material was immediately precipitated in the water after their melt mixing in DMF. Thus, the conformation of the polymer chains changed from a relatively expanded state in DMF to a collapsed state in the DMF/water mixture.

Figure 3 showed the AFM image of a single PC-1.5% MWNT fiber, which further indicated the alignment of the MWNTs within the polymer matrix along the fiber axis. Hence, the coagulation approach after melt mixing provided a better dispersion of MWNTs in the polymer matrix and also prevented the rebundling of nanotubes. It was found that spinning into fibers became difficult when the content of MWNTs in PC was increased. The flow properties of the material was reduced, so spinning was not smooth and the spun fibers were found broken. Thus, up to 1.5% of MWNTs, continuous fibers in meters were produced without any breakage. But, on further increasing the concentration of MWNTs up to 2%, breakage occurred during the drawing process. This may be due to non-uniform dispersion of MWNTs in the PC matrix at higher concentration. The tensile strength and modulus of neat PC fiber as well as PC-MWNTs fiber were given in Table 1.

It can be seen from Table 1 that the diameter of neat PC fiber was found to be 10-15 µm. The tensile strength was 900 MPa and modulus was found to be 1372 MPa. On addition of 1% of MWNTs in PC, the tensile strength and modulus increased to 1310 MPa and 1680 MPa, respectively, in spite of the increase in diameter. In case of 1.5% addition of MWNTs, the tensile strength as well as modulus were found to increase significantly resulting in 1890 MPa and 2090

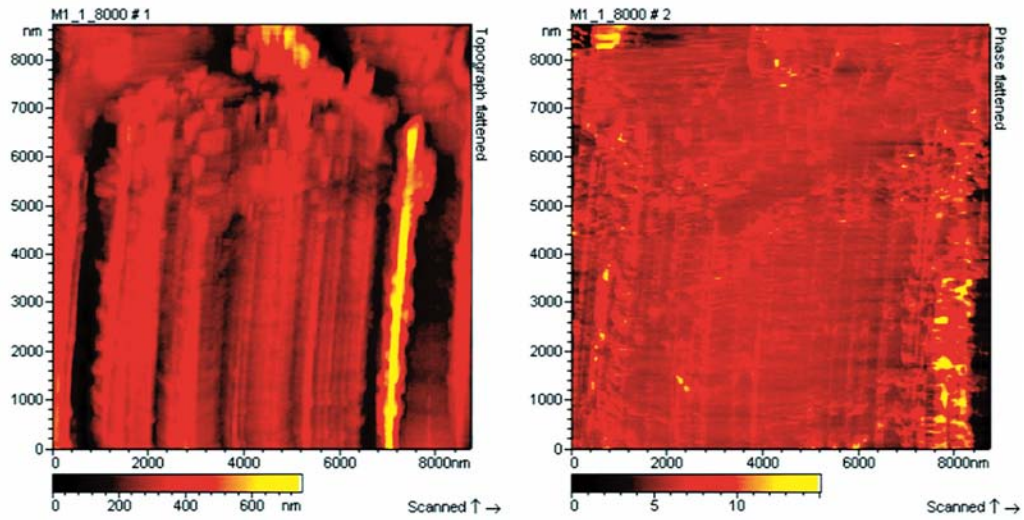

Fig. 3—AFM of single PC-1.5% MWNT fiber

Table 1—Mechanical properties of neat PC and PC-MWNTs fiber

Sample	Diameter (μm)	Tensile strength (MPa)	Tensile modulus (MPa)	Elongation at break (%)
Neat PC	10-15	900	1372	118
PC-1% MWNTs	15-18	1310 (+31)	1680 (+18)	115
PC-1.5% MWNTs	24-25	1890 (+52)	2060 (+33)	94
PC-2% MWNTs	25-27	1850 (+51)	1935 (+29)	109

MPa, respectively. However, on further addition up to 2%, the strength and modulus slightly decreased showing the tensile strength and modulus values of 1850 MPa and 1935 MPa, respectively. The increase in the mechanical properties of the composite fibers up to 1.5% were obviously due to the homogeneous dispersion of MWNTs in PC matrix and proper alignment of the MWNTs along the fiber axis, resulting in smooth spinning of fibers. The decrease in the mechanical properties in case of 2% addition of MWNTs in PC was may be due to non-uniform dispersion of carbon nanotubes within the polymer matrix, which distorted the alignment of the carbon nanotubes. When the amount was more than 5 wt%, it was difficult to obtain continuous fibers and they became very brittle.

Conclusions

PC-MWNT composite fibers were produced by melt mixing-coagulation method followed by melt spinning. The spinning was done smoothly up to 1.5% addition of MWNTs in PC, resulting in significant increase in the mechanical properties of the single fiber. Thus, the coagulation method along with melt mixing was found to be effective for uniform dispersion of carbon nanotubes in the polymer matrix and hence can be used to produce nanotube-polymer composite fibers. This method has great advantage for continuous and large-scale production of nanocomposite fibers.

Acknowledgments

One of the author (Mitali Saha) is grateful to AICTE, New Delhi, for financial assistance.

References

1 Guo W, Liu C, Sun X, Yang Z, Kia H G & Peng H, *J Mater Chem*, 22 (2012) 903-908.
2 Chae H G, Sreekumar T V, Uchida T & Kumar S, *Polymer*, 46 (2005) 10925-10935
3 Sahoo N G, Sravendra R, Cho J W, Li Lin & Chan S H, *Progr Polym Sci*, 35 (2010) 837-867.
4 Ma C, Zhang W, Zhu Y, Ji L, Zhang R, koratkar N & Liang J, *Carbon*, 46 (2008) 706-710.
5 Moniruzzaman M & Winey K I, *Macromolecules*, 39 (2006) 5194-5205
6 Sen R, Zhao B, Perea D, Itkis M E, Hu H, Love J, Bekyarova E & Haddon R, *Nano Lett*, 4 (2004) 459-464.
7 Paipetis A, Kostopoulos V, (Eds), *Carbon Nanotube Enhanced Aerospace Composite Materials. A New Generation of Multifunctional Hybrid Structural Composites Series: Solid Mechanics and Its Applications*, (Springer), 2013, 188.
8 Laforgue1 A, Champagne1 M F, Dumas J & Robitaille L, *J Eng Fibers Fabrics*, 7 (3) 2012)118-124.
9 Song K, Zhang Y, Meng J, Green E C, Tajaddod N, Li H & Minus M L, *Materials*, 6 (2013) 2543-2577.
10 Panamoottil1 S M, Pötschke P, Lin R J T & Bhattacharyya D, Fakirov S, *eXPRESS Polym Lett*, 7(7) (2013) 607–620.
11 Haslam M D & Raeymaekers B, *Compos Part B*, 60 (2014) 91-97.

12 Jin L, Bower C & Zhou O, *Appl Phys Lett*, 173(9) (1998) 1197-1199.
13 Prolongo S G, Meliton B G, Del Rosario G & Ureña A, *Compos Part B-Eng*, 46 (2013) 166-172.
14 Ajayan P, Stephan O, Colliex C & Trauth D, *Science*, 265 (1994) 1212-1214.
15 Lagerwall J, Scalia G, Haluska M, Dettlaff-Weglikowska U, Roth S & Giesselmann F, *Adv Mater*, 19 (2007) 359-364.
16 Fan Z & Advani S G, *Polymer*, 46 (2005) 5232-5240.
17 Strobl C J, Schaflein C, Beierlein U, Ebbecke J & Wixforth A, *Appl Phys Lett*, 85 (2004) 1427-1429.
18 Zhang S, Koziol K K, Kinloch I A & Windle A H, *Small*, 4 (2008) 1217-1222.
19 Gao J, Itkis M E, Yu A, Bekyarova E, Zhao B & Haddon R C, *J Am Chem Soc*, 127 (2005) 3847-3854.
20 Li Y L, Kinloch I A & Windle A H, *Science*, 304 (2004) 276-278.
21 Dubey P, Muthukumaran D, Dash S, Mukhopadhyay R & Sarkar S, *Pramana – J Phys*, 65 (2005) 681-697