The present status and key problems of carbon nanotube based polymer composites

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Abstract. The state-of-art and key problems of carbon nanotube (CNT) based polymer composites (CNT/polymer composites) including CNT/polymer structural composites and CNT/polymer functional composites are reviewed. Based on the results reported up to now, CNTs can be an effective reinforcement for polymer matrices, and the tensile strength and elastic modulus of CNT/polymer composites can reach as high as 3600 MPa and 80 GPa, respectively. CNT/polymer composites are also promising functional composite materials with improved electrical and thermal conductivity, etc. Due to their multi-functional properties, CNT/polymer composites are expected to be used as low weight structural materials, optical devices, thermal interface materials, electric components, electromagnetic absorption materials, etc. However, the full potential of CNT/polymer composites still remains to be realized. A few key problems, such as how to prepare structure-controllable CNTs with high purity and consistently dependable high performance, how to break up entangled or bundled CNTs and then uniformly disperse and align them within a polymer matrix, how to improve the load transfer from matrix to CNT reinforcement, etc, still exist and need to be solved in order to realize the wide applications of these advanced composites.

Keywords: polymer composites, carbon nanotubes, mechanical properties, electrical properties, thermal properties

1. Introduction

Since Iijima's report on CNTs in 1991 [1], scientists have been attracted by CNT's unique atomic structure and properties, such as high aspect ratio, high strength-to-weight ratio, extraordinary mechanical properties (their axial elastic modulus and tensile strength were theoretically predicted to be as high as 1–2 TPa and 200 GPa [2–6], respectively), superior thermal and electrical properties (thermally stable up to 2800°C in a vacuum or inert atmosphere, thermal conductivity about twice as high as diamond [7], and electric-current-carrying capacity 1000 times higher than copper wires [8]). These properties of CNTs endow themselves with new scientific and technological opportunities as an

ideal filler material in composites. To date, various composite materials have been prepared by incorporating SWNTs or MWNTs into a metal matrix [9–12], a ceramic matrix [13–16] or a polymer matrix [17–33]. Polymers can be easily processed and fabricated into intricately shaped components without damaging CNTs during processing using conventional methods, and hence the manufacturing cost can be certainly reduced. Therefore, CNT based polymer composites (CNT/polymer composites) stimulate great interests and have been extensively investigated.

According to the different application purposes, CNT/polymer composites can be classified as structural composites and functional composites,

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respectively. CNTs play a different role in the two kinds of composites. For CNT/polymer structural composites, the unique mechanical properties of CNTs, such as the high elastic modulus, tensile strength and strain to fracture, the ability to withstand cross-sectional and twisting distortions and compression without fracture, are generally explored to obtain structural materials with lightweight, high elastic modulus, high tensile and compressive strength and stiffness, etc. While for CNT/polymer functional composites, many other interesting properties of CNTs, such as the high electrical and thermal conductivity, are used to develop functional materials possessing heat resistance, chemical sensing, electrical conducting, thermal conducting, photoemission, electromagnetic absorbing or energy storage performances, etc.

In recent years, many polymers, such as epoxy [17–19], PMMA [20–22], PVA [23], PVC [24, 25], PP [26–28], PE [29, 30], PA12 [31], and PS [32, 33], have been employed as matrices to prepare CNT/polymer composites. The mechanical properties combining with electrical, thermal, optical and many other properties have been extensively investigated by many research groups for a wide range of applications. However, the reported results about these properties are scattered and still far from satisfactory. Analyses of these research results show that the performance of these composites depends on many factors, such as the types of CNTs (SWNTs or MWNTs) and their morphology and structure (diameter, length and chirality, for example) [34], the processing method, the choice of matrix [34, 35], the CNT dispersion within matrix [36], the interfacial interaction between CNTs and matrix, and so on.

In this article, we will present a comprehensive review about the preparation and processing, the properties and application of CNT/polymer composites. Some fundamental challenges, which have to be overcome in order to enable the wide applications of these advanced composites, are discussed.

2. Preparation of CNT/polymer composites

In the published reports, the common fabricating methods of CNT/polymer composites are solution mixing, melt blending and *in situ* polymerization. In solution mixing, CNTs are generally dispersed in solvent and then mixed with polymer solution by mechanical mixing, magnetic agitation or high-energy sonication. Subsequently, the CNT/polymer composites can be obtained by vaporizing the solvent at a certain temperature. This method is considered as an effective measure to prepare composites with a homogenous CNT distribution and often used to prepare composite films. Epoxy is a kind of thermosetting resins and is usually employed in CNT/epoxy composites by solution mixing. Many researchers, such as Xu *et al.* [37] and Lau *et al.* [38], have fabricated the CNT/epoxy composite using this method and reported their properties. Beside the epoxy resin, other polymers such as PS, PVA, PMMA and PVC can also be processed into CNT/polymer composites by solution mixing. Mamedov *et al.* [39] applied a new deposition technique called LBL assembly to make SWNT/polyelectrolyte multilayer composites, which can be considered as an improved solution mixing method. The resulting SWNT/polyelectrolyte membranes were very strong and the ultimate tensile strength was found to be 220±40 MPa with some readings as high as 325 MPa.

Melt blending is a versatile and commonly used method to fabricate polymeric materials, especially for thermoplastic polymers. CNTs are dispersed within the polymer matrix by rheological shear stress generated from the blending of CNTs with melt polymer. Its well-known disadvantage is that CNTs can easily be damaged to a certain extent or broken in some cases [40, 41]. All the CNT/polymer composites using thermoplastic polymers as matrix can be processed in this manner. Jin *et al.* [42] fabricated MWNT/PMMA composite using this method and studied its dynamic mechanical behavior. The results indicate that CNTs were well dispersed in the polymer matrix with no apparent damage or breakage. The storage modulus of the polymer was significantly increased by the incorporation of CNTs particularly at high temperatures. The melt blending method is frequently used to prepare CNT/polymer composite fibers. Haggenmuller *et al.* [22] and Kumar *et al.* [26] prepared SWNT/PMMA composite fibers and CNF/PP composite fibers using melt spinning process and the obtained fibers showed improved mechanical properties.

In situ polymerization is considered as a very efficient method to significantly improve the CNT dispersion and the interaction between CNTs and polymer matrix. Generally, CNTs are firstly mixed with monomers and then CNT/polymer composites

can be obtained by polymerizing the monomers under certain conditions. Jia *et al.* [20] initially reported the fabrication of CNT/PMMA composites by in situ polymerization. The results showed that a strong binding interface was formed between the CNTs and PMMA because the CNTs could be initiated by AIBN to open their π bonds and participate in the PMMA polymerization. It was reported that for a surface modified CNT/PMMA composite fabricated by an improved in situ process, not only was the dispersion ratio of CNTs in the PMMA matrix improved, but also the mechanical properties of the composites were enhanced when the CNT content was lower than 10 wt%. Ying *et al.* prepared surfactant-coated CNT/epoxy composites by in situ polymerizing epoxy resin with polyamide curing agent, and then their mechanical properties were studied [43–45]. The results showed that the tensile strength and ultimate strain of the composites were increased by 20% and 70%, respectively. Li *et al.* [46] reported the fabrication and characterization of SWNT/PANI composites by *in situ* polymerization of an aniline solution containing different SWNT contents. Electron microscopy and conductive analysis showed that they had uniform CNT dispersion and high electric conductivity.

In order to bring out the anisotropic nature of CNTs, it is important to align CNTs in a polymer matrix during processing of CNT/polymer composites. This has been done by several groups [47–57] based on slicing the composite [51], mechanical stretching [52, 53], filtrating the CNT dispersion under a high magnetic field [54], melt-processing and electrospinning [55], etc. Gao *et al.* [56] described that aligned conducting CNTs can be used to make novel CP-NT coaxial nanowires by electrochemically depositing a concentric layer of an appropriate conducting polymer uniformly onto each of the constituent aligned CNTs. For a conducting-polymer sheathed CNT with an outer diameter of 221 nm, inner diameter of 94.7 nm and length 9.3 μm, its elastic modulus was determined to be 13.6 GPa, much higher than that of the polymer. Li *et al.* [24] first reported to successfully synthesize long ropes of SWNT bundles with rope lengths up to 30 mm. These ropes consist of roughly aligned SWNT bundles, which consist of well-aligned SWNTs. Then such aligned SWNT ropes were impregnated with PVC resin to form composite ropes. These ropes showed significantly improved mechanical properties with the average

tensile strength as high as 3.6 GPa. Dalton *et al.* [25] used a modified coagulation-based CNT spinning method to spin a reel of nanotube gel fiber and then convert it into 100 m long solid CNT composite fiber in a continuous process, at a rate of more than 70 cm·min⁻¹. The obtained composite fibers were about 50 μm in diameter and contained around 60 wt% SWNTs. They had a tensile strength of 1.8 GPa and tougher than most natural or synthetic organic fibers described so far.

3. Present status of CNT/polymer composites

3.1. CNT/polymer structural composites

CNTs are considered to be the ultimate mechanical filler material [58] and open the way for a new generation of high performance structural composites. In recent years, many studies with different focuses on mechanical properties of CNT/polymer structural composites have been published. However, the most impressive themes seem to be enhancement of elastic modulus and tensile strength. Another important property–fracture toughness has also been reported to be significantly improved in many works. We therefore will focus on the elastic modulus, tensile strength and toughness enhancement below.

Table 1 and 2 show some representative mechanical properties of randomly oriented and aligned CNT/polymer composites, respectively. From these two tables, it can be found that the elastic modulus of CNT/polymer composites can always be obviously increased comparing with those of pure polymer matrices. However, most of randomly oriented CNT/polymer composites show only a moderate or no strength enhancement, especially for CNT/polymer composites using untreated CNTs as reinforcements, mainly attributed to poor CNT dispersion within the matrix and poor load transfer between CNTs and polymer matrix. Homogeneous dispersion of CNTs throughout a matrix without destroying their integrity is essential for CNT reinforced polymer composites. Moreover, good interfacial bonding is required to achieve effective load transfer across CNT/matrix interface, a necessary condition for improving the mechanical properties of polymer composites [59–64]. From Table 1, it can be seen that the strength of nearly all CNT/polymer composites using modified CNTs as reinforce-

Table 1. Representative mechanical properties of randomly oriented CNT/polymer composites

Author	Sample	Tensile strength [MPa]	Elastic modulus [GPa]	Remarks
Gong *et al.* [65]	Epoxy+1wt% MWNTs		Increased by more than 30%	
Ying *et al.* [44]	Pure Epoxy	52	1.18	
	Epoxy + 2wt% MWNTs	46	1.18	Untreated MWNTs
	Epoxy + 2wt% MWNTs	55	1.35	MWNTs were treated with PVA
	Epoxy + 2wt% MWNTs	62	1.39	MWNTs were treated with poly-oxyethylene-9-alkyl ether
	Epoxy + 2wt% MWNTs	58	1.34	MWNTs were treated with poly-oxyethylene-7-alkyl ether
Andrews *et al.* [66]	PS + 0.05 to 0.5 vol.% MWNTs	Significant reduction	Little change	MWNTs were heated in an inert atmosphere to a high temperature in the range 1800 to 2600°C
	PS + 2.5 to 25 vol.% MWNTs		1.9~4.5	
	High impact PS & ABS + 0.05 to 0.5 vol.% MWNTs	Little effect on tensile strength of either	Small increase	
Zhang *et al.* [67]	HDPE + 2 wt% SDS	~14.5	~0.47	SWNT aqueous solution prepared at the aid of SDS was sprayed onto the surface of HDPE. The composites were obtained by melt processing
	HDPE + 2 wt% SDS + 0.5 wt% SWNT	~19	~0.57	
	HDPE + 2 wt% SDS + 2.6 wt% SWNT	~23	~0.7	
Qian *et al.* [68]	PS	~12.8	~1.19	
	PS + 1 wt% MWNTs	~16	~1.62	CNT with 15 µm average length
	PS + 1 wt% MWNTs	~16	~1.69	CNT with 50 µm average length
Jia *et al.* [20]	PMMA + 0.08 wt% AIBN	54.9		
	PMMA + 0.24 wt% AIBN + 1 wt% CNTs	21.2		Untreated CNTs
	PMMA + 0.28 wt% AIBN + 3 wt% CNTs	20.3		
	PMMA + 0.1 wt% AIBN + 1 wt% CNTs	58.7		CNTs were ground in a ball mill and the improved *in situ* process was used to fabricate the composites
	PMMA + 0.1 wt% AIBN + 3 wt% CNTs	66.8		
	PMMA + 0.1 wt% AIBN + 5 wt% CNTs	71.7		
Jin *et al.* [42]	PMMA + MWNTs	The storage modulus was significantly increased, particularly at high temperature		
Jin *et al.* [69]	PMMA + MWNTs	The storage modulus of composite containing 0.5 wt% PVDF was almost twice as that of MWNT/PMMA without PVDF at 50°C		
Geng *et al.* [70]	PEO		0.06	
	PEO + 1 wt% fluorinated SWNTs		0.15	
Tong *et al.*[29]	Pure PE	34.1	0.81	
	PE + 0.5 wt% SWNTs	28.9	0.64	Untreated SWNTs
	PE + 0.5 wt% SWNTs	33.3	0.8	SWNTs were modified with PE prepared by in situ Ziegler-Natta polymerization
Paiva *et al.* [71]	PVA		~4	
	PVA + 2.5 wt% SWNTs		~5.6	CNTs were functionalized with low molecular weight PVA
	PVA + 5 wt% SWNTs		~6.2	
	PVA + 2.5 wt% SWNTs		~5.4	Untreated SWNTs

ments can be increased to some extent. It means that the surface modification of CNTs plays an important role in improving the mechanical reinforcement of the composites.

Compared to randomly oriented CNT/polymer composites, aligned configuration has a higher efficiency of reinforcement. In most cases, both tensile strength and elastic modulus of aligned CNT/polymer composites are significantly improved. In Figure 1, the tensile strength (Figure 1a) and elastic modulus (Figure 1b) of randomly oriented and aligned CNT/polymer composites are summarized. It can be seen that the tensile strength and elastic modulus of randomly oriented CNT/polymer composites are generally less than 100 MPa and 6 GPa, respectively. They are much lower than those of aligned CNT/polymer composites, which can even reach as high as 3600 MPa and 80 GPa, respec-

Table 2. Representative mechanical properties of aligned CNT/polymer composites

Author	Sample	Tensile strength [MPa]	Elastic modulus [GPa]	Remarks
Gao *et al.* [56]	PANI + MWNTs		13.6	Prepared by electrochemical deposition
Andrew *et al.* [48]	Isotropic petroleum pitch macrofiber	~450	~33	Produced by extrusion. The average diameter of fibers was ~18 μm
	Isotropic petroleum pitch + 1% SWNT macrofiber	~620	~42	
	Isotropic petroleum pitch + 5% SWNT macrofiber	~850	~78	
Haggenmuelle *et al.* [22]	Pure PMMA fiber		~3.1	Produced by the melt spinning process
	PMMA + 1 wt% SWNT nanocomposite fiber		~3.3	
	PMMA + 5 wt% SWNT nanocomposite fiber		~5	
Kumar *et al.* [26]	PP macrofiber	490	4.6	Spun using conventional melt spinning technique
	PP + 5 wt% MWNT macrofiber	570	7.1	
Bhattacharyya *et al.* [50]	PP	430	4.2	Significant CNT aggregation
	PP + 0.8 wt% SWNTs	420	4	
Li *et al.* [24]	PVC + SWNTs	3600		
Vigolo *et al.* [49]	PVA + SWNT composite fibers		9~15	
Dalton *et al.* [25]	PVA + 60 wt% SWNT composite fiber	1800	80	Produced by modified coagulation-based CNT spinning method
Siochi *et al.* [72]	PI	105	2.2	SWNT alignment in the composite fiber direction was induced by shear force present during the melt extrusion and fiber drawing processes
	PI + 0.1 wt% SWNTs	105	2.6	
	PI + 0.3 wt% SWNTs	105	2.8	
	PI + 1 wt% SWNTs	105	3.2	
Chen and Tao [73]	TPU	~12.4	~7.7	Prepared by a solution process
	TPU + 0.5 wt% SWNTs	~13.3	~14.5	
Ruan *et al.* [74]	UHMWPE	~3510	~122.6	Gel-spun composite fibers were drawn at draw ratio of 30
	UHWMPE + 5 wt% MWNTs	~4170	~136.8	

tively. Moreover, the tensile strength and elastic modulus are always higher for aligned CNT/polymer composites containing higher CNT content, while this is not the case for randomly oriented CNT/polymer composites.

In many reports, a critical CNT content in matrix can be found when the CNT strengthening effect on randomly oriented CNT/polymer composites are investigated. Below this content, the strengthening effect for randomly oriented CNT/polymer composites increases with increasing CNT content. Above this content, the strength of CNT/polymer composites decreases, and in some cases, even lower than that of the polymer matrix. The work of Bai *et al.* [34] shows that the critical CNT loading percentage was about 0.5 wt% for CNT/epoxy composites. This ratio is also very close to the percolation threshold for electrical conductivity. The same tendency was reported by Meguid and Sun [75] in their work on the tensile and shear strength of nano-reinforced composite interfaces. The

excess CNTs increase the viscosity of polymers, and can also cause some surface of the CNTs not be completely covered by polymer matrix due to the large specific surface area of CNTs. This makes uniform dispersion and load transfer more difficult. Moreover, it is very difficult for high quantity of polymers to intercalate among CNTs when the CNT content becomes high. Shaffer *et al.* [76] reported that, by trivial estimation, even a 1 vol% loading of SWNTs ensures that all of the polymer molecules are within one radius of gyration (say 5 nm) of a nanotube. This result implies that complete wetting of high loading fraction of SWNTs will be difficult, at least by conventional means.

Although much higher than those of randomly oriented CNT/polymer composites and, in some cases, representing the highest mechanical properties amongst all ultra-strong fibers reported so far, the obtained tensile strength and elastic modulus of aligned CNT/polymer composites are still far from satisfactory as far as the mechanical properties of
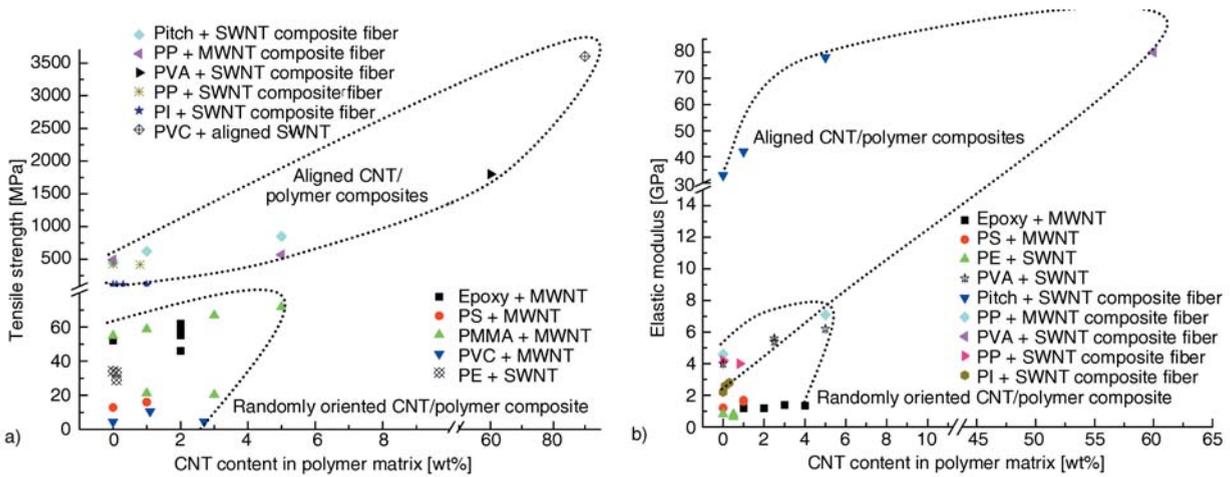
Figure 1. Tensile strength (a) and elastic modulus (b) of CNT/polymer composites

CNTs are considered. The main possible reason is sliding between CNTs under macroscopic tensile loading and between CNT and matrix.

Figure 2 shows several representative stress-strain curves of CNT/polymer composites with different polymers as matrices. It can be found that the curvilinear trend of CNT/polymer composites strongly depends on the matrix nature. Most of the reported stress-strain curves show improvement in strength and loss of strain to failure (Figure 2a, c, d). The strain to failure usually decreases with the increase of CNT content. However, CNT/polymer composites using PE and PEO as matrices show a substantial enhancement in strain to failure and toughness (Figure 2b). This indicates that significant toughening of some polymer matrices through the incorpo-
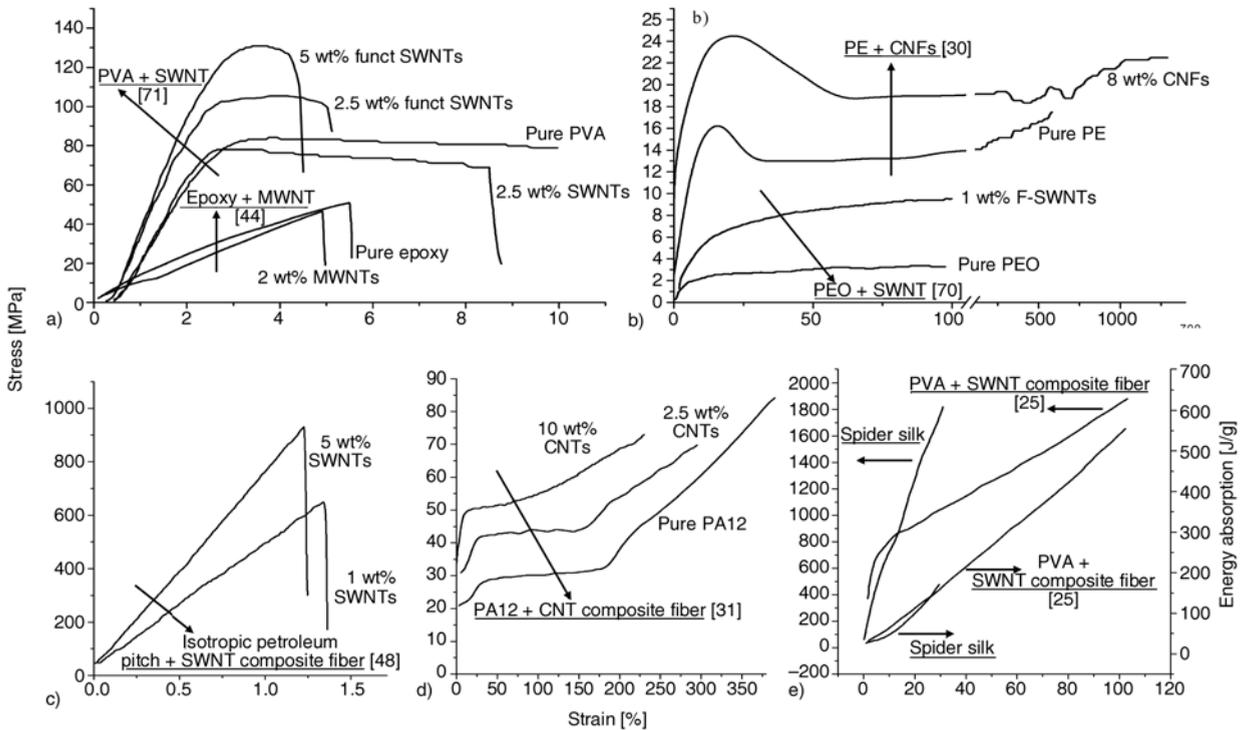
Figure 2. Representative strain-stress curves of CNT/polymer composites using different polymers as matrix. a, b) randomly oriented CNT/polymer composites [30, 44, 70, 71] and c, d, e) aligned CNT/polymer composites [25, 31, 48]

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[30, 31, 71] Reused with permission from Elsevier
[48] Reused with permission from American Institute of Physics
[70] Reused with permission from Wiley-VCH

ration of CNTs can be obtained. In such instances, the commonly used polymers including PE, PEO and PVA, are semi-crystalline thermoplastic polymers. Certain conditions, such as the presence of modified CNTs, preparation temperature of composites, pre-drawing treatment of CNT/polymer composite fibers at the temperature above the T_g of polymer matrix generally can influence their crystallization behaviour, and hence significantly affect the toughness characteristics of the composites. Ruan *et al.* [77] first reported that MWNT/PE composite films with high pre-drawing ratios showed an up to 150% increase in strain energy density together with a simultaneous increase of tensile strength of ~25% and an increase of ductility up to 140% in comparison with the pure PE film at similar pre-drawing ratio. Secondary crystallites, which were nucleated from the MWNTs and might grow during hot drawing, were much more mobile than the extended PE crystals and led to a significantly enhanced ductility and hence an increase in strain energy of the composite film [78]. Lozano *et al.* [30] reported that CNFs, which can also be considered as MWNTs since the diameter of fibers ranging between 40 and 150 nm, and PE could act in a synergistic manner to increase both strength and strain to failure of CNF/polymer composites (Figure 2b). They prepared the composite by mixing the functionalized CNFs and PE at 180°C and then pressed at 200°C. Dalton *et al.* [25] reported that their pre-drawn 60 wt% SWNT/PVA composite fibers matched the energy absorption of spider silk up to the breaking strain of the toughest silk (30%), and continued absorbing energy until reaching an energy-to-break (570 J/g) which is much higher than that of spider dragline silk (165 J/g [79]) (Figure 2e). While SWNT/PVA composite film, which was prepared by wet-casting [71] and no pre-drawing treatment and crystallinity, shows no increase in toughness (Figure 2a).

Since CNTs can enable polymer matrix to obtain a stronger and more lightweight composite due to their superior mechanical properties and small size, the obvious application of CNT/polymer composite can be as structural materials in several fields including aerospace, sports, automotive, infrastructure and defense. For example, super strong CNT/polymer composite fibers will have many applications including body and vehicle armor, transmission line cables, woven fabrics and textiles, etc. CNTs' high strength and toughness-to-weight char-

acteristics may also prove valuable as part of composite components in fuel cells that are deployed in transport applications, where durability is extremely important. Inspired partly by science fiction, NASA scientists are seriously considering space elevators as a mass-transit system. The material of choice for a space elevator is a super-strong CNT/polymer composite fiber, which is the only known substance at present that can successfully be used and make a space elevator possible. Several manufacturers of sports equipment have advertised CNTs as reinforcements of some of their top sports products. Easton Sports, the leading manufacturer of sporting goods, manufactured the world's first CNT two-piece all-composite bats, which represents the best bats Easton Sports has ever made [80].

3.2. CNT/polymer functional composites

3.2.1. Electrical properties and application of CNT/polymer composites

Many experimental results have shown that the combination of CNTs with polymers offers an attractive route to introduce new electrical properties [17, 23, 66, 81], and conductive composites can be constructed at low loading of CNTs due to low percolation thresholds originated from the high aspect ratio and conductivity of CNTs. Figure 3 and Table 3 summarize some reported electrical properties of CNT/polymer composites. It can be found in Figure 3 that all CNT/polymer composites exhibit a typical percolation behavior and introduction of CNTs to polymers can increase the conductivity of the resulting composites to several orders of magnitude or even higher than ten orders of magnitude. However, very different percolation thresholds (from 0.0025 wt% for CNT/epoxy composite to 11 wt% for SWNT/P3OT composite) have been obtained by different research groups (see Table 3). Sandler *et al.* [17] measured the electrical conductivity of untreated CNT/epoxy composites, and found that the percolation threshold was between 0.0225 and 0.04 wt% CNTs. Subsequently, they further presented the lowest threshold (0.0025 wt% for aligned SWNT/epoxy composites) observed for CNT/polymer composites yet reported [83]. Allaoui *et al.* [84] also studied the electrical performance of MWNT/epoxy composites and revealed that the presence of a percolation thresh-
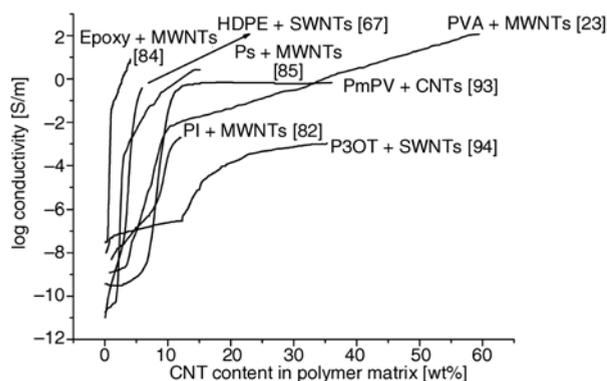
Figure 3. Representative semilogarithmic plot of the electrical conductivity of CNT/polymer composites with various weight percents of CNTs in different polymer matrices [23, 67, 82, 84, 85, 93, 94].
[23] Reused with permission from Wiley-VCH
[67, 82, 84, 94] Reused with permission from Elsevier
[85] Reused with permission from Springer
[93] Reused with permission from
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old between 0.5 and 1 wt% CNTs. When adding 1 wt% CNTs into epoxy matrix, the composite became conductive with a conductivity as high as 10^{-3} S/cm. Shaffer *et al.* [23] fabricated CNT/PVA composites and measured their electrical conductivity using impedance spectroscopy in a four-point configuration. The percolation threshold for these composites lay between 5 and 10 wt%. At the CNT concentration of 10 wt%, the electrical conductivity of the composite was about 1 S/cm, an increase

Table 3. Representative percolation threshold of CNT/ polymer composites

Author	Sample	Percolation threshold [wt%]
Sandler *et al.* [17]	Epoxy + CNTs	0.0225~0.04
Sandler *et al.* [83]	Epoxy + aligned MWNTs	0.0025
Allaoui *et al.* [84]	Epoxy + CNTs	0.5~1
Shaffer *et al.* [23]	PVA + MWNTs	5~10
Dufresne *et al.* [85]	PS + MWNTs	≤3
Andrews *et al.* [66]	PS + MWNTs	0.25 vol.%
Barraza *et al.* [86]	PS + SWNTs	8.5
Zhang *et al.* [67]	HDPE + SWNTs	4
McNally *et al.* [87]	PE + MWNTs	7.5
Hu *et al.* [88]	PET + MWNTs	0.9
Seo *et al.* [89]	PP + CNTs	1~2
Seo *et al.* [90]	PP + CNTs	2
Tchmutin *et al.* [91]	PP + SWNTs	4.5 vol.%
Meincke *et al.* [92]	PA6 + CNTs	4~6
Coleman *et al.* [93]	PmPV + CNTs	8.5
Kymakis *et al.* [94]	P3OT + SWNTs	11

by about seven orders of magnitude. Kymakis *et al.* [94] presented a percolation threshold of approximately 11 wt% SWNTs in a P3OT polymer. As the CNT concentration increased from 0 to 20 wt%, the conductivity of the composites was increased by five orders of magnitude.

Based on the reported results up to now, CNT/ epoxy composites show a much lower percolation threshold, although the reported values are very different. For CNT/PS composites, the percolation threshold was reported ranging from 0.25 vol% to 8.5 wt% by different groups. These differences can be attributed to many factors. The electrical properties of CNTs used by different research groups are generally significantly different depending on their purity, morphology, aspect ratio, etc. In addition, the interaction between matrix and CNTs influences the quality and quantity of electrical contacts between the CNTs. As reported by Shaffer *et al.* [23], it seems that the P3OT is easier to form an adsorbed layer on the surface of CNTs than epoxy resin, which reduces the quality and quantity of electrical contacts, thus the percolation threshold of CNT/P3OT composite cannot be reached at a relatively lower CNT loading. In Table 3, it can also be found that the electrical percolation threshold of CNT/polymer composites with semicrystalline thermoplastic matrices (PP, PE, PEO, PA) is always higher than those of CNT/epoxy composites. Hu *et al.* [88] first used coagulation method to prepare MWNT/PET composites and found that the percolation threshold of the composite was 0.9 wt%. Seo *et al.* [89, 90] presented that the conductivity of CNT/PP composites was increased with increasing the CNT content and the electrical percolation threshold was formed between 1 and 2 wt% CNTs. Meincke *et al.* reported that percolation threshold of CNT/PA6 composite at CNT loading 4~6 wt%. Additional complexity arises in case of semi-crystalline matrices, where it is envisaged that the dispersion of CNT is significantly affected due to the crystallization induced phase separation and subsequent rejection of CNTs by the advancing crystalline fronts [95]. On the other hand, purity and defect concentration of CNTs, dispersion and alignment of CNTs within a matrix, are also the main factors influencing the electrical properties of CNT/polymer composites.

Nevertheless, compared with CB and carbon fiber based composites, whose electrical percolation threshold are usually up to 15–20 wt% and 9–

18 wt%, respectively, CNT/polymer composites still have much lower electrical percolation threshold due to their high aspect ratio and small diameter. Therefore, in many fields, CNTs are expected to substitute CB as conductive fillers. In 2005, He *et al.* [96] reported the temperature-resistivity characteristics of MWNT/HDPE composites. They found that when temperature was between 120 and 130°C, the electrical resistivity of the 5.4 wt% MWNT-filled HDPE composite increased sharply with the increase of temperature, showing a strong PTC effect. While for CB/HDPE composite, the loading concentration of CB was nearly three times as high as that of MWNTs to obtain the PTC effect. Moreover, when temperature was above T_m of HDPE, the CB/HDPE composite exhibited obvious NTC effect. However, the NTC effect in MWNT/HDPE composites was quite small. So, CNT instead of CB based polymer composites are expected to be used in self-controlled heaters, overcurrent protectors, sensors, etc.

Besides, given high conductivity and super mechanical properties of CNT/polymer composite, they can also be exploited as EMI shielding composites, coatings for enclosures, ESD composites, antistatic materials, conductive coatings, electromagnetic absorption materials for low-observable applications, electrode materials for supercapacitor and fuel cell, etc.

### 3.2.2. Thermal properties and application of CNT/polymer composites

Many researches indicate that CNTs may be the best heat-conducting material that has ever been known. Scientists predict that the thermal conductivity can reach as high as 6600 W/mK at room temperature for a SWNT [97] and experimental value is 3000 W/mK for an isolated MWNT at room temperature [98]. It is expected, therefore, that CNTs can significantly enhance the thermal conductivity and thermal stability of some polymers at high temperature [99–102]. Wei *et al.* [101] carried out classical molecular dynamics simulations employing Brenner potential for intra-nanotube interactions and van der Waals force for polymer-nanotube interface to investigate the thermal expansion and diffusion characteristics of CNT/PE composites. The addition of CNTs was found to increase T_g and thermal expansion and diffusion coefficients in the composite above T_g. Xu

et al. [103] fabricated SWNT/PVDF composites by dispersion of SWNTs in an aqueous surfactant solution, followed by mixing with PVDF powder, filtration and hot pressing. The measured thermal diffusivity of the composites increased with increase of the SWNT content, but decreased with increasing temperature because the phonon scattering was enhanced with increasing temperature for polymers. Kashiwagi *et al.* [99] fabricated MWNT/PP composites with good dispersion, and CNTs were found to significantly enhance the thermal stability of PP in nitrogen at high temperatures. Especially, the CNT addition significantly reduced the heat release rate of PP and they were found to be at least as effective as clay/PP composite for flame-retardance.

Many efforts have also been devoted to the use of CNTs as thermal conducting fillers in polymer composites and some enhancements in the thermal conductivity are usually observed. Thostenson *et al.* [104] found that the thermal conductivity of CNT/epoxy composites increased linearly with CNT concentration to a maximum increase of 60% at 5 wt% CNTs. Liu *et al.* [105] reported an enhancement of 65% in thermal conductivity with 4 wt% CNT loading in silicone elastomer. Biercuk *et al.* [102] used SWNTs to augment the thermal transport properties of epoxy and found that epoxy loaded with 1 wt% SWNTs exhibited a 70% increase in thermal conductivity at 40 K and 125% at room temperature. But these enhanced values are still far below those predicted using the 'rule of mixtures' and unlike electrical conductivity, which is based on a percolation mechanism, the increase in thermal conductivity with increasing CNT concentration is nearly linear in many cases, while in practice this has been observed to be nonlinear in certain cases. The important reasons probably rest in lower intrinsic thermal conductivity of the CNTs used, scattering of phonons by interactions with the surroundings or with defects. An alternative or probably main reason is that high resistance at the CNT-matrix or CNT-CNT interfaces limits thermal transport along percolating networks of CNTs and this leads to a rapid increase in the overall thermal resistance.

In order to investigate the effect of interface thermal resistance at CNT-matrix, which may be due to the weak bonding between polymer matrix and CNT walls, many numerical simulations have been carried out to predict the interfacial heat flow

between the CNTs and the matrix. Shenogin *et al.* [106] studied the effect of thermal boundary resistance on the heat flow in CNT composites. They found that the effective thermal conductivity of CNT/polymer composites was limited by interface resistance. A theoretical model was proposed by Huxtable *et al.* [107] to evaluate the thermal conductivity of the composite with interface resistance, and it was found that the interface played an important role in the thermal conduction through CNT composites. Nan *et al.* [108] developed a simple formula for the thermal conductivity enhancement in composites by incorporating the interface thermal resistance with an effective medium approach, and found that a large interface thermal resistance across the CNT-matrix interface caused a significant degradation in the thermal conductivity enhancement, even for the case with ultrahigh intrinsic thermal conductivity and aspect ratio of the CNTs embedded. On the other hand, the relationship between thermal conductivity and functionalization of the surface of CNTs are also be predicted by many groups because surface modification to CNTs is a possible approach to improve the interfacial adhesion and then reduce the interfacial thermal resistance between the CNTs and the matrix. However, many calculations indicate that the interface thermal resistance has a smaller effect on the overall conductivity of the composites, which is especially sensitive to the length and diameter of CNTs [109, 110]. Consequently, some experiments are designed to investigate the functionalized CNTs on the thermal conductivity of composites. Liu *et al.* [111] prepared the composites of the polydimethylsiloxane rubber with 2 wt% chemical-treated CNTs to investigate the influence of chemical modifications and defects on the thermal conductivity of CNT composites. This study reveals that moderate chemical modifications are beneficial to improve the thermal conductivity of CNT composites. However, their negative effects to the thermal conductance are more remarkable than the theoretical anticipation. Figure 4 shows the measured thermal conductivity of epoxy matrix composites with different fillers and content [112]. It can be seen that the largest improvement is non-functionalized CNT composites. These experimental results substantiate the requirement of a weak interfacial adhesion, since the composites containing amino-functionalized CNTs show relatively lower thermal conductivity compared to the corre-
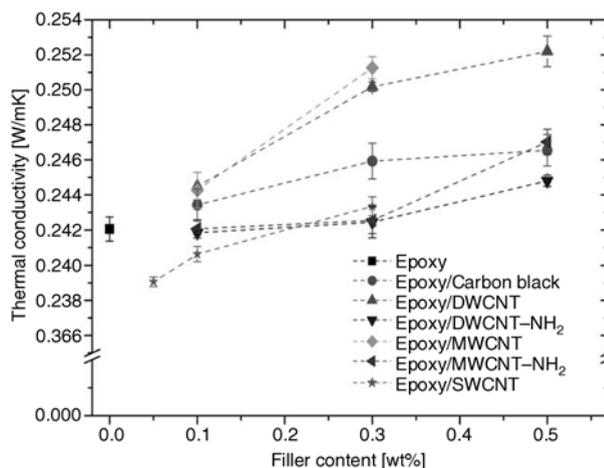

Figure 4. Thermal conductivity as a function of filler content for epoxy composites[112]. Reprinted with permission from elsevier

sponding composites with non-functionalized CNTs. Based on these theoretical and experimental results, it is obvious that to reduce interfacial thermal resistance by functionalization of CNTs is not preferable, and some new techniques therefore should be developed.

To avoid the effect of interface thermal resistance between the overlaps in CNT passage, an ideal CNT composite structure (TIM Model) was proposed by Huang *et al.* [113] to take advantage of the high thermal conductivity of CNTs. In their structure, all the CNTs embedded in the matrix were aligned from one surface to the opposite side with all the CNT tips revealed on both surfaces. According to this model, the aligned CNT composite film was prepared by *in situ* injection molding method. The enhanced thermal conductivity as a function of CNT loading for both aligned CNTs and randomly oriented CNTs are plotted in Figure 5. It can be seen that with only a small fraction of aligned CNTs (0.3%), the enhanced value of thermal conductivity is fairly high (0.65 W/mK). However, from the inset of Figure 5, it can be seen that there is only a marginal enhancement using the same loading of randomly oriented MWNTs, and the enhanced value is only 4.6% of that of an aligned CNT array. Obviously, the new structure is very effective to improve the thermal conductivity of CNT composites.

The prominent thermal properties of CNTs make them promising materials for future application as thermal management materials, such as TIMs, which are commonly used to fill air gaps between two surfaces of electronic components and base
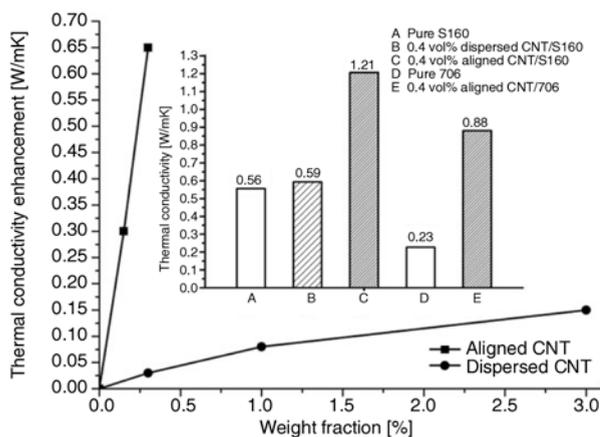
Figure 5. The enhanced values of thermal conductivity of CNT/polymer composites vs. weight fractions of CNTs [113]. Reprinted with permission from Wiley-VCH

plates of heat sinks, and thus minimize the thermal contact resistance between them and dissipate heat efficiently [114]. Based on the ideal TIM model Huang *et al.* [113] predicted that if the volume fraction of CNTs can be as high as 10% in the composites, the value of thermal conductivity would increase to an order of magnitude higher than that of most current commercial materials. They can also be used as temperature sensors, resistance heating and flame-retardance materials, etc.

3.2.3. Other functional properties and application of CNT/polymer composites

Beside electrical and thermal properties, optical and many other functional properties of CNT/polymer composites, are also fascinating. Jin *et al.* [115] studied the nonlinear optical properties of MWNT/PPA-C16TAB composite with 532 nm nanosecond laser pulses and obtained an effective nonlinear refractive index of $6.8 \cdot 10^{-18}$ m²/W. Similarly, a value of $1.2 \cdot 10^{-17}$ m²/W was obtained for MWNT/PEO composite [116]. They predicted that MWNT/PPA-C16TAB composite could be a potential candidate for solid nonlinear optical devices because of its ease to prepare a cast film. Davey *et al.* [63] prepared MWNT and SWNT/PmPV composites, and found that SWNTs could be used to enhance the luminescent properties of the conjugated polymer PmPV. The enhancement of luminescence and thus an increase in quantum yield opens up the area of luminescent polymers. Davey *et al.* [63] also proposed that the main contribution of CNTs to optical nonlinearity would

arise from 1-D polarization of delocalised π-electrons in a fixed lattice configuration. They found that MWNT/PmPV composite showed a strong nonlinear signal, which is very applicable in the area of high performance nonlinear optical devices. Chen *et al.* [117] reported that SWNT/polymer composites, especially SWNT/PI composite, not only had an optical decay time of less than 1 ps, but also a high third-order nonlinear polarizability, which makes the composite a promising material for high-quality subpicosecond all-optical switches. A few papers have studied the electromagnetic and microwave absorption properties of polymer composites reinforced with CNTs. Fan *et al.* [118] studied electromagnetic and microwave absorption applications of CNT/PET, PP, PE and varnish composites in the range of 2–18 GHz, and found that 4 wt% CNT/PET and 8 wt% CNTs/varnish composites had considerable absorbing peak at 7.6 GHz, 15.3 GHz and achieved maximum absorbing value of 17.61 dB, 24.27 dB, respectively. In addition, the frequency ranges for absorbing values exceeding 5 dB of CNT/(PET, PP, varnish) composites are 13 GHz, 10 GHz and 6 GHz, respectively. The microwave absorption between 2 GHz and 18 GHz can be mainly attributed to the dielectric loss rather than magnetic loss. The results indicate that these composites are good candidates as microwave absorption materials.

Recently, Wei *et al.* [119] synthesized PANI nanotubes doped with MWNT-$(OSO_3H)_n$ through self-assembly method and Zhang *et al.* [120] developed a facile approach to noncovalently functionalize graphitized MWNTs with the water-soluble conducting polymer of SPAN. The functionalization was based on the *in situ* polymerization of aniline and MWNTs followed by sulfonation with chlorosulfonic acid in an inert solvent and by hydrolysis in water. The lengths and surface structure of MWNTs were not perturbed with the incorporation of SPAN. The SPAN functionalized MWNTs (MWNT/SPAN composite) were highly soluble in water. These reports open many new possibilities for the prospective technological applications of CNT/polymer composites in nanoelectronic and biomedical devices and sensing, etc. In addition, the addition of CNTs to the polymer matrices can also modify the electromagnetic interference shielding effectiveness, optoelectronic properties [121], thermoelectric energy conversion, etc.

4. Key problems of CNT/polymer composites

Based on the review about the properties and application of CNT/polymer structural composites and functional composites, it is obvious that CNT/polymer composites are muti-functional and are expected to be used as automotive components, sports goods, and electronic products. They also can be used in textile and aerospace industry, etc. In a word, the application of CNT/polymer composites is at an embryonic stage of development but the scope is large and growing. The present status demonstrates that the results of enhancement in performance of CNT/polymer composite are scattered, especially for CNT/polymer structural composites. Many key issues should be addressed to realize the wide applications of these advanced composites in practice.

4.1. Structure control of CNTs

It is well known that the most important factor that determines the ultimate mechanical and other functional properties of CNT/polymer composites should be the inherent properties of CNTs. Presently, a large amount of CNTs can be prepared by electric arc discharge, laser ablation and chemical vapor deposition. But the obtained CNTs are not consistent in their structure, morphology, aspect ratio, crystallinity, crystalline orientation, purity, electric conductivity and mechanical properties, etc. Therefore, the properties of resulting CNT/polymer composites will naturally be very different even if the composites are fabricated by the same method and with the same polymer matrix.

To obtain the CNTs of high purity and uniformity is one of the big issues that still impact the applications of CNT/polymer composites. The as-prepared CNTs are usually not pure and contain a lot of impurities, some of them, such as amorphous carbon and fullerenes, can hardly be completely removed from the raw materials, and the purity is difficult to be quantified. Thus, the measured physical and chemical properties of CNTs are peculiar to different research groups. In addition, the structures of the obtained CNTs always possess surface defects and even are not identical in geometrical structure, which cause the actual mechanical strength, electrical and thermal conductivity as well

as other properties far from the theoretical predictions. Furthermore, CNTs are not continuous so that the macroscopic samples will not have the same good performance as the single CNT. Therefore, to start with, the most important problem is to prepare structure-controllable CNTs with high purity, geometrical uniformity and consistently dependable high performance.

In addition, CNTs normally are randomly oriented and entangled with each other. Especially, CNTs prepared by electric arc discharge tend to form bundles. It is very difficult to break up these entangled and bundled CNTs to produce individual ones and then disperse them uniformly in polymer matrices. Therefore, a revolutionary progress of CNT application can only be realized when we know how to break up the entangled and bundled as-prepared CNT products to produce individual CNTs without damaging their properties or to directly produce individual CNTs.

4.2. Dispersion of CNTs within polymer matrix

Uniform dispersion of CNTs in polymers is also a fundamental challenge. In fact, no matter what preparation method is adopted, the main purpose is to uniformly disperse CNTs within a polymer matrix and obtain composites with intrinsic properties [59, 60, 122]. In general, several factors that influence the dispersion of CNTs in a polymer matrix have to be considered in the preparation process of CNT/polymer composites. The entangling of CNTs during growth process and the attraction between CNTs by van der Waals force make CNTs themselves tend to aggregate. So it is difficult for CNTs to be separated from one another within the polymer matrix in mixing stage. These factors prevent CNTs from being well dispersed within polymers. The obtained CNT/polymer composites are poor in their mechanical performance and the intrinsic properties of the composites cannot be achieved.

Presently, there are two main kinds of surface treatment for CNTs to improve their dispersion within a polymer matrix. One is surface functionalization: Some functional groups, which can improve the interaction between CNTs and polymer matrix, are covalently bonded directly to the surface of CNTs. Gojny *et al.* [19] treated MWNTs with oxidizing inorganic acids. The oxidized CNTs were embed-

ded in epoxy resin and the resulting composite was investigated by TEM. The results indicated that through the treatment with oxidizing inorganic acids, the outer shells of CNTs were damaged to some extent and carboxylic groups were formed on the surface, which enables a better dispersion of CNTs. However, surface functionalization may disrupt the bonding between graphene sheets, and thereby reduce the properties of functionalized CNTs in the final composites. Another method is to have CNTs physically coated by some surfactants, which is a non-covalent approach and may be a more facile and practical processing technique. Gong *et al.* [63] added some surfactants in CNT/epoxy composites to assist the dispersion of CNTs. Experimental results showed that CNTs were dispersed more evenly with the aid of the surfactant, and the elastic modulus of the composites was increased by more than 30%. Nevertheless, CNTs treated by different surfactants may decrease the contact between CNTs, and thereby reduce the conductive properties of CNT/polymer composites.

In recent years, an interesting and successful non-covalent measure to obtain well-dispersed CNT/polymer composites was developed by establishing 'π-cation'. Bellayer *et al.* [123] prepared well-dispersed MWNT/PS composites by melt extrusion, using trialkylimidazolium tetrafluoroborate-compatibilized MWNTs. Evidence for a 'π-cation' interaction between the CNT and imidazolium was found. In addition, the use of imidazolium salts in controlling the dispersion of SWNT was also reported [124]. Kodgire *et al.* [95] prepared for the first time CNT/PA6 composites through specific interactions between Na-AHA, which assisted debundling the MWNT and leading to homogeneous dispersion within matrix during melt-mixing. The composite fabricated by this route exhibited low electrical percolation threshold of 0.5 wt% at room temperature, the lowest reported value in this system so far. Reactive coupling was found to be a dominant factor besides 'π-cation' interaction in achieving low electrical percolation in MWNT/PA6 composites.

It is worthy to mention the other efforts of synthesizing hybrid multi-scale reinforcement such as CNTs on carbon fibers [125–131] or on different ceramic particles [132]. In this case, CNTs can be homogeneously dispersed at microscale in the matrix with the help of these microscale carbon fibers or ceramic 'vehicle' because the microscale reinforcements can be much easily handled [132].

4.3. Interfacial interaction between CNTs and polymer matrix

Achieving suitable CNT-matrix interfacial bonding that provides effective stress transfer is another critical challenge for fabrication of CNT based polymer composites, especially for CNT/polymer structural composites. There are three main possible mechanisms of load transfer from a matrix to a reinforcement. The first is the weak van der Waals bonding between the reinforcement and the matrix, which is the main load transfer mechanism for CNT/polymer composites. In this case, interfacial energies normally amount to ~50–350 mJ/m^2 [133]. The second is micromechanical interlocking, which can be marginal in CNT/polymer composites if the CNTs have atomically smooth surface. The third is chemical bonding between CNTs and matrix, which is not guaranteed in many cases.

It is possible to enhance the load transfer from a matrix to a reinforcement by different treatments to improve the mechanical performance of the composite, based on the above three main mechanisms of load transfer. The van der Waals bonding can be increased by using small size reinforcement and close contact at the interface. From this point of view, individual SWNTs well dispersed in a matrix is helpful. In the case of the micromechanical interlocking between CNTs and the matrix's molecular chains, it seems that the ideal situation is that: CNTs are strong enough and inter-connected or long enough to block the movement of the polymer chains [34, 134]. The contribution of this mechanism may reach saturation at a relatively low CNT content [134]. It is unclear at this stage whether or not this critical content depends on the CNT orientation in the matrix. The chemical bonding between CNTs and matrices can be enhanced or created by surface treatments such as controlled oxidization, surface functionalization and physical coating. The question is whether or not such surface modification induces damage to the mechanical properties of the CNTs. The surface modification of the oxidized MWNTs was achieved by refluxing MWNTs with multi-functional amines by Gojny *et al.* [19] The TEM images of functionalized CNTs in the epoxy resin showed that CNTs were completely covered by the matrix. Telescopic pull-outs showed

the outermost layer, which was directly bonded to the matrix, remained in the matrix, while the inner-tubes bridged the crack. All these results substantiate the evidence of improved interaction between CNTs and matrix.

Molecular dynamic simulation results given by Frankland *et al.* [135] showed that chemical cross-links between SWNTs and polymer matrix could increase the shear strength of the SWNT-polymer interface by over an order of magnitude in comparison to the weak nonbonded interactions. Wagner *et al.* [136] examined stress-induced fragmentation of MWNTs in a polymer matrix. From the estimated values of CNT axial normal stress and elastic modulus, they concluded that the interfacial shear strength of CNT/polymer composites was at the order of 500 MPa. Wagner also calculated the interfacial shear strength of SWNT/polymer composites using a traditional force balance approach modified for a hollow tube, and examined the effect by varying some of the model parameters [137]. It was shown that high values of the interfacial shear strength (compared to those in current advanced fiber based polymer composites) were attainable. Cooper *et al.* [138] quantified the interfacial interaction between CNTs and epoxy matrix by detaching individual SWNT bundles and MWNTs from the epoxy matrix using a scanning probe microscope tip. The values for the interfacial shear strength were determined varying from 35 to 376 MPa. These results from both experimental measurements and theoretical simulations show that high values (up to several hundreds of MPa under certain conditions) of the interfacial interaction between CNTs and polymer matrices are in principle attainable.

4.4. Contacts between individual CNTs

As mentioned in sections 4.2 and 4.3, surface treatment generally is used to improve CNT dispersion within the matrix and interfacial interaction between CNTs and matrix and it is very efficient to enhance the mechanical properties of the composite. However, surface treatment is adverse to a CNT/polymer functional composite in many cases. As we know, surface functionalization may disrupt the bonding between graphene sheets, and thereby reduce the intrinsic properties of CNTs in the final composites. Also, CNTs treated by different surfactants may decrease the contact between CNTs, and

thereby reduce the final conductive properties of CNT/polymer composites. In fact, for a CNT/polymer functional composite, especially for an electric conducting composite, the effective conducting CNT path is primary, and good contact between individual CNTs is more important. An existing CNT conducting path does not guarantee good conductivity if the contact resistance between the individual CNTs is too high. Only when excellent electrical or thermal contact is established between the individual CNTs, the good electrical, thermal, wave or some other signals can be well transmitted. Several main factors may influence the contact resistance between CNTs, which finally determines the transport properties of CNT conducting path. The first is the intrinsic transport properties of CNTs, surface treatment for CNTs, therefore, is not desirable. The second is the orientation of CNTs within the matrix and the way that one CNT contacts with the others, overlap, end to end, one to one or one to more. The third is an insulating layer coating around CNT as well as its possible thickness, which is determined by the type of CNTs or polymers, the surface treatment of CNTs, and the processing method of the composites. Based on these analyses, it is possible to decrease the contact resistivity and improve the transport properties between individual CNTs by some measures. At first, high temperature treatment for CNTs to improve their intrinsic transport properties through improving their structural integrity and reducing the functional groups on their surface is preferable. To design and construct the contact means between CNTs by adjusting the content and orientation of CNTs within a matrix is also an effective measure. For example, to avoid the effect of contact resistance caused by the overlaps of CNTs in CNT passage, Huang *et al.* [113] proposed an ideal CNT composite structure, in which all the CNTs embedded in the matrix are aligned form one surface to the opposite side with all the CNT tips revealed on both surfaces. Another method is to weaken interfacial interaction between CNTs and matrix in order to decrease the interfacial interaction and then the quantity and quality of the polymer coating around CNTs during the processing of the composites [112, 139]. From this view of point, CNT/polymer functional composites may be applied in practice before CNT/polymer structural composites since it is very difficult to improve interfacial interaction between CNTs and matrix but do not damage the

intrinsic structure and the mechanical properties of CNTs.

4.5. Other problems

Besides above discussions, a few other problems exist for applied research of CNT/polymer composites. For instance, it is important to avoid interlayer slippage within MWNTs and slippage within SWNT bundles in order to fully take advantage of extra-high mechanical properties of CNTs in composites. Because interlayer slippage is an inherent structural property of MWNTs, no effective method has been found to solve this problem. This becomes even more critical when the outer layers of MWNTs are discontinuous. We know that no interlayer slippage can occur in SWNTs because they only have one shell. However, the slippage within SWNT bundles can also decrease the performance of CNT/polymer composites. Thus, isolation of individual SWNTs may be highly desirable. However, the binding energy of SWNTs in a bundle is high enough so that separation of SWNTs by polymer infiltration in the melt seems unlikely, unless some external force, such as that from ultrasonication for example, is utilized. In addition, the good CNT orientation gives rise to CNT/polymer composites with higher electrical conductivity and mechanical reinforcement, control of the alignment of CNTs towards the pre-determined direction, therefore, becomes another important issue for the design of these composites.

5. Conclusions

This article reviewed the present status of CNT/polymer composites. Based on the results reported up to now, CNTs are promising for fabricating both structural and functional composites which can be widely used in various important fields. However, it is obvious that the potential as reinforcing fillers in polymers especially for CNT/polymer structural composites have not been fully brought into play. The state-of-art indicates that great efforts should be continuously made before widely employing these advanced composites into practical usage. The critical challenges lie in how to prepare structure-controllable CNTs with high purity, geometrical identity and consistently dependable high performance, how to break up CNT products to obtain isolated CNTs, how to fab-ricate composites with uniformly dispersed CNTs, and then how to enhance load transfer from a matrix to CNT reinforcement, etc. Scientists in different fields are worming their way to improve the status by innovative chemical techniques for CNT treatment and processing techniques for CNTs and CNT/polymer composites. Nevertheless, CNT/polymer composites present significant opportunities to basic science and nanotechnology, and pose significant challenges for future work in nanocomposite field.

Glossary of abbreviation and symbols used in this article

CB	carbon black
CNT	carbon nanotube
CP-NT	conducting polymer-carbon nanotube
SWNT	single-walled carbon nanotube
MWNT	multi-walled carbon nanotube
CNF	carbon nanofiber
PA6	polyamide-6
PA12	polyamide-12
PANI	polyaniline
PE	polyethylene
HDPE	high density polyethylene
UHMWPE	Ultrahigh molecular weight polyethylene
PEO	poly(ethylene oxide)
PET	poly(ethylene terephthalate)
PI	Polyimide
PMMA	poly(methyl methacrylate)
PmPV	poly(m-phenylenevinylene-co-2,5-dioctoxy-p-phenylenevinylene)
P3OT	poly(3-octylthiophene)
PP	polypropylene
PPA-C16TAB	poly(acrylic acid)-hexadecyltrimethy-ammonium bromide
PS	polystyrene

PVA	poly(vinyl alcohol)
PVC	polyvinyl chloride
PVDF	poly(vinylidene fluoride)
SPAN	sulfonated polyaniline
TPU	Thermoplastic polyurethane
AIBN	2, 2'-azobisisobutyronitrile
Na-AHA	sodium salt of 6-aminohexanoic acid
EMI	electromagnetic interference
ESD	electrostatic dissipation
NTC	negative temperature coefficient
PTC	positive temperature coefficient
TIM	thermal interface material
K	thermal conductivity
T_g	glass transition temperature
T_m	melting temperature
LBL	layer-by-layer

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Appendix: list of published review papers related to CNT/polymer composites

[A1] Gibson R. F., Ayorinde E. O., Wen Y. F.: Vibrations of carbon nanotubes and their composites: A review. Composite Science and Technology, **67**, 1–28, (2007).

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