

## Review Article

# Recent Advances in Characterization Techniques for the Interface in Carbon Nanotube-Reinforced Polymer Nanocomposites

Junjie Chen , Xuhui Gao , and Deguang Xu 

*Department of Energy and Power Engineering, School of Mechanical and Power Engineering, Henan Polytechnic University, Jiaozuo, Henan, China*

Correspondence should be addressed to Junjie Chen; [cjj@hpu.edu.cn](mailto:cjj@hpu.edu.cn)

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The current state of characterization techniques for the interface in carbon nanotube-reinforced polymer nanocomposites is reviewed. Different types of interfaces that exist within the nanocomposites are summarized, and current efforts focused on understanding the interfacial properties and interactions are reviewed. The emerging trends in characterization techniques and methodologies for the interface are presented, and their strengths and limitations are summarized. The intrinsic mechanism of the interactions at the interface between the carbon nanotubes and the polymer matrix is discussed. Special attention is given to research efforts focused on chemical functionalization of carbon nanotubes. The benefits and disadvantages associated with covalent and noncovalent functionalization methods are evaluated, respectively. Various techniques used to characterize the properties of the interface are extensively reviewed. How the mechanical and thermal properties of the nanocomposites depend on the physical and chemical nature of the interface is also discussed. Better understanding and design of the interface at the atomic level could become the forefront of research in the polymer community. Potential problems going to be solved are finally highlighted.

## 1. Introduction

Since the discovery of carbon nanotubes and, in particular, the realization of their unique performance [1–4], much effort has been devoted to develop advanced nanocomposites with carbon nanotubes as reinforcement [5–10]. Full understanding of the excellent behavior of carbon nanotube-reinforced nanocomposites requires knowledge of the interactions at the interface between the carbon nanotubes and the matrix [11–16]. There are no essential differences between such a requirement and that for conventional fiber-reinforced composites, but the characteristic length scale of the reinforcement varies from micrometer to nanometer. The fundamental challenge for applied research on carbon nanotube-reinforced nanocomposites can be illustrated by the electron microscope images shown in Figure 1, where multiwalled carbon nanotubes (decade nanometers in diameter) are deposited on the surface of carbon fibers (a few microns in

diameter) in yarn bundles (a few millimeters in diameter) [17, 18]. The variation in the characteristic length scale of the reinforcement presents both new challenges and significant advantages in the development of fabrication techniques for carbon nanotube-reinforced nanocomposites [19–22] and the development of characterization techniques for such nanocomposites [23–28]. The nanometer scale of the reinforcement represents a significant challenge in the fundamental research of mechanics, since the interfacial interactions at the atomic-scale must be accounted for [29–32].

Various attempts have been made in recent years to develop ceramic-matrix and metal-matrix nanocomposites with carbon nanotubes as reinforcement [33–36], but much more attention has been focused on the development of carbon nanotube-reinforced polymer nanocomposites [37–40]. The outstanding physical and mechanical properties of this new form of carbon have led to significant advances in the development of such nanocomposites for

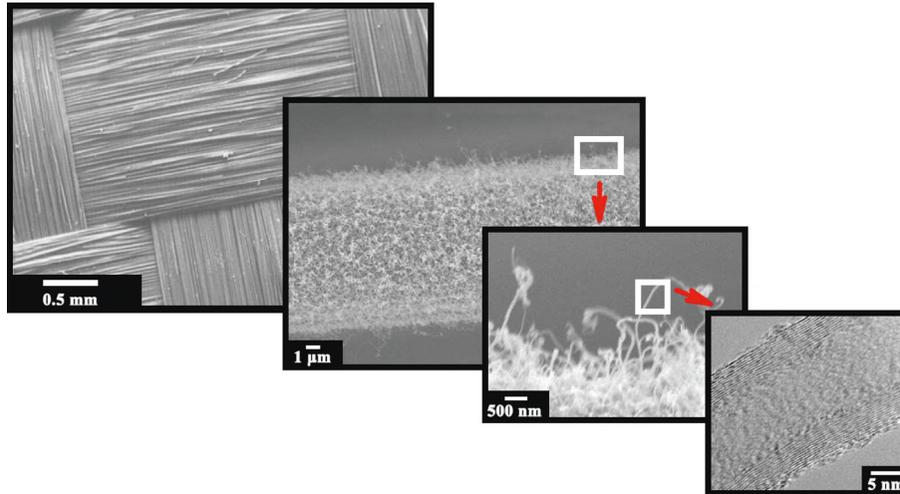


FIGURE 1: Evolution of the characteristic length scale of reinforcement from millimeters to nanometers: (from upper left to lower right corner) from woven fabric of yarn bundles, to an individual carbon fiber with multiwalled carbon nanotubes grown and entangled on its surface, to such a carbon nanotube (adapted with permission from references [17, 18]; copyright 2005, Elsevier Ltd. and copyright 2002, AIP Publishing LLC).

both functional and structural applications [41–44]. However, the potential of carbon nanotubes as reinforcement for polymers has not yet been fully realized, especially in the resulting mechanical properties [17, 20]. The application of carbon nanotube-reinforced polymer nanocomposites depends strongly on how to effectively address the issues and challenges associated with the fundamental properties of the interface between the carbon nanotubes and the polymer matrix [45, 46].

Increasing attention is focused on the properties of the interface in carbon nanotube-reinforced polymer nanocomposites. From the point of view of micromechanics, stress is transferred across such an interface [47, 48]. In addition, the mechanical performance of the nanocomposites depends critically on both the stress transfer across the interface and the shear strength of the interface [49–52]. Furthermore, the thermal resistance of the interface is the most critical factor influencing the thermal performance of the nanocomposites [53–56]. Consequently, knowledge of the properties of the interface becomes particularly essential to the development of carbon nanotube-reinforced polymer nanocomposites. To unlock the full potential of carbon nanotubes and to further improve the overall performance of such nanocomposites, it is important to understand the fundamental properties of the interface [57–60].

Research on carbon nanotube-reinforced polymer nanocomposites is an emerging area, which was first introduced by Ajayan and coworkers [61, 62]. Their work certainly represents a significant milestone towards the development of the nanocomposites and, together with other early work [63, 64], clearly demonstrates that the extraordinary properties inherent to carbon nanotubes can be transferred into the polymer matrix efficiently. Since then, the literature related to the nanocomposites is growing explosively, always accompanied by scientific debates [65–70]. Significant insights have been gained through

nanotechnology research, with over 20 papers each day currently appearing in relation to carbon nanotube-reinforced polymer nanocomposites alone, as shown in Figure 2. One of the most interesting avenues of research involves manipulation of the chemistry of carbon nanotubes [71–74], which offers great opportunities for developing multifunctional polymer nanocomposites with the ability to tailor the properties of the interface [75, 76]. As yet, the physical and chemical nature of the interface, along with its influence on the bulk properties of carbon nanotube-reinforced polymer nanocomposites, remains a poorly understood area of research.

Significant progress has been made in understanding the nature of the interface in carbon nanotube-reinforced polymer nanocomposites. Unfortunately, there are only several review articles focused on the topic available in the literature. Chen et al. [77] have provided an overview of the current state of the properties of the interface in carbon nanotube-reinforced polymer nanocomposites. However, full understanding of the nature of the interface requires knowledge of both interfacial properties and characterization techniques [78]. Both complement each other naturally, be short of one cannot, making it easier to understand the nature of the interface. It is therefore of great significance to provide a literature review about the characterization techniques for the interface, in addition to interfacial properties. This review is devoted to characterization techniques for the interface in carbon nanotube-reinforced polymer nanocomposites. Particular emphasis is placed on how to determine the properties of the interface at the nanometer scale for such nanocomposites.

## 2. Techniques for Interface Characterization

Measurement and control of the strength of the interface are important for developing carbon nanotube-reinforced polymer nanocomposites with tailored interfacial structures and

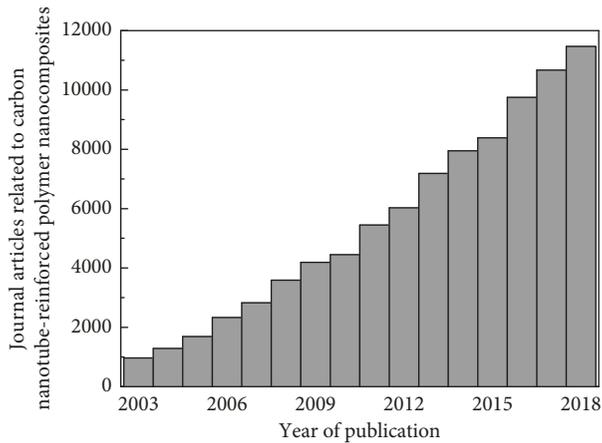


FIGURE 2: Refereed journal articles published in English related to carbon nanotube-reinforced polymer nanocomposites. Data collected from Web of Science by the end of 2018.

properties [79, 80]. To fully realize the benefits of carbon nanotubes and to further improve the overall performance of the nanocomposites, it is essential to obtain accurate information about the microscopic structure and properties of the interface [81, 82]. Electron microscope techniques such as scanning electron microscopy [83, 84] and transmission electron microscopy [85, 86] have been extensively used for the general visual identification of carbon nanotube-reinforced polymer nanocomposites. Additionally, optical microscope techniques such as confocal laser scanning microscopy are effective in evaluating the quality of the dispersion of carbon nanotube in a polymer matrix [87–90]. However, the above microscope techniques are usually not sufficient enough to characterize the properties of the interface at the atomic level. High-resolution transmission electron microscopy could achieve the atomic resolution and is being used widely in the field of the nanocomposites to provide more information about the microscopic structure of the interface [91–94].

Atomic force microscopy [95, 96] and Raman spectroscopy [97, 98] have served to quantitatively characterize the properties of the interface. Atomic force microscopy may be the preferred technique for characterization of the interface due to its advantages for high-precision and high-sensitivity measurements [99, 100]. This microscope technique can also serve as a means to elucidate the properties of the interface in polymer nanocomposites [101, 102]. Two particularly powerful approaches to characterize the properties of the interface are the pull-out method [103] and the peeling method [104]. Attempts have also been made in recent years to develop more convenient ways such as Raman spectroscopy to directly assess the properties of the interface [97, 98].

**2.1. Atomic Force Microscopy.** In developing carbon nanotube-reinforced polymer nanocomposites, it is essential to focus on the properties of the interface, as they determine the efficiency of interfacial load transfer [41–44] and can significantly influence the resulting mechanical performance

[1–4]. The strength of the interface can serve as a quantitative indicator in evaluating the mechanical properties of the nanocomposites. This physical parameter is usually measured through individual carbon nanotube pull-out tests using atomic force microscopy [99, 100]. Individual carbon nanotube pull-out tests are ideal measurements to quantitatively characterize the interfacial load transfer between carbon nanotubes and the polymer matrix. Such experiments involve the use of the probes of atomic force microscopy *ex situ*, or are carried out *in situ* within a chamber of scanning electron microscopy. Based on such nano-pull-out test technology, several nanomechanical methods are being developed and tested for the nanocomposites [58, 99, 103, 105–112]. For example, carbon nanotubes can be bonded to the probe of an atomic force microscopy, embedded in a polymer melt, and then pulled out from the matrix after solidification [103, 105, 106], as shown in Figure 3. Partially embedded carbon nanotubes were observed on the fatigue fracture surface in a polymer matrix and pulled out by a nanomanipulator [58, 99, 107].

Barber et al. [103, 105, 106] attached an individual multiwalled carbon nanotube to the sharp tip of an atomic force microscope. The carbon nanotube was then pulled from a polymer matrix, while the critical force necessary to remove such a carbon nanotube was recorded by the atomic force microscope. Shear strength of the interface [103] and fracture energy for the interface [105] were predicted by measuring the critical pull-out force and the carbon nanotube embedment length. Poggi et al. [113, 114] measured the interfacial adhesion between specific functional groups and the outer surface of an individual single-walled carbon nanotube using a chemically modified chemical force microscope, where the tip of the cantilever of an atomic force microscope was chemically modified. The interaction characteristics of the interface were then measured using this chemical force microscopy [113, 114].

However, there are still some critical issues that need to be addressed before performing such an individual carbon nanotube pull-out test using atomic force microscopy [108]. Images may be limited by misalignment during the cantilever-tip assembly load. Additionally, the intensity of the force signal recorded by atomic force microscopy is based on the assessment of cantilever stiffness as well as the determination of cantilever deflection from low-resolution scanning electron microscope images, both of which may eventually lead to considerable errors [103, 105, 106]. Finally, the embedment depth of an individual carbon nanotube cannot be controlled and measured easily using this nano-pull-out test technology. To observe the embedded portion of such a carbon nanotube after the process of a nano-pull-out test, Nie et al. [101] developed a carbon nanotube-polymer nanobridge structure in a polymer film, as described below.

The strength of the interface can serve as an indicator of interfacial load transfer for the nanocomposites, as it can greatly affect their effective mechanical performance. Unfortunately, inevitable entanglement or kinks of carbon nanotubes inside a polymer matrix would complicate the characterization of the interface as probed and measured by

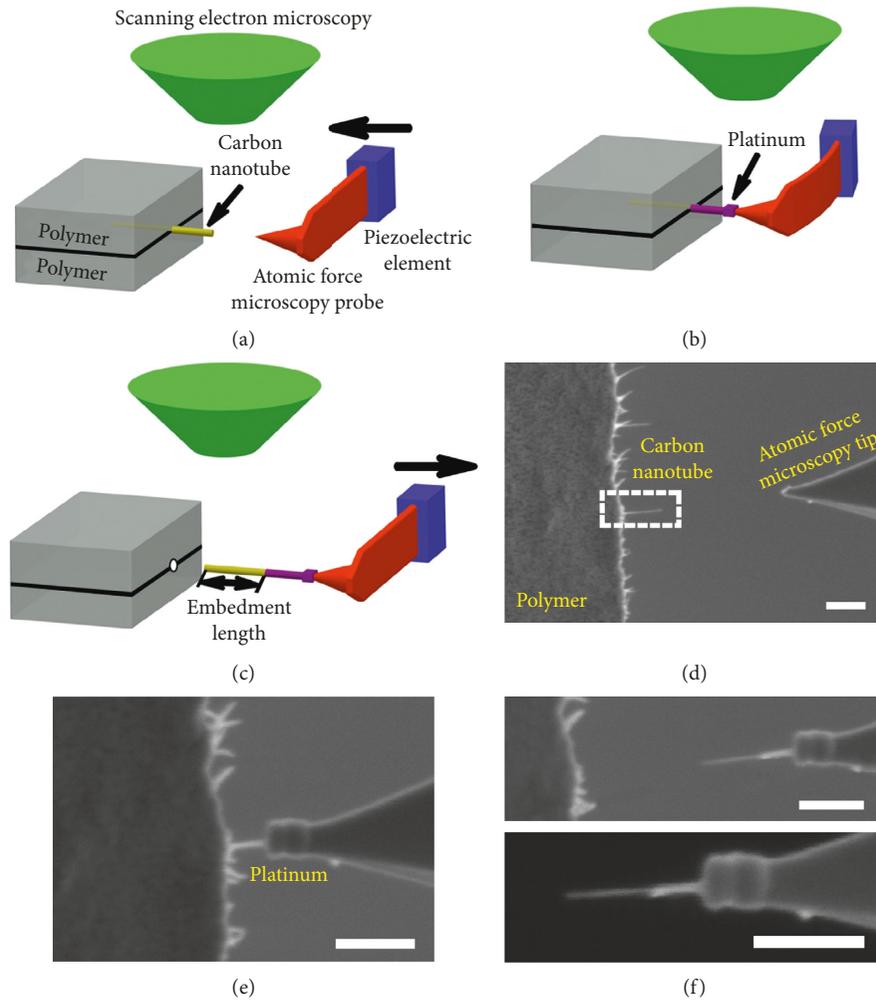


FIGURE 3: Schematic diagram of an individual carbon nanotube pull-out experiment and scanning electron microscope images of one representative individual carbon nanotube pull-out test: (a) a tip fixed to the cantilever of an atomic force microscope approaches the free end of an individual carbon nanotube embedded within a polymer matrix in a controlled way, (b) such a free end has been attached to the tip of the cantilever by means of electron beam-induced deposition of platinum, (c) the carbon nanotube has been pulled out from the polymer matrix completely, and (d)–(f) three scanning electron microscope images of the individual carbon nanotube pull-out test, corresponding to the schematic diagrams illustrated in (a)–(c), respectively. The lower right image in (f) is a zoom-in view of the carbon nanotube after being pulled out. Each of the scale bars represents 500 nm (adapted with permission from reference [99]; copyright 2015, Elsevier Ltd).

individual carbon nanotube pull-out tests. To observe an embedded carbon nanotube before or during the process of a nano-pull-out test, there is a need for a beam of electrons with high energy in a transmission electron microscope or a scanning electron microscope [115–118], which may damage or cross-link the polymer matrix and the interface. To address this issue, Nie et al. [101] developed a controllable method to measure the strength of the interface with the help of an individual nanobridge in a suspended polymer film and proposed an effective way to evaluate the efficiency of interfacial load transfer, as illustrated in Figure 4. Such a microscopic structure is constructed on the basis of the self-assembly of multiwalled carbon nanotubes in liquid flow and the phase separation of polymer blends and the ductile fracture of such blends after thermal and chemical treatment, as shown in Figure 5(a). The microscopic structure of such a nanobridge can be characterized

by a nanomechanical method, by which the force applied to the individual multiwalled carbon nanotube in a suspended state in the transverse direction leads to the pull-out force in the axial direction, as shown in Figures 5(b)–5(d). An atomic force microscope is used to apply such transverse force in a controlled way to the suspended carbon nanotube to produce the force in the axial direction. Such axial force can be applied to pull a segment of the embedded part of an individual carbon nanotube out of a polymer matrix. The pull-out force in the axial direction can be obtained through the control signal recorded by the deflection of the cantilever. The average strength of the interface formed by an individual multiwalled carbon nanotube and a poly(-methyl methacrylate) matrix is about 48 MPa, measured by using this method [101]. Such a controllable method can be used to measure the strength of the interface for the nanocomposites due to the ability to form an image of the

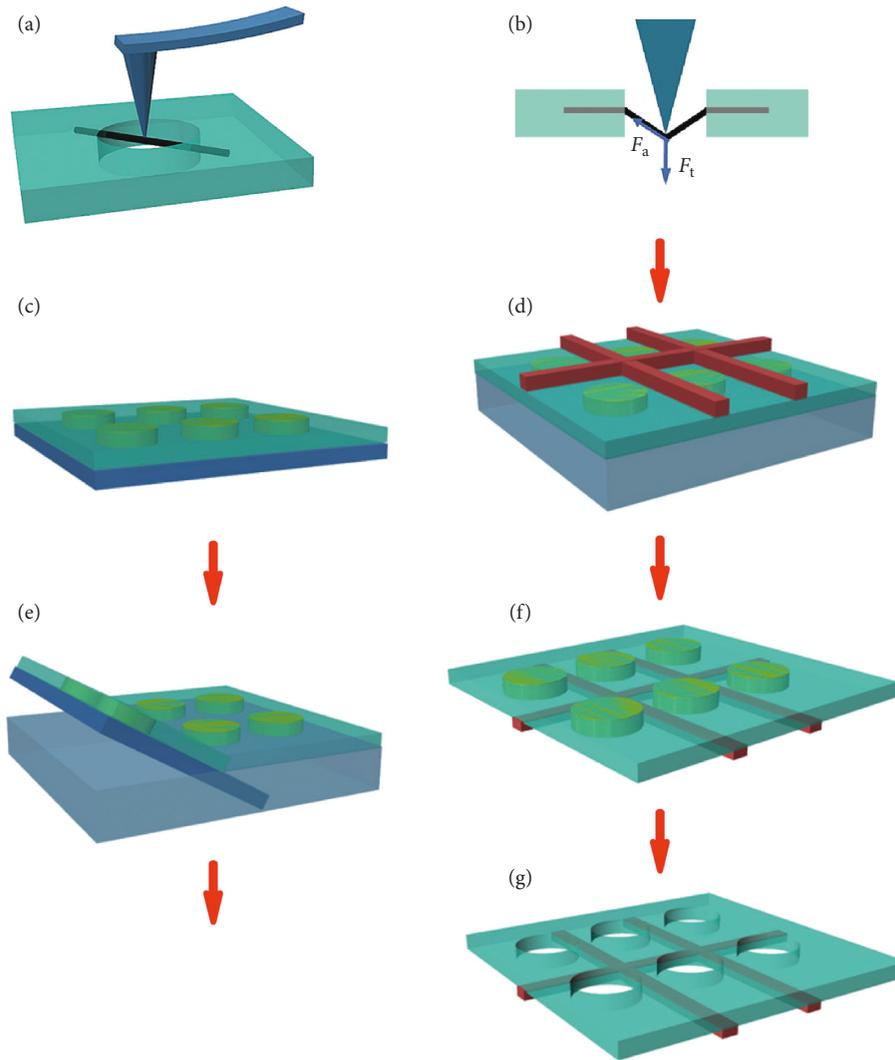


FIGURE 4: (a and b) Schematic diagram of the pull-out process applied to an individual carbon nanotube suspended within a free-standing polymer thin film using an atomic force microscope. Schematic diagram of the fabrication process of a suspended holey film as being formed from a polymer blend: (c) a hydrophobic polymer thin film coated on the surface of a hydrophilic glass slide using a dip-coating method, (d) release of such a film from the glass slide by water, (e and f) transfer of the film to a grid made of copper, and (g) hole-opening using cyclohexane as a solvent and then using a heat treatment method (adapted with permission from reference [101]; copyright 2017, Elsevier Ltd).

embedded portion of an individual carbon nanotube and to the high-resolution imaging during the loading process [101]. It is important to note that this technique is only applicable to the polymers with the ability to form a phase separation structure.

Atomic force microscopy also provides an efficient way to gain insight into the adhesive mechanics of an individual carbon nanotube peeled from a polymer matrix by means of the peeling method described in the literature [104]. Strus et al. [104] used such a method to attach an individual multiwalled carbon nanotube to the peeling probe of an atomic force microscope, as illustrated in Figure 6. The critical force required to peel the multiwalled carbon nanotube off the surface of a polymer matrix was recorded, and the fracture energy of the interface was then quantitatively measured. This method has the ability to measure the

properties of the interface, but it is incapable of measuring the interfacial energy per unit area [104].

**2.2. Raman Spectroscopy.** Raman spectroscopy can serve as a powerful tool to characterize the properties of the interface by utilizing the unique electronic and vibrational structure of carbon nanotube-reinforced polymer nanocomposites [119–130]. This spectroscopic technique was found to be more effective in revealing the level of disorder and defect sites on the surface of carbon nanotubes [129, 130]. Raman spectroscopy is one of the most effective techniques used to analyze the microscopic mechanism of interfacial load transfer for the nanocomposites [63]. There is a shift in Raman frequency with the stress or strain applied to the nanocomposites [131, 132]. If such a stress is transferred to

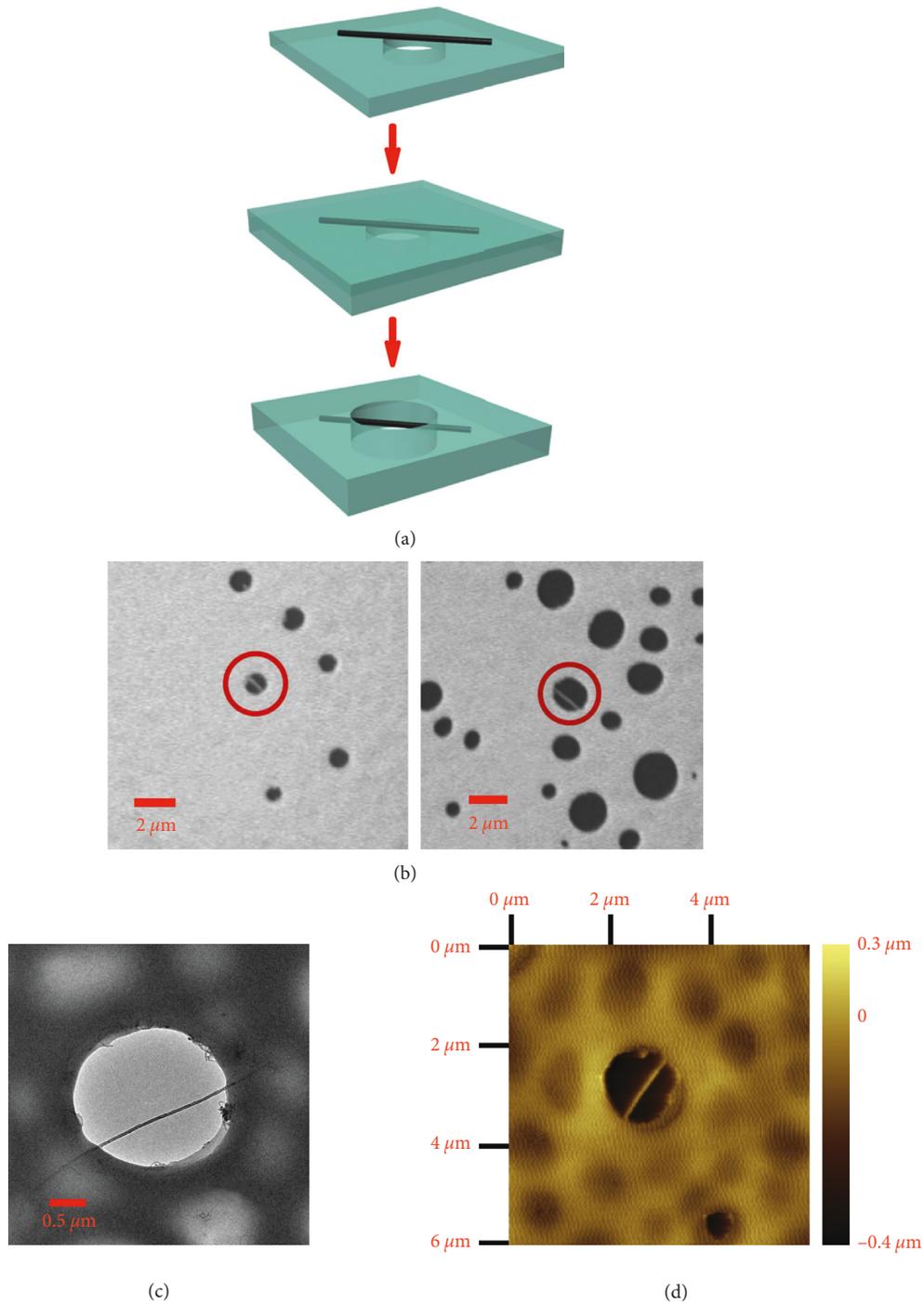


FIGURE 5: (a) Schematic diagram of the formation and evolution process of a nanobridge structure for an individual carbon nanotube-polymer nanocomposite, (b) scanning electron microscope images of such a carbon nanotube suspended across a hole in a layer of the polymer blend film made of poly(methyl methacrylate) and polystyrene using a self-assembly method (left) and the holes opened by using thermal and solvent treatment techniques after adding a second layer of poly(methyl methacrylate) film (right), (c) transmission electron microscope image of the nanobridge structure in a polymer film, and (d) atomic force microscope image of an individual, suspended carbon nanotube with two ends embedded in a poly(methyl methacrylate) matrix (adapted with permission from reference [101]; copyright 2017, Elsevier Ltd).

the carbon nanotubes across the interface, the Raman peak shifts by a few  $\text{cm}^{-1}$ . The shift in Raman frequency is linear with the strain applied to the nanocomposites, and the slope of the Raman shift-strain response curve is dependent on the

properties of the polymer matrix as well as the degree of alignment of carbon nanotubes [133, 134].

Raman spectroscopy can provide information about the elastic modulus of polymer-based nanocomposites, alignment

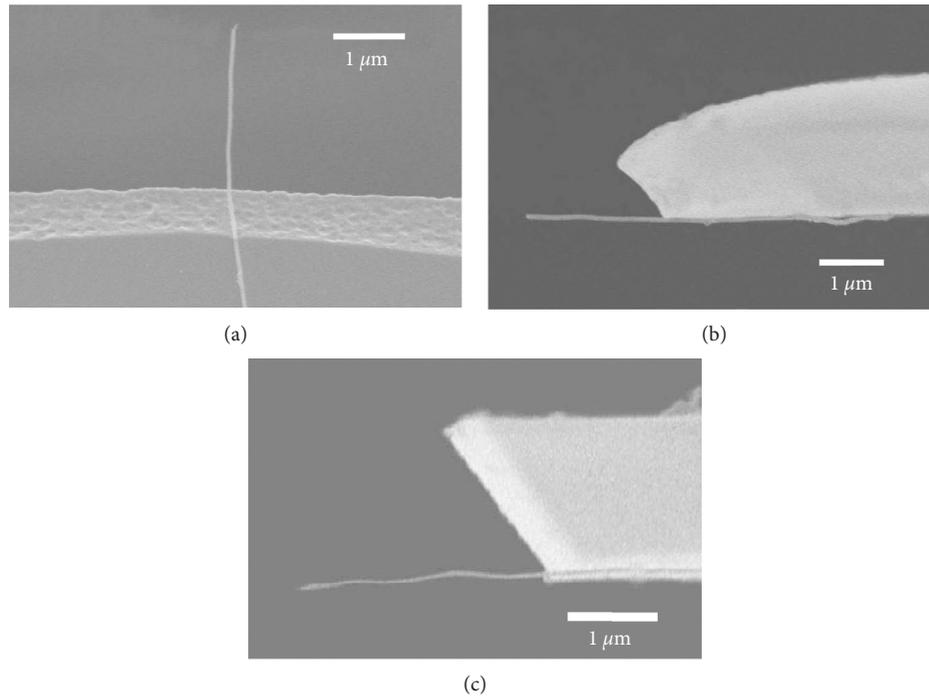


FIGURE 6: Scanning electron microscopy images of the (a) bottom and (b) side of a peeling atomic force microscope probe with a length of  $2.2\ \mu\text{m}$  and of the (c) side of a peeling atomic force microscope probe with a length of  $2.5\ \mu\text{m}$ . The individual carbon nanotube used to attach such probes is  $10 \pm 2\ \text{nm}$  in inner diameter and  $40 \pm 6\ \text{nm}$  in outer diameter, respectively (adapted with permission from reference [104]; copyright 2009, Elsevier Ltd).

and dispersion of carbon nanotubes, interfacial load transfer, and interfacial interaction characteristics [135–140]. Such interactions are usually reflected by a shift in Raman frequency or a peak width change [91, 141]. However, there are some disadvantages associated with this spectroscopic technique [135–140]. For example, since the diameter of the Raman laser spot is much larger than that of an individual carbon nanotube, the Raman signal is averaged over carbon nanotubes in all directions [91, 141].

Earlier publications associated a shift in Raman frequency to a lower frequency or to a higher frequency from the peak about  $2700\ \text{cm}^{-1}$  with tensile strain and compressive strain in a carbon nanotube-reinforced polymer nanocomposite [64, 142]. Ruan et al. [143, 144] monitored load transfer across the interface from an ultrahigh molecular weight polyethylene matrix to multiwalled carbon nanotubes, with a shift in Raman frequency at  $2691\ \text{cm}^{-1}$ . They also suggested four regimes of strain behavior, as outlined in Table 1. Another work [145] related a shift in Raman frequency at  $1594\ \text{cm}^{-1}$  with the compressive strain in single-walled carbon nanotubes embedded in an epoxy matrix.

### 3. Interactions at the Interface

It is of great significance to understand the interactions at the interface [146–149]. Knowledge of such interactions is necessary to gain insight into the properties of the nanocomposites [146]. An optimum interfacial interaction is a critical step in achieving the full potential of carbon nanotubes [147], which has become a primary goal of this emerging area of research [146–149].

Chemical functionalization of carbon nanotubes has proved effective in improving the characteristics of interfacial interactions and the performance of the resulting nanocomposites [150–153]. Chemical functionalization of carbon nanotubes, such as wrapping of polymer around carbon nanotubes [154], as illustrated schematically in Figure 7, or introducing covalent interfacial bonding [155], has been achieved. In general, covalent and noncovalent interfacial interactions [156–159] are the two main chemical functionalization methods.

**3.1. Covalent Interactions.** Covalent chemical functionalization of carbon nanotubes involves the covalent attachment of functional groups onto either the sidewalls or ends of carbon nanotubes [160, 161]. The open ends of carbon nanotubes can be stabilized by hydroxide and carboxylic acid groups, and the sidewalls of carbon nanotubes can be chemically modified by reactive elements [42]. For example, nitric acid treatment was reported to successfully oxidize the surface of carbon nanotubes as detected by Fourier transform infrared spectroscopy [162, 163]. Covalent chemical functionalization of carbon nanotubes can also be accomplished through diazonium salts [164] or Diels–Alder reactions [165].

Recently, an extensive review of various covalent functionalization methods of nanoparticles by means of tailoring surface properties to improve their wettability and compatibility with polymer matrices has been made by Mosnáčková et al. [166]. Different methods of surface modification by polymer chains have been described in

TABLE 1: Shift in Raman frequency ( $2691\text{ cm}^{-1}$ ) as a function of the tensile strain applied to an ultrahigh molecular weight polyethylene nanocomposite reinforced with multiwalled carbon nanotubes.

Regime*	Tensile strain	Shift	Interpretation
Regime (i): elastic region	0–1%	Clear shift to a lower frequency	Elastic response; tensile loading of multiwalled carbon nanotubes
Regime (ii): viscoelastic and plastic deformation occurs	1–10%	Much less apparent shift to a lower frequency	Slip and stick at the interface
Regime (iii): strain hardening occurs	10–15%	Somewhat more apparent shift to a lower frequency	Multiwalled carbon nanotube knots preventing further stretching of the chain of the polymer; tensile loading of multiwalled carbon nanotubes
Regime (iv): partial failure occurs in the polymer matrix at the microscopic level	Larger than 15%	Shift to a higher frequency	Elastic recovery from local failure occurred in the polymer matrix; compressive loading of multiwalled carbon nanotubes

\*Four regimes of behavior are available for a shift in Raman frequency when different tensile strains are applied to an ultrahigh molecular weight polyethylene nanocomposite reinforced with multiwalled carbon nanotubes. Interpretation of data is available in reference [143].

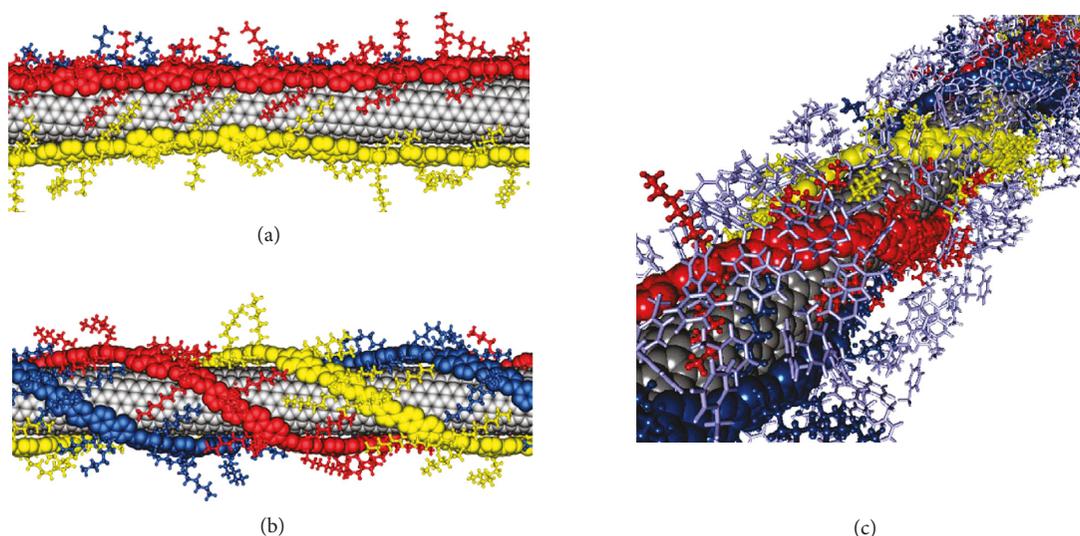


FIGURE 7: Single-walled carbon nanotube wrapped by three poly(9,9-dioctylfluorenyl-2,7-diyl) chains with different geometries: (a) polymer chains aligned to the axis of the carbon nanotube and (b) periodic helical conformation. The polymer chains are represented by different colors: red, blue, and yellow. (c) The side chains of the polymer covered by the solvent system that contains enough toluene molecules. The toluene molecules are represented by light gray (adapted with permission from reference [154]; copyright 2011, American Chemical Society).

detail with some examples of functionalization of various types of nanoparticles to obtain advanced properties of polymer nanocomposites. Various polymerization mechanisms have been discussed in detail. The current state of various grafting methods has also been summarized. The effect of introduced functional groups on the properties of carbon nanotubes and polymer nanocomposites has recently been reviewed by Wei et al. [167].

Covalent functionalization can enhance the interactions at the interface effectively, making it possible to significantly improve the performance of the resulting nanocomposites [16, 42, 49]. Gojny et al. [168, 169] gave direct evidence of significant interactions existed at the interface, as illustrated

in Figure 8. Amino acids were successfully used to modify carbon nanotubes by heating the oxidized carbon nanotubes with excess triethylenetetramine. Their works clearly demonstrated that a covalent bond was formed at the interface through introducing functional groups [168, 169]. The chemically modified carbon nanotubes were completely covered with an epoxy matrix, as indicated by the transmission electron microscope images of functionalized carbon nanotubes in the polymer matrix in Figure 8. Upon the expansion of the crack formed in the polymer matrix heated by a beam of electrons from a transmission electron microscope, the outer layer of an individual, functionalized multiwalled carbon nanotube was fractured and embedded

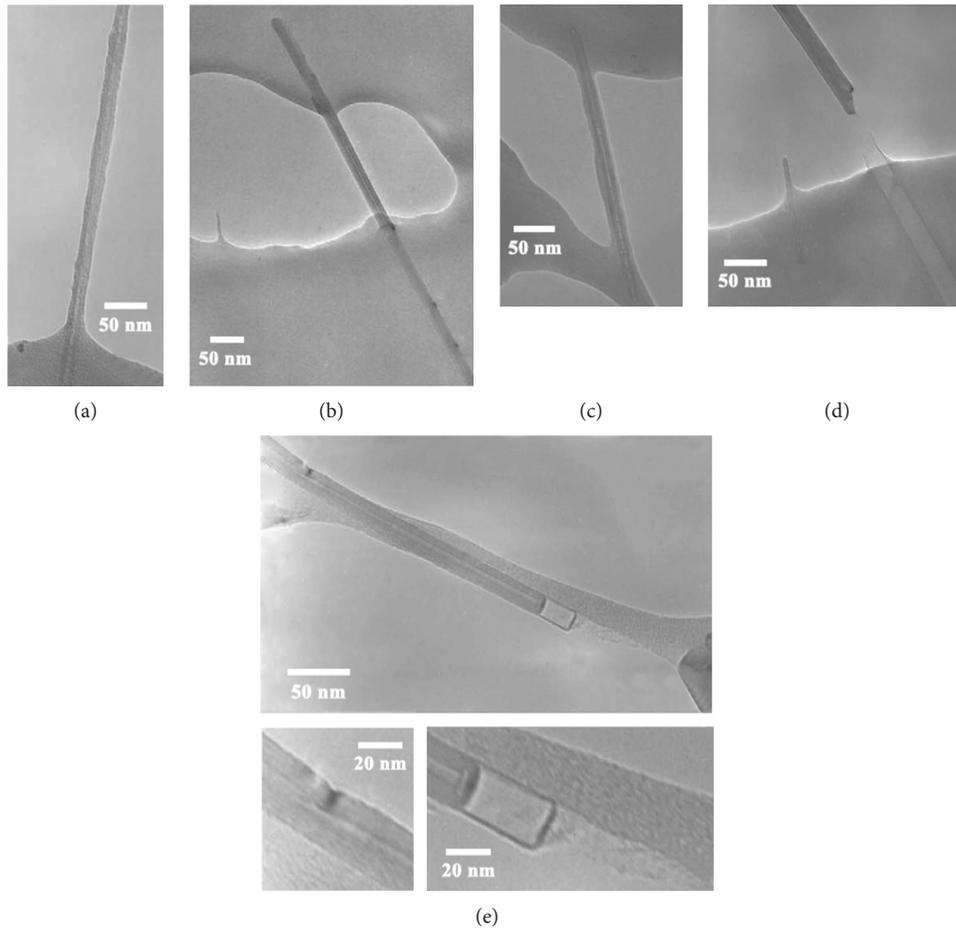


FIGURE 8: Transmission electron microscope images of an individual, functionalized multiwalled carbon nanotube in an epoxy matrix. (a) Chemically modified carbon nanotube covered by the polymer matrix completely, indicating improved interactions at the interface through a covalent chemical functionalization method. (b and c) Fracture toughness improved by such carbon nanotubes through bridging microcracks and pores in the polymer matrix. (d and e) Evidence of improved interactions at the interface through pull-out tests (adapted with permission from references [168, 169]; copyrights 2003 and 2004, Elsevier Ltd).

in an epoxy matrix, as shown in Figure 8(d). In contrast, the inner shells of the functionalized multiwalled carbon nanotube, which were bounded together by the relatively weak van der Waals forces between these shells, were pulled out from the polymer matrix through a sword-in-sheath mechanism [168, 169].

Covalent chemical functionalization has been proved to be an effective way for improving the properties of the interface [168, 169], such as the efficiency of interfacial load transfer, making it possible to significantly improve the properties of carbon nanotube-reinforced polymer nanocomposites [168, 169]. However, this functionalization method would introduce structural defects into the surface of carbon nanotubes, thus lowering their intrinsic properties [170, 171]. As a consequence, there is often a tradeoff between the properties of carbon nanotubes and those of the interface. In addition, covalent functionalization may cause severe disruption of the electronic network and structure of carbon nanotubes [170, 171], which may lead to decreased performance of the final nanocomposites.

**3.2. Noncovalent Interactions.** Alternatively, noncovalent functionalization of carbon nanotubes can serve to improve the properties of the interface. This functionalization method utilizes  $\pi-\pi$  interactions and van der Waals forces by adsorption of polymers [172–175]. Noncovalent functionalization of carbon nanotubes is particularly attractive, since it does not deteriorate their intrinsic properties [176–179]. This functionalization method mainly involves the polymer wrapping of carbon nanotubes. Wrapping is typically denoted as the enveloping of the surface of carbon nanotubes by polymers. Three possible multihelical wrapping conformations can be found in the work of O’Connell et al. [175], as shown in Figure 9. Recent studies demonstrated that single-walled carbon nanotubes can be wrapped in a number of adsorbing polymers, such as biopolymers such as DNA [180, 181], block copolymers [182, 183], and semiconjugated polymers [184, 185].

Polymer wrapping of carbon nanotubes has proven to be quite effective in improving the properties of the interface, making it possible to greatly improve the properties of

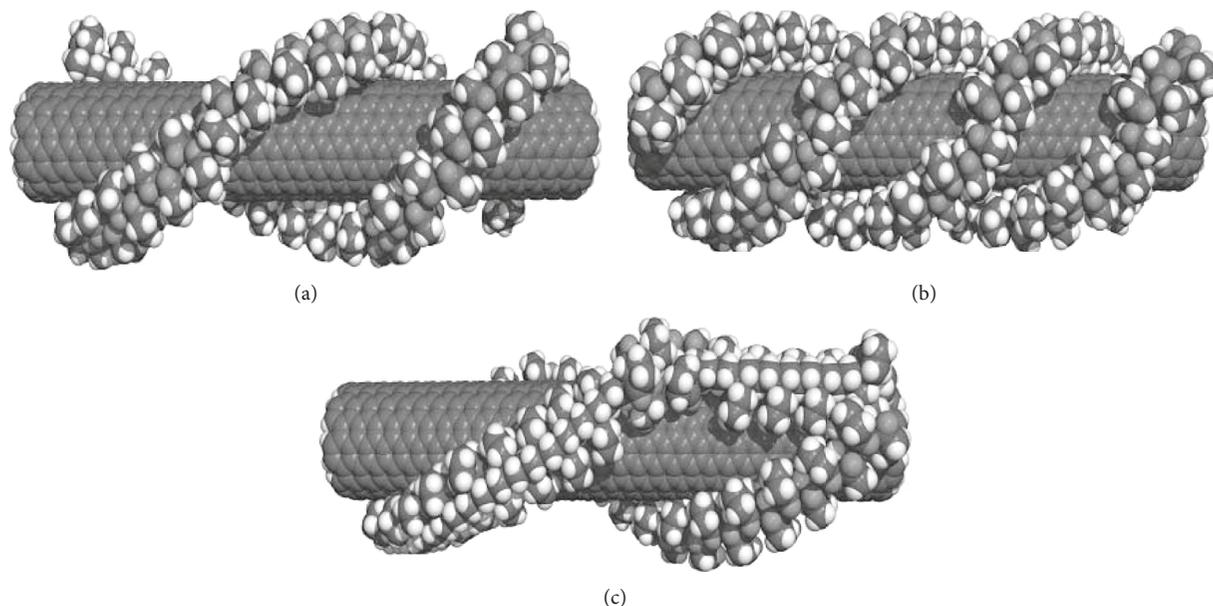


FIGURE 9: Three possible multihelical wrapping conformations of polyvinyl pyrrolidone around an individual single-walled carbon nanotube. (a) A double helix and (b) a triple helix. (c) Backbone bond rotations can cause a sharp bend, which allows multiple parallel wrapping strands to come from the same chain of the linear polymer. Low-backbone strain with high-surface area coverage is allowed for multihelical wrapping (adapted with permission from reference [175]; copyright 2001, Elsevier Ltd).

carbon nanotube-reinforced polymer nanocomposites [186, 187]. For example, interfacial interactions can be significantly improved by using conjugated polymers with a specific molecular configuration [187]. Yang et al. [187] utilized molecular dynamics to investigate the physical interactions at the interface for different polymer matrices. Their modeling work suggested that polymers with a main chain containing aromatic functionalities hold great promise for the noncovalent binding of carbon nanotubes into the structure of the nanocomposites, as shown in Figure 10, since these aromatic rings are able to provide strong interactions at the interface. Such polymers could serve as a building block for amphiphilic copolymers not only to enhance the interactions at the interface but also to improve the mechanical performance of the resulting nanocomposites. Polymer wrapping of carbon nanotubes also holds great promise for the manipulation and organization of carbon nanotubes into ordered structures and even for their noncovalent functionalization [187].

Unfortunately, the possible mechanisms underlying the noncovalent interactions have not yet been fully understood [146]. It has been found that wrapping of single-walled carbon nanotubes by polymers is a general interfacial phenomenon [175]. Furthermore, recent molecular dynamics studies suggested that such interactions depend strongly on the specific structure of polymer matrices, and the effect of polymer affinity and flexibility is important [188–193]. For example, stiff-chain polymers tend to wrap around single-walled carbon nanotubes in a more distinct conformation than polymers with a high degree of flexibility in the backbone structure [188, 189], as shown in Figure 11, making it possible to improve the interfacial properties, such

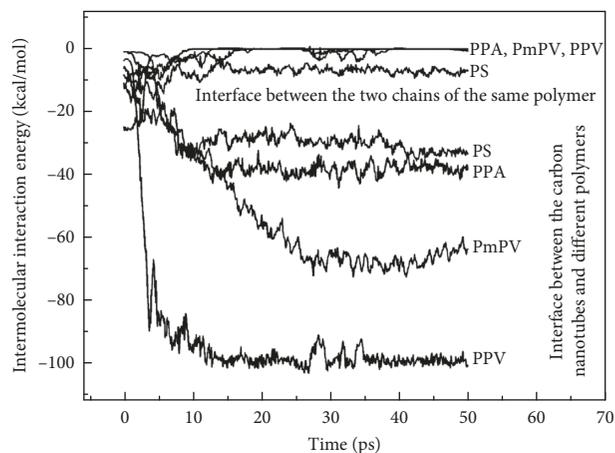


FIGURE 10: Time evolution of the intermolecular interaction energy at the interface between single-walled carbon nanotubes and different polymer matrices as well as between the two chains of the same polymer during the wrapping procedure for polystyrene (PS), poly(*p*-phenylenevinylene) (PPV), poly(*m*-phenylenevinylene-co-2,5-dioctyloxy-*p*-phenylenevinylene) (PmPV), and poly(phenylacetylene) (PPA) (adapted with permission from reference [187]; copyright 2005, American Chemical Society).

as the intermolecular interaction energy at the interface, for the resulting nanocomposites.

Noncovalent functionalization offers a number of advantages for improving the properties of the interface and the performance of the resulting nanocomposites, without affecting the intrinsic properties of carbon nanotubes [194–197]. However, it is often difficult to achieve the

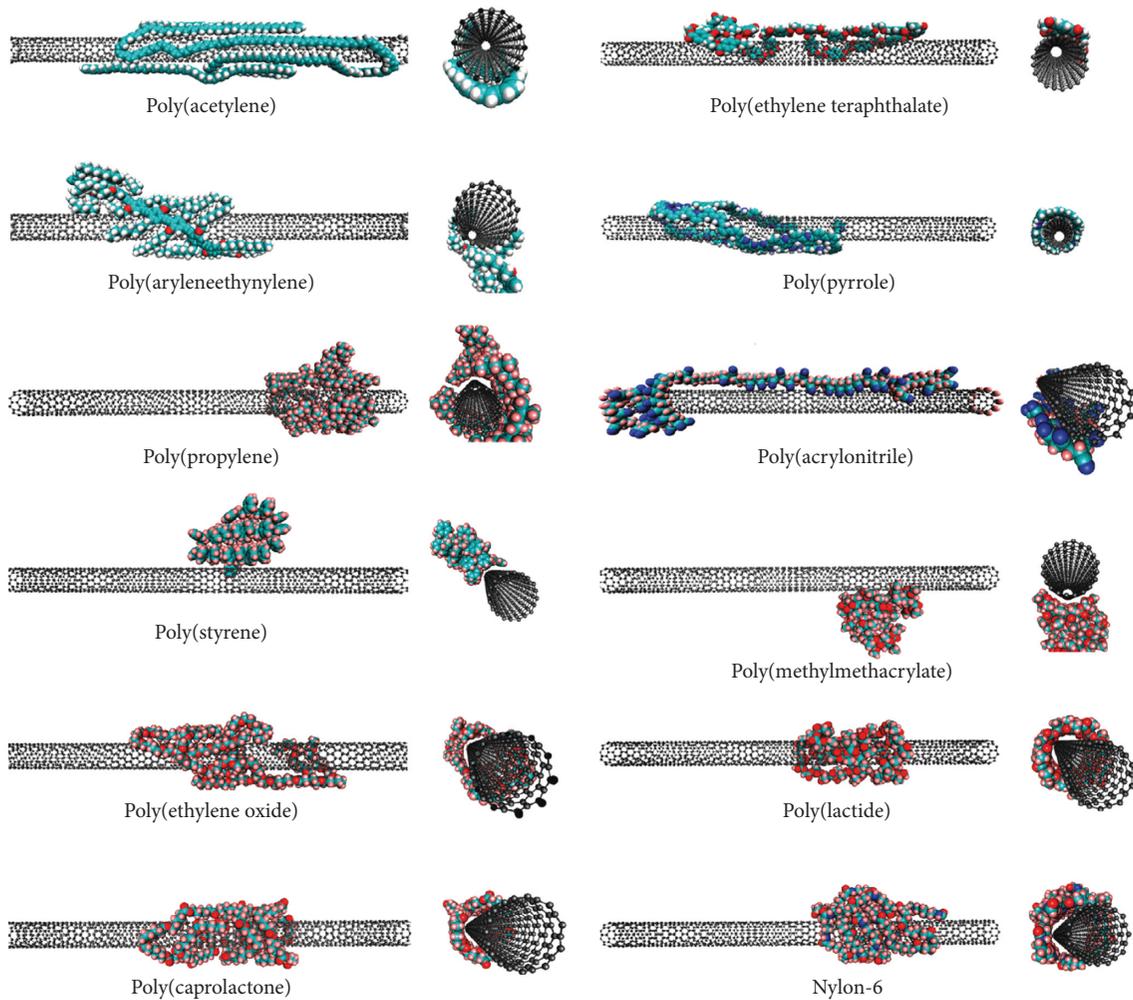


FIGURE 11: Possible wrapping conformations obtained from a model used to illustrate potential interactions at the interface between single-walled carbon nanotubes and different polymer matrices at the 3.2 ns time step in molecular dynamics simulations. The polymers have different degrees of flexibility in the structure of their respective backbone (adapted with permission from references [188, 189]; copyright 2010, American Chemical Society).

desirable strength of the interface so as to ensure the load transfer across the interface with high efficiency for carbon nanotube-reinforced nanocomposites [198–201].

#### 4. Characterization of Interfacial Mechanical Properties

The nature of the interface is of particular importance to the interfacial stress transfer and the consequent improvement in the mechanical properties for the nanocomposites [202, 203]. For example, good interfacial bonding is necessary to ensure the load transfer across the interface with high efficiency, which is a necessary condition for further development of the nanocomposites with high mechanical performance [204, 205]. Over the past few decades, it has become increasingly clear that considerable modification of the properties of the interface can be an effective way to improve the overall mechanical performance for the nanocomposites [206–209].

The work of Wong et al. [204] has shown that the interfacial shear stress measured for multiwalled carbon nanotube-reinforced polymer nanocomposites is larger than that measured for conventional microfiber-reinforced composites by orders of magnitude. Furthermore, there were no broken carbon nanotubes based on an analysis of the interfacial morphology measured by both transmission electron microscopy and scanning electron microscopy for carbon nanotube-reinforced epoxy nanocomposites [204], indicating that the load transfer across the interface was not sufficient to fracture the multiwalled carbon nanotubes. In contrast, recent experiments have demonstrated that the failure of multiwalled carbon nanotube-reinforced polymer nanocomposites appears to arise from mechanical fracture of the polymer matrix and from the pull-out of such carbon nanotubes from the polymer matrix [106, 210, 211].

Qian et al. [212] characterized the properties of the polystyrene nanocomposites reinforced with multiwalled carbon nanotubes to investigate the behavior of the load transfer across the interface and deformation mechanisms of

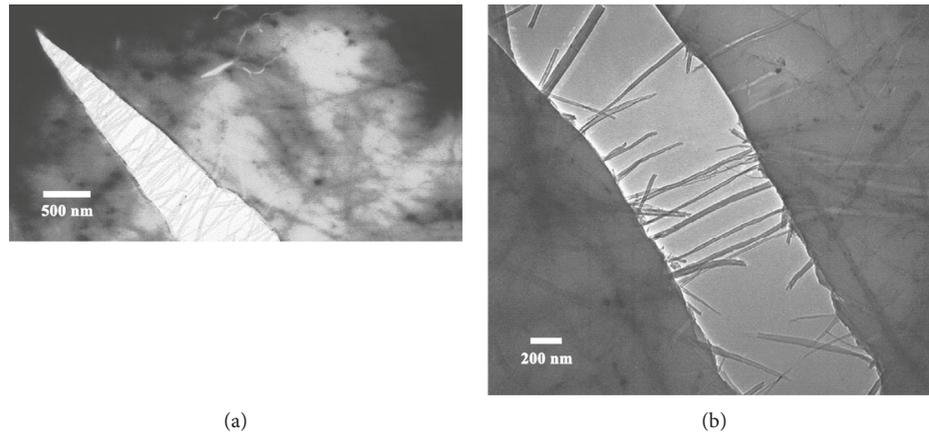


FIGURE 12: *In situ* transmission electron microscopy images of crack nucleation and propagation induced by thermal stresses in a polystyrene thin film reinforced with multiwalled carbon nanotubes. (a) Such a crack propagates along relatively low density regions of carbon nanotubes or along weak interfaces. (b) The multiwalled carbon nanotubes tend to align and bridge the crack wake, and subsequently to break and/or pull out from the polymer matrix (adapted with permission from reference [212]; copyright 2000, American Institute of Physics).

the nanocomposites. *Ex situ* mechanical tensile tests on films of the nanocomposites shown that after adding 1.0% by weight of carbon nanotubes, the tensile strength is improved by about 25.0% and the elastic stiffness is increased by about 36–42%, indicating that there are considerable load transfer across the interface and significant improvement in the mechanical performance of the polymer. In this context, an external load can be transferred from the polymer matrix to carbon nanotubes effectively. Furthermore, *in situ* transmission electron microscopy provided essential information about the interfacial bonding and deformation mechanisms of the nanocomposites described above, as illustrated in Figure 12. As for commercial carbon fiber-reinforced polymer composites, a fracture of carbon nanotubes and crack bridging by carbon nanotubes were observed during pull-out tests. In contrast, multiwalled carbon nanotubes pulled out from the polymer matrix, while less fracture was observed, as shown in Figure 12. Furthermore, there are strong interactions at the interface [212], as evidenced by the significant improvement in the mechanical performance of the nanocomposites given above. Qian et al. [212] also demonstrated that 10% by weight of carbon fibers in the previous work of Tibbetts and McHugh [213] was required to achieve the same effectiveness of the improvement in the elastic modulus of the nanocomposites containing only 1.0% by weight of multiwalled carbon nanotubes by utilizing short-fiber composite theory.

The force required to pull an individual multiwalled carbon nanotube out from a polyethylenebutene matrix was measured by atomic force microscopy [103]. The mean interfacial shear strength of 47 MPa was sufficiently high, indicating that there exists covalent bonding at the interface. Furthermore, the chain of the polymer close to the interface behaved differently from its bulk. A sheathing layer of a polymer on pulled out multiwalled carbon nanotubes, i.e., an interfacial region, was imaged [107], as shown in Figure 13, which gave direct evidence of significant interfacial interactions. In addition, an additional increase was observed in the diameter of the polymer sheathing layer after chemical functionalization of carbon

nanotubes, possibly indicating chemical augmentation of interfacial bonding. Chemically functionalized carbon nanotubes could improve the interactions at the interface [214, 215]. Orders of magnitude improvement in interfacial shear strength would be required to achieve more efficient reinforcement for the nanocomposites [216]. While optimization of the interfacial properties for the mechanical reinforcement is still poorly understood at the nanoscale, the recent evidence available clearly suggests that chemical functionalization can be effective for such reinforcement [217], as shown in Figure 14, and this method appears to be a major focus of improvement in the performance of the nanocomposites.

Strong interfacial interactions are usually necessary to ensure successful stress transfer across such an interface. An effective strategy for the improvement in the interactions at the interface is chemical functionalization by which polymer chains are attached onto the surface of carbon nanotubes. However, the unique mechanical performance of carbon nanotubes is dependent on their specific chemical structure [218, 219]. Covalent functionalization would introduce internal stresses and atomic defects into the surface of carbon nanotubes [220, 221], thereby deteriorating their inherent mechanical properties. On the other hand, noncovalent functionalization offers an alternative strategy for regulating the properties of the interface [222, 223]. Such a strategy does retain the intrinsic properties of carbon nanotubes, but, unfortunately, interfacial stress transfer might not be so effective in the treatment for the nanocomposites. It is usually difficult for this modification strategy to improve the interfacial properties efficiently [224, 225], and considerable improvements remain to be exploited.

## 5. Characterization of Interfacial Thermal Properties

The thermal resistance of the interface, also known as Kapitza resistance, plays an important role in determining

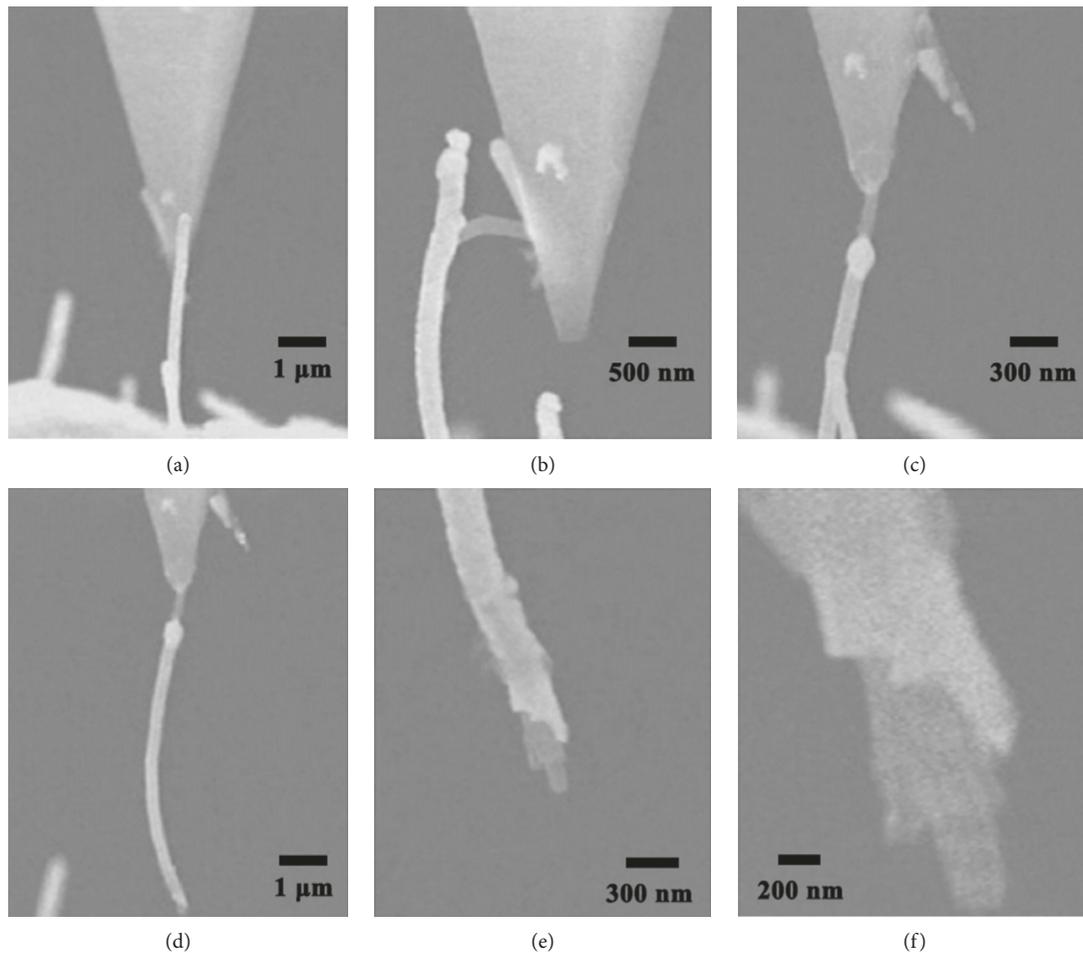


FIGURE 13: Direct evidence of multiple polymer sheathing layers in a polycarbonate nanocomposite reinforced with multiwalled carbon nanotubes, confirmed by the nanomanipulation contact experiments carried out under a scanning electron microscope. (a) Initial contact with an individual, protruding multiwalled carbon nanotube by a tip of the cantilever of an atomic force microscope. (b) The inner structure of the nanocomposite has become detached partially from the outer polymer sheathing layer in the process of bending. (c) Such a structure is clamped to a tip of the cantilever of an atomic force microscope by electron beam-induced deposition, and the outer polymer sheathing layer balls up. (d) The inner structure has been broken close to the fracture surface of the nanocomposite during tensile loading. (e) The fracture broken end of the coated multiwalled carbon nanotube provides clear evidence of a double polymer sheathing layer. (f) Higher magnification image of the fracture broken end of the coated multiwalled carbon nanotube (adapted with permission from reference [107]; Copyright 2003, American Chemical Society).

the thermal properties of the nanocomposites [226, 227]. The interfacial thermal resistance has been investigated both experimentally and theoretically [228–237]. The recent results measured and predicted for the interfacial thermal resistance of the nanocomposites are summarized in Table 2. Such resistance is typically on the order of magnitude of  $10^{-8} \text{ m}^2 \cdot \text{K}/\text{W}$ .

Since transport of heat in carbon nanotube-reinforced polymer nanocomposites is by way of elastic vibrations of the lattice, heat transfer based on this mechanism is limited by the elastic scattering of acoustic phonons at lattice defects [238, 239]. As a consequence, the interfacial thermal resistance is caused by the scattering of acoustic phonons across the interface [238, 239]. It is therefore of great significance to gain insight into such a thermal property for the nanocomposites, making it possible to further improve their overall thermal

performance [53, 54]. To address issues associated with the interfacial thermal resistance, it is usually necessary to use the method of molecular dynamics simulations to understand the interfacial thermal resistance and to evaluate the benefits and disadvantages associated with new ways to reduce such resistance [227, 229, 231].

Large thermal resistance at the interface has emerged as a significant challenge, and such a technical issue has not been addressed yet. Much effort has been devoted to estimate the interfacial thermal resistance [227], as well as to evaluate the effect of chemical functionalization and the role of interfacial interactions [240, 241]. Unfortunately, the influence of interfacial properties on the thermal conductivity of the nanocomposites has not yet been fully understood. Chemically functionalized multiwalled carbon nanotubes were found to have the potential to realize carbon nanotube-

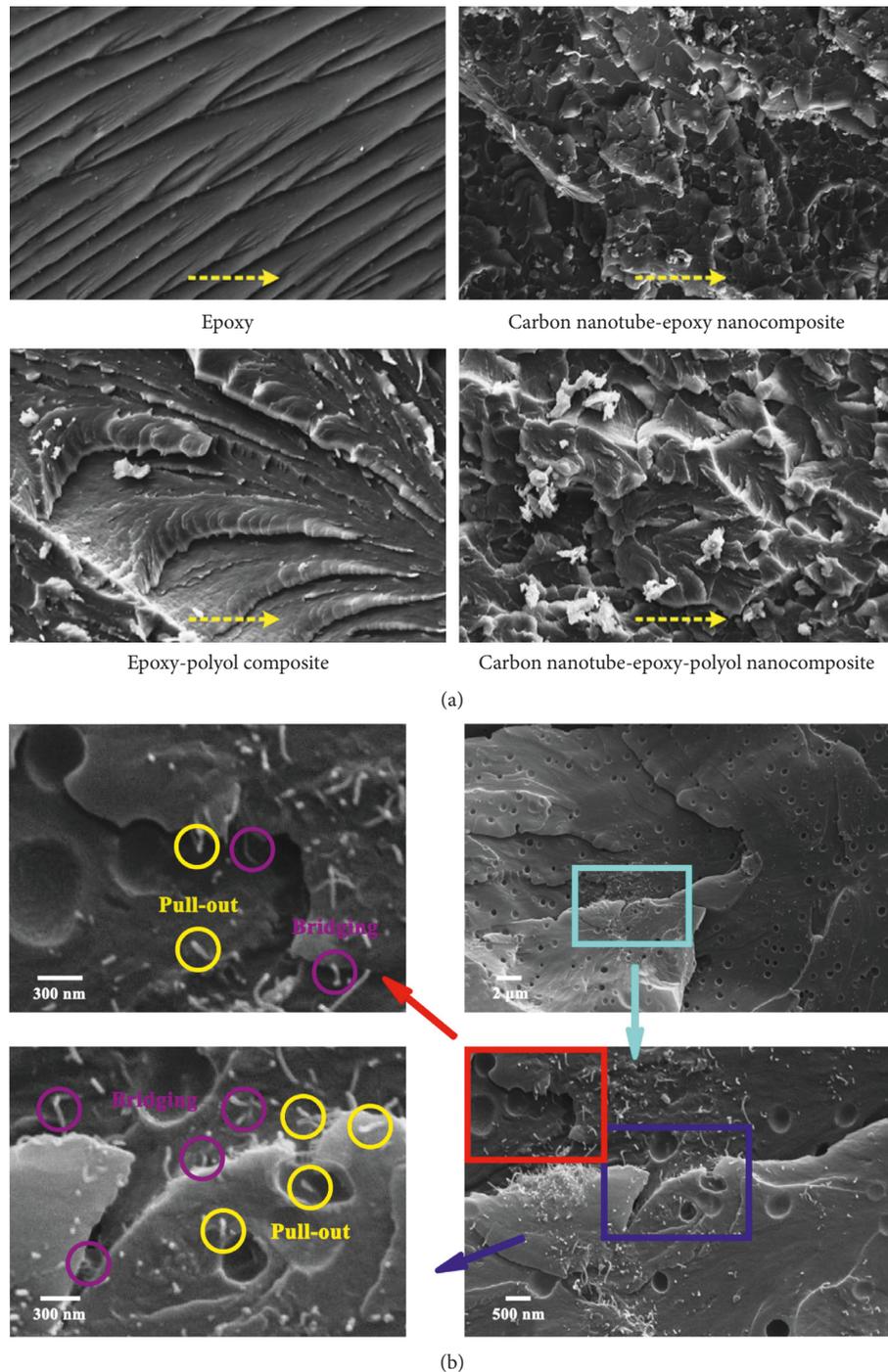


FIGURE 14: (a) Morphology of the quasistatically fractured surfaces characterized by a scanning electron microscope for neat epoxy and chemically modified epoxy composites. (b) Morphology of the quasistatic fracture surface characterized by a scanning electron microscope for an epoxy-polyol hybrid nanocomposite reinforced with multiwalled carbon nanotubes, illustrating a bridging phenomenon as well as the pull-out process of multiwalled carbon nanotubes from the polymer matrix (adapted with permission from reference [217]; copyright 2014, Elsevier Ltd).

reinforced polymer nanocomposites with improved thermal properties because this type of carbon nanotubes can provide unique structure advantages [240, 241].

Covalent functionalization of carbon nanotubes have proven to be very effective in improving the interfacial thermal conductance for carbon nanotube-reinforced polymer nanocomposites [242, 243], eventually leading to

their improved overall thermal performance [228]. For example, a significant improvement in interfacial thermal conductance was reported as large as one order of magnitude for the nanocomposites by means of this chemical method [229]. Molecular dynamics simulations can serve as a powerful tool to explore the effect of covalent functionalization of carbon nanotubes on the thermal properties of the

TABLE 2: Thermal resistance of the interface in carbon nanotube-reinforced polymer nanocomposites.

Nanocomposites	Interfacial thermal resistance	Method	Reference
Carbon nanotube-octane	$3.3 \times 10^{-8} \text{ m}^2 \cdot \text{K/W}$	Simulation	[226]
Carbon nanotube-octane	$4.0 \times 10^{-8} \text{ m}^2 \cdot \text{K/W}$	Simulation	[227]
Carbon nanotube-EVA*	$(0.2-9.6) \times 10^{-8} \text{ m}^2 \cdot \text{K/W}$	Simulation	[229]
Carbon nanotube-epoxy	$(0.24-2.6) \times 10^{-8} \text{ m}^2 \cdot \text{K/W}$	Experimental	[230]
Carbon nanotube-epoxy	$(0.77-2.5) \times 10^{-8} \text{ m}^2 \cdot \text{K/W}$	Simulation	[231]
Carbon nanotube-polyimide	$1.58 \times 10^{-8} \text{ m}^2 \cdot \text{K/W}$	Theoretical	[232]

\*EVA: poly(ethylene vinyl acetate).

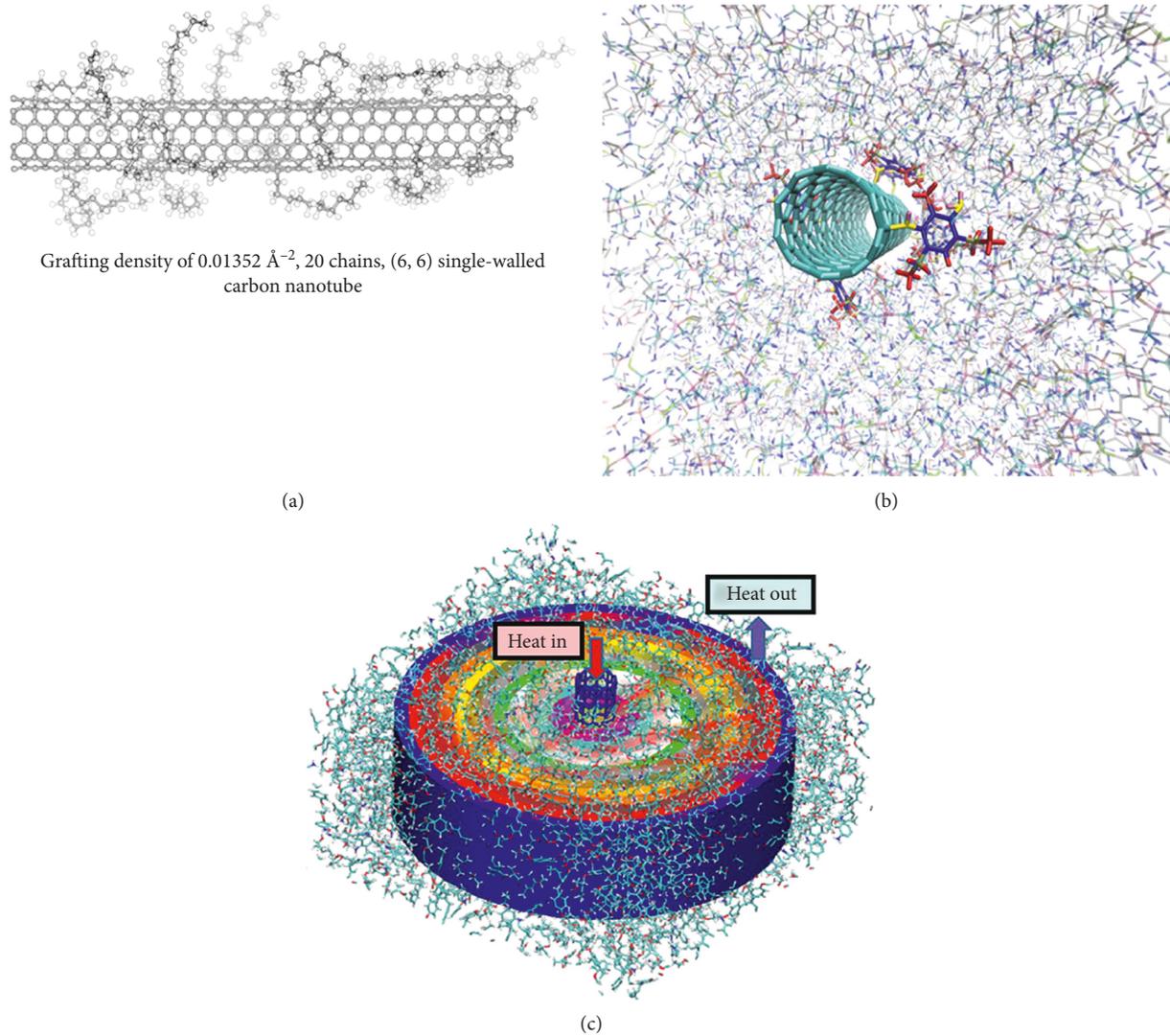


FIGURE 15: (a) An individual single-walled carbon nanotube with short end-grafted chains. (b) Schematic diagram of an individual single-walled carbon nanotube functionalized with crosslinker diethylenetoluenediamine and embedded in an epoxy matrix. Carbon nanotubes are rendered in cyan. The color scheme for the functionalized diethylenetoluenediamine molecule (red: hydrogen and carbon atoms contained in methyl groups, yellow: nitrogen atoms, and blue: carbon atoms contained in benzene rings). (c) Schematic diagram of a single-walled carbon nanotube-epoxy nanocomposite with different interfacial thicknesses. The nanocomposite exists in an epoxy environment, with the carbon nanotube as heat source and the interfacial region close to the polymer matrix as heat sink (adapted with permission from references [229, 231]; copyright 2006, Elsevier Ltd. Copyright 2012, TMS).

resulting nanocomposites [229, 231], as particularly well illustrated in Figure 15. Covalent functionalization can significantly improve the overall thermal performance for carbon nanotube-reinforced polymer nanocomposites due

to a great increase in the thermal conductance of the interface [229, 231]. However, excessive chemical functionalization will damage the intrinsic thermal properties of carbon nanotubes [244–247].

## 6. Concluding Remarks

Characterization is a fundamental process in the field of materials science, without which no scientific understanding of engineering materials could be ascertained. Characterization of the interface in carbon nanotube-reinforced polymer nanocomposites has been reviewed. Special emphasis is placed on how to determine the properties of the interface at the nanometer scale.

Tremendous research efforts are ongoing to characterize the properties of the interface in the polymer community. The state of research on the chemical functionalization of carbon nanotubes has been presented, followed by considerable details given over to the mechanical and thermal properties being pursued or envisioned in this emerging area. Various emerging characterization techniques used to determine the microscopic structure and properties of the interface are summarized.

New characterization techniques and methodologies are constantly emerging. In particular, the advent of atomic force microscopy in recent decades has revolutionized the field of carbon nanotube-reinforced polymer nanocomposites, allowing the imaging and analysis of the structures of the interface on much smaller scales than was previously possible, resulting in a significant increase in the level of understanding as to why different carbon nanotube-polymer interfaces show different bulk properties and behaviors. Atomic force microscopy can be combined with a variety of optical microscopy and spectroscopy techniques, thus further expanding its applicability.

Progress towards carbon nanotube-reinforced polymer nanocomposites will require continued development of characterization techniques, providing potential for tailoring the properties of the interface. The characteristic scale of the carbon nanotube reinforcement presents significant challenges to the development of methods for characterizing the properties of the interface. To address the issues associated with interfacial properties, it will be essential to understand the critical nature of the interface, develop accurate nanomechanics-based models, elucidate the interactions at the interface, and acquire knowledge of the relationship between nanocomposite properties and interfacial optimization, aiming at improving the performance of the nanocomposites effectively.

Carbon nanotubes pose both tremendous opportunities and significant new challenges for the development of fundamentally new polymer-based nanocomposites. Understanding the essence of the nanometer-scale reinforcement mechanism for the nanocomposites certainly warrants further experimental and theoretical research. Multidisciplinary research efforts will be required to bridge the gap in the existing knowledge between technological developments and fundamental understanding. There has never been a more exciting time to be involved in the field with new fundamental discoveries to be made regularly. It is hoped that this review would contribute to the development of this emerging area in some small way and would stimulate new research enthusiasm to enrich the content of an increasingly exciting discipline.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Acknowledgments

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