



Recent Advances in Carbon Nanotube-Polymer Composites

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Abstract: Carbon nanotubes demonstrate remarkable mechanical, thermal, and electrical properties, which allow a number of exciting potential applications. In this article, the most recent progress in research on the development of carbon nanotube-polymer composites is critically reviewed, with emphasis on recent advances in the principles and techniques for carbon nanotube functionalization. Various functionalization approaches and their role in the preparation of carbon nanotube-polymer composites with improved mechanical and electrical properties are discussed. The interaction between carbon nanotubes and polymers is also reviewed. Various techniques of carbon nanotube-polymer interaction measurements, including experimental and modelling studies, are described. Different methods of interaction improvement, mainly categorized under covalent and noncovalent interactions, are also described afterwards. An optimum carbon nanotube-polymer interaction is a crucial factor towards reaching the full potential of carbon nanotubes in nanocomposites. Potential topics of oncoming focus along with the potential applications of carbon nanotube-polymer composites are highlighted.

Keywords: Nanocomposites, Polymer Composites, Carbon Nanotubes, Physical Properties, Dispersion, Reinforcement, Functionalization, Interaction

1. Introduction

Carbon nanotubes were first observed by Iijima [1, 2], almost two decades ago, and since then, extensive work has been carried out to characterize their properties [3, 4]. Carbon nanotubes themselves have remarkable electrical, thermal, and mechanical properties. For example, carbon nanotubes theoretically have exceptional mechanical properties such as elastic modulus and strengths 10-100 times higher than the strongest steel at a fraction of the weight. Outstanding electrical, thermal, and mechanical properties of carbon nanotubes makes them a promising candidate for a wide variety of applications [1-4]. Carbon nanotubes have the potential to impact a wide range of fields and thus have emerged as one of the most promising materials in nanoscience research. Structural nanocomposites, in particular, aimed for taking advantage of the superior mechanical properties of carbon nanotubes as well as their high aspect ratios and surface areas [5-8]. In recent years, carbon nanotubes are prime candidates for use in the development of polymer composite materials [5-12]. Considerable efforts

have been devoted to fabricate advanced carbon nanotube composite materials that exhibit one or more of the above-mentioned excellent properties [13-22]. The combination of remarkable properties makes carbon nanotubes potentially ideal candidates for the formation of polymer composites with improved mechanical properties and electrical conductivity [23-28]. Carbon nanotubes can be thought of as the ultimate carbon fiber with break strengths reported as high as 200 GPa, and elastic moduli in the 1000 Gpa range [29-32]. This, coupled with approximately 500 times more surface area per gram and aspect ratios of around 1000, has spurred a great deal of interest in using carbon nanotubes as a reinforcing phase for polymer matrices.

Carbon nanotube-polymer nanocomposites have the advantage of size compatibility between their constituents [9-12]. Introducing carbon nanotubes to polymer matrices modifies electrical, thermal, mechanical, and morphological properties of the produced nanocomposite [13-22]. In order to obtain the best performance from carbon nanotube-polymer nanocomposites, there are a lot of technical challenges to overcome. For example, purification of carbon nanotubes is a

necessary step to remove metallic and amorphous carbon impurities, while avoiding serious damage to the carbon nanotubes [13-15]. Furthermore, carbon nanotube content and alignment as well as the void content in the nanocomposite should be controlled [16-22]. The dispersion of carbon nanotubes in the polymer matrix plays an important role in the performance of nanocomposites. In general, it is a challenge to obtain the optimum dispersion, which requires further researches and exploration at different scales, particularly for nanocomposites with relatively high carbon nanotube content [35, 36].

Lacking direct manipulation, when used as reinforcement in polymers, carbon nanotubes are typically first randomly dispersed in a solvent or polymer fluid, melt by sonication or shear mixing followed by further processing to create the nanocomposite. The energy input to disperse the carbon nanotubes tends to break them into shorter segments [35, 36], decreasing their aspect ratio in the final composite while simultaneously increasing their dispersibility. However, insufficient dispersion is often cited as a process limitation [37] and the key diminishing factor [33, 38] on the mechanical properties of nanocomposite. Efforts to improve carbon nanotube dispersion include the use of surfactants [39, 40], as well as the oxidation or chemical functionalization of the surface of carbon nanotubes [11, 18, 35, 36].

Adding carbon-based fillers to polymers to improve mechanical properties, reduce weight, and act as heat conductors is not a novel idea. Carbon black has been widely utilized to reinforce rubber and plastics, for example, it is extensively used in racing tires to reduce thermal damage. Carbon fibers are also hugely popular industrial materials that are used in cars, airplanes, bicycles, etc. However, the carbon-based polymer additives with the greatest potential are carbon nanotubes. The ability to harness the mechanical strengths of carbon nanotubes has already given rise to new industrial products [3-8]. For example, sports equipment, such as tennis racquets and golf clubs containing carbon nanotubes, has been produced and marketed. With carbon nanotubes becoming easier to produce and cheaper to buy, the carbon nanotube industry could potentially overtake the carbon fiber

industry and becomes one of the major additives for polymer-composite fabrication [22, 32].

There are a lot of studies and a limited number of literature reviews on the conductive and mechanical properties of carbon nanotube-polymer composites [6-8, 19, 21, 32, 36]. However, a significant progress has been made in the area of the preparation and utilization of carbon nanotube-polymer composite materials lately. In this article, the most recent progress in research on the development of carbon nanotube-polymer composites is critically reviewed, with emphasis on recent advances in the principles and techniques for carbon nanotube functionalization.

2. Carbon Nanotube Materials and Properties

Since the discovery of single-walled carbon nanotubes [2, 41], there has been a flurry of research activity aimed at understanding their physical properties [42], elucidating their growth mechanisms [43], and developing novel applications for them [44]. Carbon nanotubes and related nanomaterials along with their physical properties have been discussed extensively in the literature [1-8]. Consequently, a comprehensive review of this topic will not be presented here. A brief discussion of these nanomaterials and their physical properties is appropriate, however, to familiarize the reader with the typical characteristics and the most common methods of production.

There are three common types of carbon nanotube raw materials that can be employed as composite materials. These are single-walled carbon nanotubes [2, 16, 23, 24, 26, 35, 41], multi-walled carbon nanotubes [1, 11, 26, 27, 31, 32], and carbon nanofibers [18, 22, 37]. The three raw materials can also be used in combination to develop composite materials. The morphologies of these nanomaterials are illustrated in Figure 1. These carbon nanotubes and carbon nanofibers are commercially available. A summary of the characteristics of each of these nanomaterials has been given in the literature [22]. Carbon nanotube properties have also discussed in the literature [45, 46].

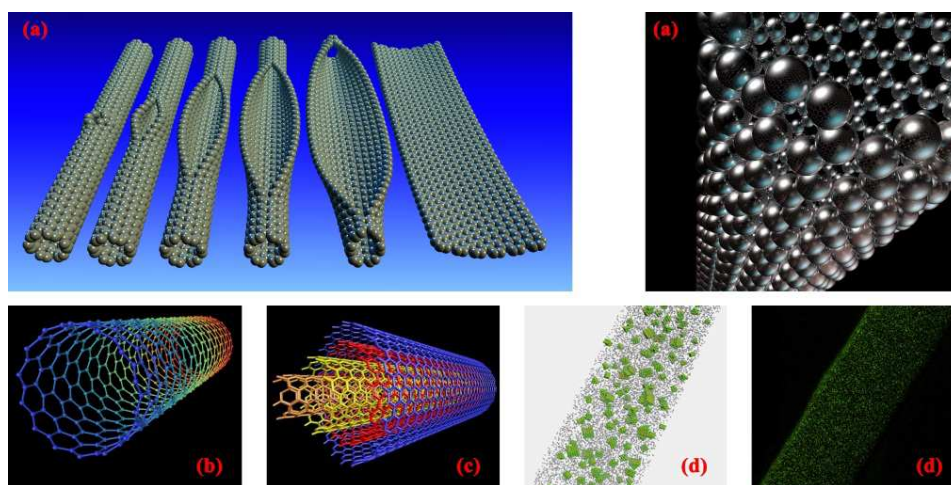


Figure 1. Schematic illustration of carbon nanotubes: (a) carbon nano-walls, (b) single-walled carbon nanotubes, (c) multi-wall carbon nanotubes, and (d) carbon nanofibers.

Carbon nanotubes are the strongest and stiffest materials yet discovered [29-32]. This strength results from the covalent sp^2 bonds formed between the individual carbon atoms. Although the strength of individual carbon nanotube shells is extremely high, weak shear interactions between adjacent shells and tubes result in a significant reduction in effective strength [47]. The electronic property of carbon nanotubes is a strong function of its chemical doping, mechanical deformation, and atomic structure. Changing these properties can induce strong changes in electrical conductance [48]. The electrical impedance has been found to be very sensitive to chemical exposure [49, 50] and mechanical deformation [51]. These properties are highly dependent on the type of carbon nanotubes [52, 53]. Carbon nanotubes have useful absorption, photoluminescence, and Raman spectroscopy properties [54]. Spectroscopic methods offer the possibility of quick and non-destructive characterization of relatively large amounts of carbon nanotubes. There is a strong demand for such characterization from the industrial point of view: numerous parameters of the carbon nanotube synthesis can be changed, unintentionally or intentionally, to alter the quality of carbon nanotubes [54].

Recently, several kinds of techniques have been developed to produce carbon nanotubes in sizable quantities, such as arc discharge, laser ablation, and chemical vapor deposition [3, 4]. Most of these processes take place in a vacuum or with process gases. The growth method of chemical vapor deposition is popular [48, 55]. Controlled synthesis of carbon nanotubes opens up exciting opportunities in nanoscience and nanotechnology, including electrical, mechanical, and electromechanical properties and devices, chemical functionalization, surface chemistry and photochemistry, and molecular sensors [48].

3. Carbon Nanotube Functionalization

The nanocomposite performance strongly depends on the dispersion of carbon nanotubes in the polymer matrix and the interfacial interactions between them [5-8]. However, the

reinforcing carbon nanotubes are inert and can interact with the surrounding polymer matrix mainly through van der Waals interactions, unable to provide an efficient load transfer across the interface [8-20]. Consequently, considerable efforts have been devoted to develop methods in order to modify surface properties of carbon nanotubes [4, 53, 56, 57]. These methods can be conveniently divided into chemical functionalization and physical methods based on the interactions between the active molecules and carbon atoms on the carbon nanotubes [15]. Major principles of these methods along with their advantages and disadvantages have been summarized in the literature [36].

It is known that carbon nanotube solubility, dispersion, and stress transfer must all be maximized to obtain optimum performance. Unless the interface between carbon nanotube and polymer is carefully engineered, poor load transfer between carbon nanotubes, when in bundles, and between carbon nanotubes and surrounding polymer chains may result in interfacial slippage. Consequently, functionalization of carbon nanotubes is extremely important for their processing and potential applications in polymer composites [19]. In general, composites based on chemically modified carbon nanotubes show the best mechanical results because functionalization enables a significant improvement in both dispersion and stress-strain transfer. The treatment of carbon nanotubes by chemical functionalization is widely used to improve the dispersion of carbon nanotubes in solvents [19].

The functionalization of carbon nanotubes not only results in the improved dispersibility in various organic solvents and polymers [58], but also increases the interface strength [58]. However, chemical functionalization may disrupt the bonding of the graphene sheet, and thereby reduces the mechanical properties [5]. Several approaches have been developed, in both molecular and supramolecular chemistry [36]. These approaches include defect functionalization, covalent functionalization of the sidewalls, noncovalent exohedral functionalization, for example, formation of supramolecular adducts with surfactants or polymers, and endohedral functionalization, as illustrated in Figure 2.

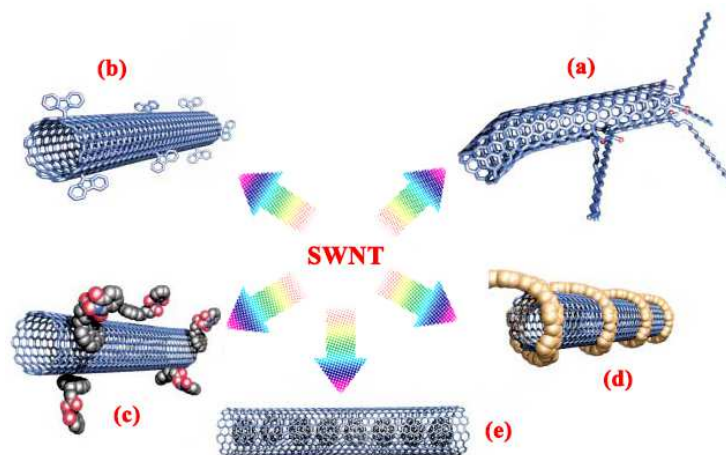


Figure 2. Functionalization possibilities for single-walled carbon nanotubes: (a) defect-group functionalization, (b) covalent sidewall functionalization, (c) noncovalent exohedral functionalization with surfactants, (d) noncovalent exohedral functionalization with polymers, and (e) endohedral functionalization with, for example, fullerenes. For methods (b)-(e), the carbon tubes are drawn in idealized fashion, but defects are found in real situations.

3.1. Chemical Functionalization

Chemical functionalization is based on the covalent linkage of functional entities onto carbon scaffold of carbon nanotubes [35, 36]. It can be performed at the termini of the carbon tubes or at their sidewalls. Direct covalent sidewall functionalization is associated with a change of hybridization from sp^2 to sp^3 and a simultaneous loss of π -conjugation system on the graphene layer. This process can be made by reaction with some molecules of a high chemical reactivity, such as fluorine [59-63]. Besides sidewall fluorination of carbon nanotubes, other similar methods, including cycloaddition, such as carbene and nitrene addition [64-66], Diels-Alder reaction, bromination [67], hydrogenation [68], chlorination, and aminobenzene acids [69], have also been successfully employed in recent years. All these methods can be regarded as the derivative of sidewall functionalization [36].

Defect functionalization is another method for the covalent functionalization [35, 36, 56, 57]. This process takes advantage of chemical transformation of defect sites on carbon nanotubes. Defect sites can be the open ends and holes in the sidewalls, pentagon or heptagon irregularities in the hexagon graphene framework. Oxygenated sites can also to be considered as defects [36]. Defects can be created on the sidewalls as well as at the open ends of carbon nanotubes by an oxidative process with strong acids such as nitric acid, sulfuric acid, a mixture of them [70, 71], or with strong oxidants [72-75]. The defects on carbon nanotubes created by oxidants are stabilized by bonding with carboxylic acid or hydroxyl groups. These functional groups have rich chemistry and the carbon nanotubes can be used as precursors for further chemical reactions, such as silanation [76], polymer grafting

[77], esterification [78], thiolation [79], and alkylation and arylation [80, 81]. The carbon nanotubes functionalized in this way are soluble in many organic solvents because the hydrophobic nature of carbon nanotubes is changed to hydrophilic one due to the attachment of polar groups. The chemically functionalized carbon nanotubes can produce strong interfacial bonds with many polymers, allowing carbon nanotube-based nanocomposites to possess high mechanical and functional properties [36].

3.2. Physical Functionalization

Functionalization of carbon nanotubes using a covalent method can provide useful functional groups onto the surface of carbon nanotubes [36]. Unfortunately, these methods have two major drawbacks: firstly, during the functionalization reaction, a large number of defects are inevitably created on the sidewalls of carbon nanotubes, and in extreme cases, carbon nanotubes are fragmented into smaller pieces [36]. These damaging effects can result in severe degradation in mechanical properties of carbon nanotubes as well as disruption of π electron system in carbon nanotubes. The disruption of π electrons is detrimental to transport properties of carbon nanotubes because defect sites scatter electrons and phonons that are responsible for the thermal and electrical conductions of carbon nanotubes, respectively. Secondly, strong oxidants or concentrated acids are usually used for the functionalization of carbon nanotubes, which are typically environmentally unfriendly [36]. Consequently, considerable efforts have been devoted to develop methods that are of low cost, less damage to the structure of carbon nanotubes, and convenient to use.

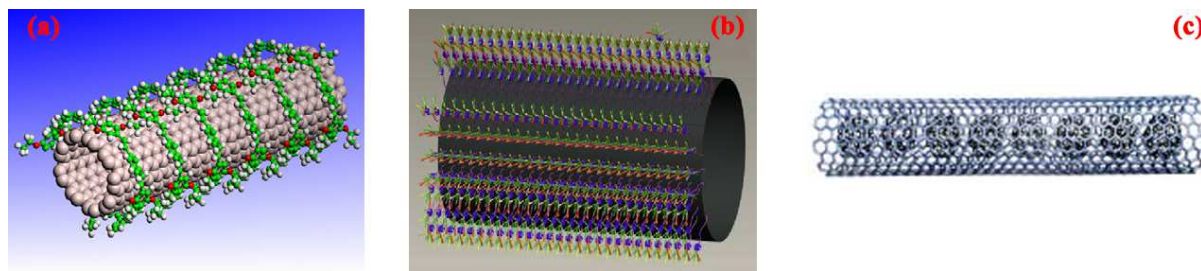


Figure 3. Schematic illustration of carbon nanotube functionalization using non-covalent methods: (a) polymer wrapping, (b) surfactant adsorption, and (c) endohedral method.

Non-covalent functionalization is an alternative method for tuning the interfacial properties of carbon nanotubes. The functionalization of carbon nanotubes using non-covalent methods is illustrated in Figure 3. The suspension of carbon nanotubes in the presence of polymers, such as polyphenylene vinylene [82] or polystyrene [83], results in the wrapping of polymer around the carbon nanotubes to form supermolecular complexes of carbon nanotubes, as shown in Figure 3(a). This is a typical example of non-covalent functionalization of carbon nanotubes. The polymer wrapping process is achieved through the van der Waals interactions and π - π stacking between carbon nanotubes and polymer chains containing aromatic rings.

Surfactants have also been employed to functionalize

carbon nanotubes, as shown in Figure 3(b). Several studies have been contributed to the in-depth study of the effects of surfactant on dispersibility and other physical properties of carbon nanotubes. The surfactants studied previously include non-ionic surfactants [84, 85], anionic surfactants [86, 87], and cationic surfactants [88, 89]. A simulation showing single-walled carbon nanotubes embedded within sodium dodecyl sulfate micelles is illustrated in Figure 4. In particular, a recent work provides a comprehensive review of the mechanisms behind the improved dispersibility of carbon nanotubes [90]. The physical adsorption of surfactant on the surface of carbon nanotubes lowered the surface tension of carbon nanotubes, effectively preventing the formation of

aggregates. Furthermore, the surfactant-treated carbon nanotubes overcame the van der Waals attraction by electrostatic-steric repulsive forces. The efficiency of this method was highly dependent on the properties of medium chemistry, surfactants, and polymer matrix. In water-soluble polymers such as polyethylene glycol, cationic surfactants had some advantages, whereas in water-insoluble polymers, the

dispersion of carbon nanotubes was promoted by a non-ionic surfactant [39, 90, 91]. The treatment of non-ionic surfactants was based on a strong hydrophobic attraction between the solid surface and the tail group of surfactant. Once the surfactant was adsorbed onto the surface of the filler, the surfactant molecules were self-assembled into micelles above a critical micelle concentration [36].

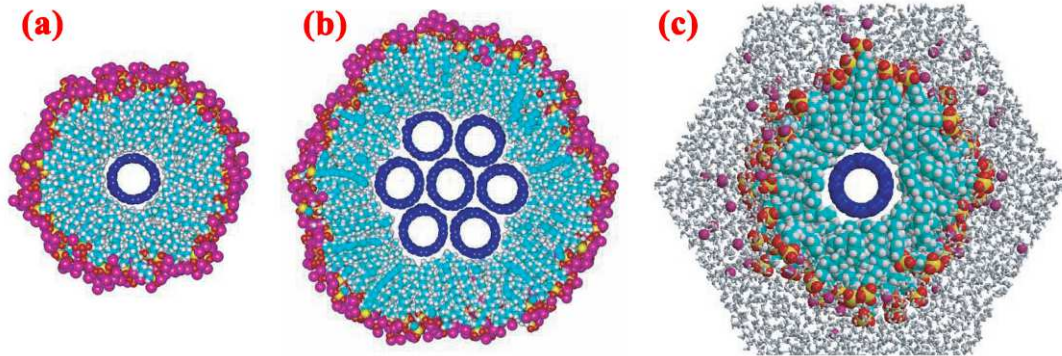


Figure 4. A simulation showing single-walled carbon nanotubes embedded within sodium dodecyl sulfate micelles: (a) an individual single-walled carbon nanotube embedded in a cylindrical sodium dodecyl sulfate micelle, (b) a seven-tube bundle of single-walled carbon nanotubes coated by a layer of sodium dodecyl sulfate, and (c) a larger region showing a water-free hydrocarbon environment as well. Adapted with permission from Ref. [86].

Another non-covalent method is the endohedral method, as shown in Figure 3(c). In this method, guest molecules or atoms are stored in the inner cavity of carbon nanotubes through the capillary effect. The insertion often takes place at defect sites localized at the ends or on the sidewalls of carbon nanotubes. The insertion of inorganic nanoparticles into the carbon nanotubes [92] is a typical example of endohedral functionalization. Small biomolecules can also be entrapped in the inner hollow channel of carbon nanotubes by simple adsorption, forming natural nano-test tubes [56, 93]. The combination of these two materials is particularly useful to integrate the properties of the two components in hybrid materials for use in energy storage, catalysis, nanotechnology, and molecular scale devices [92].

4. Carbon Nanotube-Polymer Interfaces

Particular attention is focused on the carbon nanotube-polymer interface [94-100]. From micro-mechanics, it is through shear stress build-up at this interface that stress is transferred from the polymer matrix to the carbon nanotubes [5, 8-20]. Lower-than-predicted carbon nanotube-polymer composite properties can be attributed to a lack of interfacial bonding [101, 102]. If the surface of a carbon nanotube, essentially an exposed graphene sheet, is considered, it is not surprising that interfacial traction is a concern [5]. It is the weak inter-planar interaction of graphite that provides its solid lubricant quality, and resistance to polymer matrix adhesion. This is exaggerated by the chemically inert nature of graphene structures [5]. A June 2002 publication on carbon nanotube composites noted “*In depth study on the stress transfer mechanism of the carbon nanotube composites with different chemical and geometrical properties, matrix environments and loading conditions are essential*” [103]. Since then, some

interesting published results have described progress on addressing this issue. The force required to separate a carbon nanotube from a solid polymer matrix has been measured by performing reproducible nanopullout experiments using atomic force microscopy [104]. The computed average interfacial shear strength using a Kelly-Tyson approach of 47 MPa, typical values for carbon fiber-epoxy are in the range of 30 to 80 MPa [105], was sufficiently high to suggest that covalent bonding between defects in the outer shell of the carbon nanotube and the polymer was occurring. It also suggested that the polymer chains close to the interface behaved differently than the bulk, a logical result when considering the outer diameter of the carbon nanotubes was of similar magnitude to the radius of gyration for the polymer. In a report corroborating an interfacial region of non-bulk polymer, a “sheathing layer” of polycarbonate on pulled out multi-walled carbon nanotubes was imaged, which gave further direct evidence of substantial carbon nanotube-polymer interaction [106]. Furthermore, chemical functionalization of the carbon nanotubes augmented the diameter of the polymer sheath suggesting chemical augmentation of interfacial bonding. Acid oxidation of multi-walled carbon nanotubes was again reported to attach carboxylic groups on the surface, which were then reacted with epoxide-terminated molecules up to 12 weight percent by a method of thermal gravimetric analysis [58]. These types of functionalized carbon tubes could enhance reinforcement of epoxy resins. Carboxylated tubes have also been reported to augment the cure rate of epoxy resins at lower operating temperatures [107]. Unfortunately, direct and indirect measurement of carbon nanotube-polymer interfacial shear strength suggesting good bonding exists conflict with other reports which often cite clean pull-out of carbon nanotubes and poor interfacial bonding [101, 108]. The magnitude of

carbon nanotube strength, more than 10 times that of typical carbon fiber, may preclude embedded carbon nanotube tensile failure in large numbers resulting in the dominant failure mode to be carbon nanotube pull-out. Order of magnitude increases in interfacial shear strength may be required for the most efficient strengthening of polymers with carbon nanotubes. Optimizing the carbon nanotube-polymer interface for nanoscale mechanical reinforcement remains unclear, but the evidence available indicates that chemical means can be effective, and this is likely to be a major focus in the near term.

Theoretical treatments of carbon nanotube pull-out have also been reported in the literature [108, 109]. A single-walled carbon nanotube pulled out from a crystalline polyethylene matrix, which interacted with the carbon nanotube through van der Waals forces represented by the Lennard-Jones potential, was modeled through molecular dynamics simulations and described by an interfacial friction model. The velocities and displacements of carbon nanotubes were traced in order to characterize the interfacial interactions during carbon nanotube pull-out. The interfacial friction between carbon nanotube and polymer was described with a relation between the applied force and carbon nanotube velocity. Furthermore, an interfacial friction model for the entire pull-out process was proposed, which involved an effective viscosity coefficient for the interfacial sliding. The existence of a carbon-ring-based period in carbon nanotube sliding during pull-out was identified. Linear trends in the carbon nanotube velocity-force relation were observed and used to estimate an effective viscosity coefficient for interfacial sliding at the carbon nanotube-polymer interface. A low effective viscosity of 0.2 cP was found for interfacial sliding [108]. A force of approximately 0.1 nN was required for pull-out to initiate for a carbon nanotube.

5. Strongly Coupled Carbon Nanotube-Polymer Systems

Strong coupling has been reported in conducting carbon nanotube-polymer systems [8-20]. The conduction mechanism of polymers is based on the motion of charged defects within the conjugated framework, and strongly depends on the level of doping [110-112]. Conducting polymers have been successfully utilized for commercial applications. Recently, considerable efforts have been devoted to optimization of the optical and electrical properties of conducting polymers, mainly via the development of efficient pathways for doping [110]. Recent experiments have demonstrated that multi-walled carbon nanotubes could be used for doping of a functional conjugated polymer [113], polyaniline [114], and a conjugated luminescent polymer. The electronic structure of functional conjugated polymer [115] as well as other types of conducting polymers was modified by the presence of carbon nanotubes [116, 117], indicating strong carbon nanotube-polymer coupling. In a different system [117], single-walled carbon nanotubes promoted the solution-protonation of the polymer, and thus affected its electrical properties. Conjugated polymers and carbon nanotubes were strongly associating, tightly bound systems [100]. The molecular geometry of the association was that of helical wrapping of the carbon nanotubes by the polymers [117]. The polymer-wrapped carbon nanotubes formed long-lived stable dispersions in different liquid media [118], and might be utilized for the preparation of carbon nanotube-polymer composites [119] exhibiting improved mechanical and electrical properties.

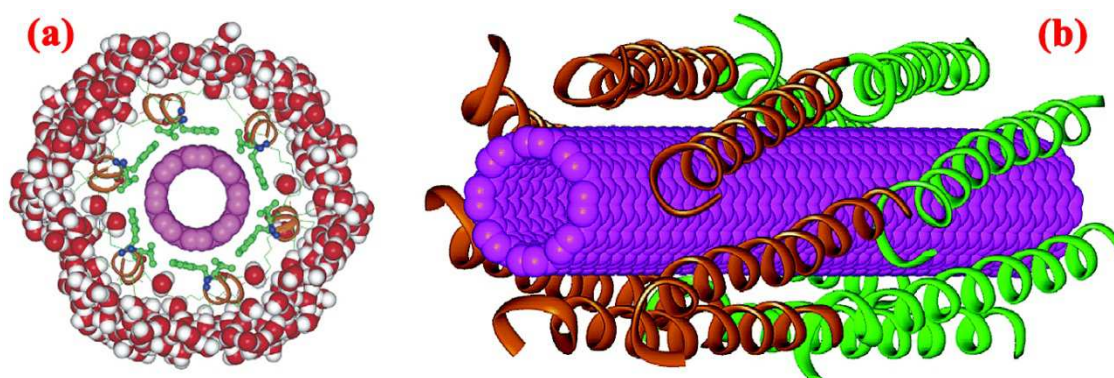


Figure 5. A model illustrating potential interactions between a synthetic, hydrophilic polypeptide and an individual single-walled carbon nanotube: (a) cross-section view of a carbon nanotube wrapped by six peptide helices and a water shell, and (b) view of a peptide-wrapped carbon nanotube illustrating the 12 peptide helices used in the model. Adapted with permission from Ref. [129]. For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.

Strong binding and carbon nanotube-polymer-wrapping were also reported for biopolymers. Interactions between specific types of deoxyribonucleic acid and carbon nanotube enabled the visualization of deoxyribonucleic acid [120] and affected the properties of carbon nanotubes [121]. Carbon nanotube-deoxyribonucleic acid complexes were found to form stable dispersions [122], enable fractionation of carbon

nanotubes [120, 123], preparation of fibers and composites [124-126], and nano-electronic devices [127]. A comprehensive review of carbon nanotube-deoxyribonucleic acid interactions and related bio-applications has been given in the literature [128]. Recently, Dieckmann *et al.* [129] reported the synthesis and application of an amphiphilic peptide, specifically designed to disperse single-walled carbon nanotubes. It has been found

that single-walled carbon nanotubes induce preferential folding of the peptide into specific configurations, and the interactions among the single-walled carbon nanotube-peptide moieties could be utilized for controlled self-assembly of the complexes. The concept is demonstrated in Figure 5. Electron microscopy and polarized Raman studies revealed that the peptide-coated carbon nanotubes assemble into fibers with the carbon nanotubes aligned along the fiber axis. Most importantly, the size and morphology of the fibers can be controlled by manipulating solution conditions that affect peptide-peptide interactions.

Dispersion of carbon nanotubes through polymer wrapping was suggested in additional systems, where it was conjectured that wrapping results in screening of the hydrophobic interaction at the carbon nanotube-water interface [130, 131]. This approach, wrapping water-soluble linear polymers around the carbon nanotubes, was robust and general, allowing pristine carbon nanotubes to be manipulated reliably by solution-phase techniques such as electrophoresis and chromatography, and simplifying their use as chemical reagents. The carbon nanotubes could be unwrapped by changing the solvent system. This solubilization process opens the door to solution chemistry on pristine carbon nanotubes [130].

6. Carbon Nanotube-Polymer Interactions in Nanocomposites

The carbon nanotube-polymer interfacial characteristics directly affect the efficiency of carbon nanotube reinforcements in improving mechanical, thermal, and electrical properties of the carbon nanotube-polymer nanocomposite [100]. An optimum carbon nanotube-polymer interaction is a critical factor towards reaching the full potential of carbon nanotubes in nanocomposites [100]. While a wide range of characteristic parameters has been reported, there are contradictory reports that show the effect of carbon nanotube on a particular property to be indifferent, improving, or even deteriorating [132]. These contradictory findings are perhaps obtained due primarily to differences between material characteristics and processing conditions of the nanocomposites. Furthermore, there are a number of other parameters such as the curvature of carbon nanotubes [133, 134], which has been found to play a significant role in achieving the optimum performance of nanocomposites. However, the experimental results or even realistic modelling predictions have always been below the rule of mixtures predictions [135]. The root cause of these under-performances may be sought in interaction behavior of the nanocomposite constituents. The interaction characteristics significantly affect the performance of nanocomposites. Furthermore, the answer to challenges, such as the dispersion of carbon nanotubes in nanocomposites, lies with the interaction characteristics of the system. Consequently, the interaction properties of the system should be tailored in order to optimize the performance of nanocomposites.

The carbon nanotube-polymer interaction characteristics

have been studied at three different levels: between different shells of a multi-walled carbon nanotube [136], between different carbon nanotubes in a bundle [137], and between carbon nanotubes and the polymer matrix [138]. The first two types of carbon nanotube-polymer interaction are only related to the characteristics of carbon nanotubes. However, the carbon nanotube-polymer interaction is dependent on the characteristics of both constituents and how they interrelate with each other. The performance of conventional composites has also been found to be affected by fiber-matrix interaction [139]. However, there are fundamental differences in interaction characteristics between them, and a general overview of the interaction studies is missing in the literature. Consequently, a brief discussion of the interaction studies on carbon nanotube-polymer nanocomposites is given as follows.

6.1. Noncovalent Interaction

The extensive efforts to improve carbon nanotube-polymer interaction may be classified under noncovalent and covalent interaction categories [100]. The noncovalent interaction in polymer nanocomposites is enhanced by using any mechanical interaction improving techniques, including bridging, increasing specific interfacial area, and wrapping [100]. Bridging happens when a polymer chain interacts with two or more reinforcements at the same time. The possibility of bridging phenomenon is determined by the ratio of polymer radius of gyration to the average distance between nearest reinforcements [140]. Consequently, increasing the content of carbon nanotubes or using polymers with higher molecular weight greatly increases the chance of bridging. A specific interaction area is the polymer-reinforcement interfacial area per unit volume, only depending on the polymer to reinforcement density ratio as well as weight fraction and diameter of the reinforcement [140]. Consequently, increasing the content of properly dispersed carbon nanotubes enhances the specific interfacial area and results in a stronger nanocomposite [141]. The wrapping mechanism [142] not only results in a better carbon nanotube-polymer interaction but also helps to improve the dispersion of carbon nanotubes [143]. It is affected by the structure of the polymer, chemical composition of the polymer molecules, and geometric parameters of the constituents in the nanocomposite. Polymers with stiff backbones tend to wrap around carbon nanotube reinforcements with more distinct conformation [144], and form a helical configuration [145]. However, flexible polymer chains with bulky and aromatic side groups form an interchain coiling rather than a helical conformation [146]. Figure 6 shows examples of flexible and stiff backbone polymers interacting with single-walled carbon nanotubes. Chemical composition of the polymer also affects the wrapping behavior of the chains. On the other hand, bulky aliphatic side chains hinder such interactions. Finally, the geometric parameters of the polymer and reinforcement determine the possibility of wrapping. When the radius of gyration of the chains is greater than the radius of carbon nanotubes, there is a significant improvement in carbon nanotube-polymer interaction properties [140]. Consequently, nanocomposites with high

molecular weight polymers and small diameter carbon nanotubes are more viable to present wrapping mechanism.

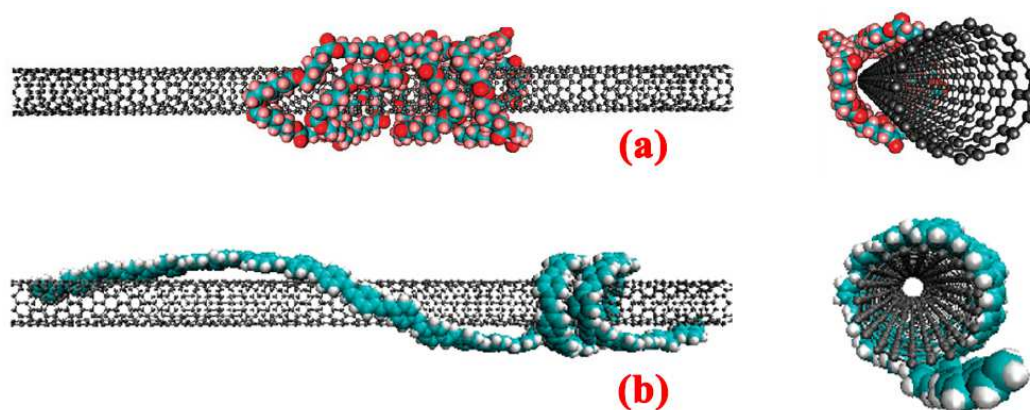


Figure 6. Molecular dynamics snapshots of polymer chains introduced to a single-walled carbon nanotube. Panel (a) shows poly(caprolactone) with flexible backbone interacting with the carbon nanotube at 3200 ps [146]. Panel (b) shows poly(paraphenylene vinylene) with a stiff backbone interacting with the carbon nanotube at 2400 ps [144]. Adapted with permission from Refs. [144, 146]. For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.

Wang *et al.* [147] reported a simple and scalable method to enrich large quantities of semiconducting arc-discharged single-walled carbon nanotubes using dithiafulvalene-thiophene copolymers. Stable solutions of highly individualized and highly enriched semiconducting single-walled carbon nanotubes were obtained. Molecular dynamics simulations of polymer backbone interactions with and without side chains indicated that the presence of long alkyl side chains gave rise to the selectivity toward semiconducting tubes, indicating the importance of the roles

of the side chains to both solubilize and confer selectivity to the polymers. Representative snapshots of the molecular dynamics simulations are shown in Figure 7. Increasing the ratio of thiophene to dithiafulvalene units in the polymer backbone could slightly improve the selectivity toward semiconducting single-walled carbon nanotubes. High on to off ratio solution-processed thin film transistors were fabricated from the sorted single-walled carbon nanotubes to confirm the selective dispersion of semiconducting arc-discharge single-walled carbon nanotubes.



Figure 7. Representative snapshots of the molecular dynamics simulations for (a) a metallic single-walled carbon nanotube and (b) a semiconducting single-walled carbon nanotube with pDTFF-1T, pDTFF-2T, and pDTFF-3T. Adapted with permission from Ref. [147]. For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.

Noncovalent interaction methods require no extra preparation steps and induce no defect in the structure of the carbon nanotubes [100]. The interaction may significantly improve by in situ crystallization of the polymer with carbon nanotubes [148, 149]. This type of interaction only provides a weak interfacial adhesion, depending on the polymer type. Furthermore, adjusting parameters such as carbon nanotube content and polymer molecular weight may affect the processability of the nanocomposite. One solution to address such problems is noncovalent grafting of carbon nanotubes with end-functionalized polymers [150]. In this technique, zwitterionic interactions or hydrogen bonds are formed between the defect sites of purified carbon nanotubes and the functional groups of the polymers [100]. Consequently, the

undesirable defects caused by carbon nanotube purification play a positive role in this method, while the interactions are still noncovalent and the carbon nanotubes are not chemically functionalized by functional groups.

6.2. Covalent Interaction

The covalent interaction occurs when polymer chains are chemically bonded to the carbon nanotube reinforcements [100]. The covalent interaction has been found to be an effective method to improve nanocomposite properties [151, 152]. It is typically achieved through open-end oxidation or sidewall functionalization [35]. Most of functionalization techniques require carbon nanotubes to be pre-modified by introducing functional groups to their surface. These functional groups

should be compatible with the polymer matrix [58, 153, 154] in order to react with reactive groups of the polymer matrix and form chemical bonds. An effective method of forming such chemical bonds is in situ polymerization where free radicals of the polymer matrix monomer interact with each other and with the carbon nanotubes functional groups simultaneously [155, 156]. These standard functionalization techniques are suitable for reactive polymers. Nonreactive polymers are functionalized through other techniques such as ozone-mediated process [157]. As a result, alkylperoxide and hydroperoxide groups are formed in the polymer chains and decompose into reactive radicals under heat. These radicals target the sp^2 hybrid carbons and result in a convenient approach to form chemical bonds between pristine carbon nanotubes and nonreactive polymers. Other techniques such as microwave irradiation [154, 158] and plasma enhanced chemical vapor deposition [159] are also reported to be effective.

Nanocomposites with covalent interaction exhibit a better dispersion of the carbon nanotube reinforcements within the polymer matrix [100]. The agglomerated bundles of carbon nanotubes are broken and thus a more uniform behavior can be found [151]. Furthermore, stronger adhesions between the carbon nanotubes and the polymer matrix result in better

mechanical performance. The effect of functionalization on different characteristics of the nanocomposites has also been evaluated [160]. During the functionalization process, reactive radicals form chemical bonds with the sp^2 carbons and convert them into sp^3 carbons. Consequently, Raman spectroscopy can distinguish functionalization through increase in the D to G band intensity ratios and in the intensity of D band which shows the disorders in the carbon nanotubes. Furthermore, the results of thermogravimetric analysis indicated negligible amount of mass loss for pristine multi-walled carbon nanotubes below 1120 K, whereas the mass loss of functionalized carbon nanotubes prior to this temperature is due primarily to the organic polymer chains incorporated to the carbon nanotubes [157]. Scanning electron micrographs of the fractured surface of functionalized nanocomposites showed broken carbon nanotubes and demonstrated a better adhesion between the carbon nanotubes and the polymer matrix [156]. Furthermore, transmission electron micrographs also revealed amorphous polymer layers covering functionalized carbon nanotubes [154, 157], as shown in Figure 8. Finally, the covalent interaction has a stronger influence of carbon nanotube properties on nanocomposite characteristics [161].

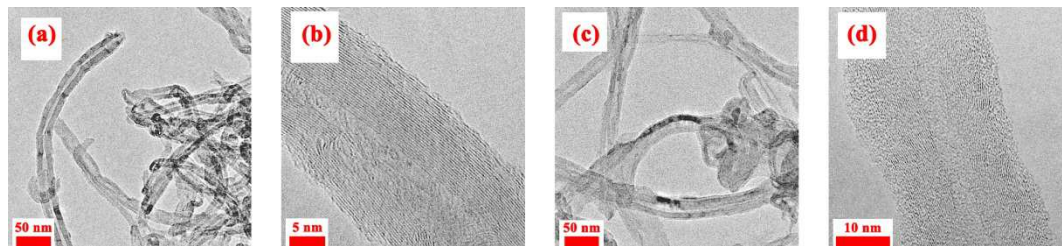


Figure 8. Transmission electron micrographs of (a, b) purified and (c, d) modified multi-walled carbon nanotubes. The polystyrene layer is applied via polymerization of styrene under microwave irradiation to form a middle layer between carbon nanotubes and matrix, resulting in a strong interfacial adhesion. Adapted with permission from Ref. [154].

Functionalization is an effective method that creates covalent interaction and incorporates carbon nanotubes into the polymer network [100]. Consequently, functionalized nanocomposites behave more uniformly and do not show phase separation and poor dispersion. Superior properties are also more likely to be harnessed with functionalization [100]. However, special attention should be paid to purification and other preparation steps during functionalization [156]. Unfortunately, creating covalent bonds between the carbon nanotubes and the polymer matrix requires several preparation steps [100], and needs to be reconsidered in detail. However, the efficiency of the method along with its promising outcomes, especially when hybrid systems of covalent and noncovalent interactions are used [162], compensates for the extra effort.

7. Property Improvements

Carbon nanotubes possess extremely high thermal and electrical conductivities, a negative coefficient of thermal expansion, and superior mechanical properties [163-170]. Incorporating carbon nanotubes into polymer matrices affects nanocomposite properties accordingly [171-178]. In particular,

the interfacial characteristics directly affect the efficiency of the reinforcements of carbon nanotubes in improving mechanical, thermal, and electrical properties of the polymer nanocomposite [179-187]. The effect of carbon nanotubes is more pronounced when the carbon nanotube-polymer interaction is stronger [188]. It should be noted that the above-mentioned interaction improvement techniques may be employed differently for thermoset [189] and thermoplastic [190] polymers. Thermoset polymers cure irreversibly by forming a three-dimensional network of cross-linked chains. On the other hand, the interaction between the polymer chains in thermoplastics is mostly through van der Waals forces, dipole-dipole interactions or hydrogen bonds [100]. Consequently, thermoplastic polymers may undergo phase transition, e.g., between liquid and solid, by applying or removing heat. Based on the polymer type, i.e., thermoset or thermoplastic, and the selected interaction improvement technique, i.e., covalent or noncovalent, each material system should be considered specifically [100]. For example, when a carbon nanotube is attached to a thermoset polymer through covalent interaction, it becomes part of the three-dimensional network of cross-linked chains. However, for a thermoplastic polymer, a chemically

functionalized carbon nanotube is covalently attached to given polymer chains that interact with other chains through weaker interactions such as van der Waals forces.

A list of nanocomposite property enhancements has been reported in the literature [100]. The amount of enhancement is a direct measure of the interaction quality. Single-walled and multi-walled carbon nanotubes have been incorporated into a variety of thermoset and thermoplastic polymer matrices [191-196]. The carbon nanotube content is usually very low, due to the effectiveness of carbon nanotubes and nanocomposite manufacturing techniques such as solution mixing. However, extremely high carbon nanotube contents have also been achieved by using different manufacturing techniques and various forms of carbon nanotubes such as buckypaper [135, 197-202]. The improvement percentages cover a very wide range up to almost 40 times improvements [100].

While experimental approaches can result in new material systems or manufacturing techniques, realistic and insightful modelling studies optimize the critical parameters and eliminate the need for impossible or costly experiments [100]. For example, monitoring the Raman spectra of nanocomposites with multi-walled carbon nanotubes revealed that the mechanism of load transfer was different under tension and compression [203]. Furthermore, a friction-like interaction was observed and the load transfer was limited to low strains, above which the carbon nanotube slippage significantly reduced the load transfer [204]. On the other hand, finite element modelling showed that nanocomposites containing carbon nanotubes with aspect ratio of 300 behaved similar to those with continuous carbon nanotubes [205]. Consequently, when the results of modelling and experimental approaches agree, even if the results are not favorable [206, 207], new lessons are learned and further studies are followed.

According to experimental data, there is a variety of successful modelling techniques that predict properties such as Mori-Tanaka [208-210], Halpin-Tsi [211, 212], self-similar [213-215], equivalent-continuum [33, 213], and multi scale models [33, 216]. Each of these modelling strategies has their own advantages, assumptions, and range of validity [100]. A proper modelling approach will result in trustable results if the assumptions and simulation setups are realistic and the input parameters are chosen properly [152]. Since the modelling studies are not limited by the state-of-the-art experimental capabilities, innovative modelling studies may result in new discoveries and guide the experimental and manufacturing efforts toward efficient and optimum material systems [100]. On the other hand, during the modelling approach, the entire picture should be considered in order to avoid results that are limited by indirect restrictions [95, 100, 162]. The other challenge involved in modelling approaches is providing the simulation with proper input parameters [100, 205].

On the other hand, experimental data are lower than theoretical predictions such as rule of mixtures [100, 207]. The carbon nanotube-polymer interaction is proposed to be an extremely important factor. The interaction defines the load transfer capacity and directly affects the dispersion of carbon

nanotubes in the polymer matrix [217]. Furthermore, the dispersion of carbon nanotubes is significantly affected by the processing parameters during the manufacturing [218]. Consequently, achieving a good dispersion during the preparation phase and before polymerization does not guarantee a strong interaction between the constituents of the final product. Overall, both experimental and modelling studies should consider the effect of processing parameters on the interaction characteristics in order to obtain good interactions in the final nanocomposite [100]. Other influential factors, such as orientation of carbon nanotubes in the polymer matrix [219] and bundle effects, are affected by dispersion [220] and thus by interaction. Consequently, optimizing interaction properties will result in direct and indirect improvements in performance.

8. Applications of Carbon Nanotube-Polymer Composites

8.1. Electrical Conductivity

Conductive carbon nanotube-polymer composites are regarded as promising materials for use in lithium batteries, supercapacitors, polyactuators, iosensors, and flexible transparent electrodes [12-20]. In particular, new transparent and electrically conductive coatings and films have a variety of fast-growing applications ranging from window glass to flat-panel displays [12-20]. Carbon nanotube-polymer composites do not have high enough electrical conductivity to replace metal wiring [5-8]. However, a number of applications exist where the electrical conductivity requirement is not as severe. Two of those applications are electromagnetic interference shielding [221-224] and electrostatic dissipation [225, 226]. The difference between them is the electrical conductivity required; the former requires significantly higher electrical conductivities than the latter. Specifically, a particular material is classified as conductive, static conductive, or static dissipative [13]. Materials classified as conductive are useful for electromagnetic interference shielding. Metal cages are used where the shielding requirements are severe. The third application is due to an inherent property of all metals: metals are not optically transparent. Specifically, the requirement is for transparent electrodes [227, 228], for example, the connection between a current source and an active component or between an active component and a current sink [14]. Very thin films of carbon nanotubes with high conductivities can be made and polymers can assist in this process. Flexible electronics, where the carbon nanotubes are deposited on a polymeric substrate, are probably the most common use of carbon nanotubes as transparent electrodes.

8.2. Thermal Properties

Significant commercial volume for carbon nanotubes in polymers exists if high thermal conductivities can be achieved [6, 8-20]. Without a doubt, infused systems offer some promise

and perhaps in the future carbon nanotubes will be used as layers and coatings designed to dissipate heat. Another application where the high thermal conductivity of carbon nanotubes may play a role in improved performance is flame retardancy [229-234]. Since the mechanism of flame retardancy for carbon nanotubes is as a protective layer [235], it is worth considering this mechanism in more detail. A protective surface layer reduces flammability by reducing the diffusion of oxygen to the surface, reducing the diffusion of gaseous degradation products to the air, and finally acting as a thermal insulating layer reducing the temperature of the polymer underneath [14]. The increase in melt viscosity caused by carbon nanotubes could also contribute to the reduction in flammability [14]. An increase in thermal conductivity could also play an important role if the coating is sufficiently conductive. Carbon nanotubes themselves are flammable at high temperatures; because polymers are less thermally stable than carbon nanotubes, carbon nanotubes are effective flame retardants for polymers. In tests that involve heat release, carbon nanotubes tend to improve the performance of nanocomposites. However, time to ignition has been found to be quicker with carbon nanotube addition, which can be attributed to heat localization due primarily to the high thermal conductivity and low specific heat [232]. Carbon nanotube-filled materials have also been reported to have problems with mass loss on ignition tests [236-238]. Carbon nanotubes have been found to be more effective retardants with respect to heat release at similar loading levels [239, 240].

8.3. Electromechanical Properties

Carbon nanotubes have the physical property that a change in their mechanical state can induce a change in their ability to conduct electricity, and vice versa [241, 242]. In electromechanical actuation, carbon nanotube-containing materials are connected to a voltage source only at one end. Since no connection exists, current is not able to flow, instead there is a buildup of charge. This buildup of charge can cause a change in the shape of the object, with the change depending on the original object shape, voltage applied, and so on. In strain sensors, a mechanical strain imposed on the carbon nanotubes alters the electronic conductivity, either in an individual sense with respect to individual carbon nanotubes or in a global sense with respect to a carbon nanotube network. Strain sensors work on the principle of measuring a change in electronic properties and converting that change to a strain. A number of theoretical and experimental studies have examined pristine carbon nanotube films as strain sensors [243, 244] including carbon nanotubes directly deposited on a polymer substrate [245, 246]. In fact, freestanding carbon nanotube films do not generally make good strain sensors due to slippage between carbon nanotubes [247].

9. Conclusion

Significant progress has been achieved in the area of carbon nanotube-polymer composites over the past two decades. Carbon nanotubes have shown outstanding mechanical,

thermal, and electrical properties. However, at the state of the art, their full potential has not been reached when combined with polymer matrices in nanocomposites. In this work, an overview of the research in carbon nanotube-polymer nanocomposites has been provided, with emphases on the principles of carbon nanotube functionalization. Many techniques have been attempted with varying success to functionalize the inherently inert nature of the surface of carbon nanotubes. There are two major approaches, i.e., chemical and physical functionalization. Fluorination, cycloaddition, carbene and nitrene addition, chlorination, bromination, hydrogenation, and silanization belong to the chemical methods that can provide covalent functional groups onto the surface of carbon nanotubes. The physical methods include wrapping of polymer around the carbon nanotubes, use of surfactants of various ionic nature, and the endohedral method. The state of research into carbon nanotube-polymer composites for mechanical reinforcement has been reviewed, and particular interest is also given to interfacial bonding of carbon nanotubes to polymer matrices as it applies to stress transfer from the polymer matrix to the carbon nanotube. The carbon nanotube-polymer interaction is believed to play an important role in determining the overall properties of the nanocomposites. The interfacial characteristics directly affect the efficiency of carbon nanotube reinforcements in improving thermal, electrical, and mechanical properties of the polymer nanocomposite. The interaction studies in carbon nanotube-polymer composites have been critically reviewed. Different techniques of measuring interaction, including experimental and modelling methods, were described, and advantages and challenges of each method were discussed. Furthermore, various techniques of interaction improvement were discussed under the two main classes of covalent and noncovalent interactions. The excellent mechanical properties of carbon nanotubes combined with unique transport properties render a huge potential for structural and functional applications of carbon nanotube-polymer composites. Although numerous studies have dedicated to the development of carbon nanotube-polymer composites for various purposes, their applications in real products are still in their early stage of realization.

Finally, there is large interest in the further development of carbon nanotube-polymer composite materials, which are promising for a broad range of important applications. A very important area in the near future will be the development of new ultra-strong carbon nanotube-polymer materials for application in bullet-proof vests, protective clothing, high performance composites for aircraft, and automotive industries. These large sectors will require huge quantities of carbon nanotubes. For these reasons the development of new cost-effective carbon nanotube-polymer composites will be necessary to meet these needs.

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