

The Electrical Properties and Conducting Mechanisms of Carbon Nanotube/Polymer Nanocomposites: A Review

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This paper reviews the mechanism of the conducting process of carbon nanotubes (CNTs)-reinforced polymer nanocomposites. Comparison of the two different mechanisms, the formation of the conducting network and the hopping of the electrons, are discussed. The paper also describes the critical factors that determine percolation thresholds or the conductivity of the nanocomposites. By summarizing the predecessors' research, some measures are put forward to improve the structure of the nanocomposites to get the samples that have the most extraordinary electrical conductivity with the lowest CNTs concentrations.

Keywords Carbon nanotube; Electrical conductivity; Nanocomposites; Polymer

INTRODUCTION

After the carbon nanotubes (CNTs) were first time synthesized by Iijima^[1] in the early 1990s, they have been widely used in some fields for their excellent physical and chemical properties^[2–10]. Thostenson^[11] and Dong^[12] have reviewed the extraordinary properties of CNTs in their works. Due to its unusual structure and extended π -electron system, CNTs exhibit a variety of outstanding electronic and electrical properties (1000–200,000 S/cm)^[13,14]. It has also been established that CNTs could exhibit amphoteric behavior by exchanging electrons with electron acceptors or donors to form the corresponding positively or negatively charged counterions^[15].

Some experimental studies on CNT-reinforced polymer materials have been reported for various kinds of organic polymers since the discovery of CNTs, such as: polyethylene^[16–18], polypropylene^[19–21], poly(methylmethacrylate)^[22–24], pitch^[25], epoxy^[26,27], polystyrene^[28], polyurethane^[29,30] and polysilsesquioxane^[31]. Many of these composites are made because when reinforced by CNTs, there will be a more sufficient level of properties compared to base polymer, especially for the electrical conductivity with a increase of about 8 to 13 orders^[15,32–35]. In

Pedroni et al.'s^[36] work, they compared the conductivity of the composites reinforced CNTs and traditional carbon black and found that in the range of filler content investigated, the necessary CNTs is 6–12 times lower than the carbon black concentration.

Sometimes, when prepared by the same polymer, it may get different electrical conductivity or percolation thresholds because there are some uncertainties such as type and quality of nanotubes, different sizes, aspect ratios, crystalline orientation, purity, entanglement, straightness, etc.^[37]. However, the nonreactive surface of the CNTs limits its applications in composites because of lacking adhesion between CNTs and various matrix polymers. Intrinsic van der Waals attraction among tubes, in combination with their high surface area and high aspect ratio, often leads to significant agglomeration, thus preventing efficient transfer of their superior properties to the matrix^[24,29,38,39]. To solve these problems, some physical and chemical approaches have been taken into effect. Nadia^[40] and Vaisman^[41] reviewed these methods, offered us a guided tour through the toolbox for the dispersion of CNTs into polymer matrixes. There are also some researchers^[42] who think that the aspect ratio of the filler, dispersibility and the ability to conglomerate are crucial parameters for the realization of conductive composites at low filler contents. It is hoped that the scholars can get a reference to make the nanocomposites with good electrical conductivity easily and successfully from this paper.

THE CONDUCTIVE MECHANISM

In the nanocomposites reinforced by CNTs, nanotubes are covalently integrated into the matrix and become part of the cross-linked structure rather than just a separated component^[43], as shown schematically in Figure 1. When the percolation threshold is reached, the formation of a percolated filler network structure, where a critical minimum distance between CNTs structures has been attained and electron conduction is facilitated through a 'hopping' or 'tunnelling' mechanism.

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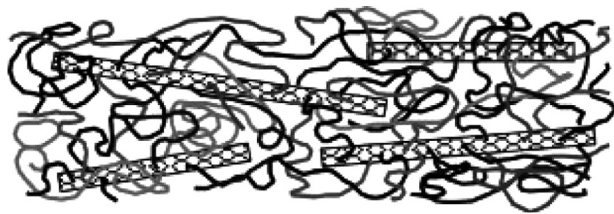


FIG. 1. Integration of nanotubes into the polymer cross-linked structure^[43].

A number of reports^[1,44,45] indicated that the incorporation of graphite with other conductive materials, especially CNTs, has been considered as an effective method to develop higher bulk electrical conductivity of the nanocomposite bipolar plates due to 3D conductive networks. The formation of CNT network^[46] or a continuous electron path^[47–49] is the main cause of the electron transport by tunneling or electron hopping which occurs along CNT interconnects. Garyd^[50] has developed a micromechanics model to assess the impact of the effects of electron hopping and the formation of conductive networks on the electrical conductivity of CNT-polymer nanocomposites.

Some researchers suggested that the low percolation concentrations observed in experiments are due to nanoscale phenomena such as electron hopping^[51–53], while many other investigators focus on dispersion arguments, indicating that the low percolation concentrations observed are due to the formation of conducting networks that allow a high percentage of electrons flow through the composites within the polymer^[54–56]. However, Garyd^[50] considered that for MWCNT nanocomposites, the formation of conductive networks is therefore seen as the principle cause for the large increases in nanocomposite conductivity observed at very low CNT concentrations, while for SWCNT nanocomposites, both the formation of conductive networks and electron hopping lead to large increases in nanocomposite conductivity at very low CNT concentrations, with the formation of conducting networks being the dominant factor at concentrations below the percolation concentration associated with electron hopping.

Ounaies^[54] observed that there are non-linear I–V relationships when the CNTs loadings are in excess of 0.1 vol%, and this non-Ohmic behavior is most likely due to a tunneling mechanism. Conduction may occur by electron hopping from a nanotube to an adjacent one when they are close enough but need not always physically touch each other^[57], conductive paths are formed through the composites due to quantum tunneling effects where the distance between the conductive inclusions is such that electron hopping can occur. Percolation theory assumes paths are made up of conductive inclusions in direct contact.

However, in the case of quantum tunneling, a contact resistance or potential barriers exists within the conductive

path, in this case between 2 adjacent CNTs, which decreases the effective conductivity of the CNTs and then result in the reducing of the conductivity of the nanocomposites^[58,59]. It is reasonable to expect that at much higher CNTs loading, a second threshold would be achieved where the CNTs inclusions contact directly, eliminating the contact resistance. In that case, the conductivity of the nanocomposites would approach the CNTs.

CRITICAL FACTORS THAT AFFECT THE CONDUCTIVITY OF NANOTUBES

Percentage of the CNTs

With increasing MWNT loading, the nanocomposites undergo transition from electrically insulative to conductive and then the electrical conductivity was increase with additional weight percent of CNTs in the matrix^[60–62]. The electrical conductivity of the composites may increase by 10–13 orders by adding the surprisingly volume fraction 0.03% or 0.06%^[42,63].

Percolation theory predicts the relationship between the composite resistivity and volume content of the conductive filler as^[64],

$$\rho = \rho_0(v - v_c)^t \quad (1)$$

where ρ , composite resistivity; ρ_0 , resistivity of conductive filler; v , volume content of filler; v_c , percolation threshold; t , critical exponent. This equation is valid at the concentration above the percolation threshold, i.e., when $v > v_c$. The critical exponent related to the system dimension ranges from 1.6 to 2.0 by theoretical prediction, while experimental values between 0.7 and 3.1 have been reported for the CNT-filled nanocomposites^[54,61,65–68].

The incorporation of a small quantity of CNTs into polymer composites could exhibit higher electrical conductivity due to the formation of an extra effective electrical network^[48]. However, higher CNTs loading in polymer composites may cause serious CNTs aggregation, and the bulk electrical conductivity will be leveled off, even decreased^[69]. In summary, choosing a suitable CNTs percentage is very important.

Dispersion of the CNTs

There are some traditional thoughts that the electrical conductivity will increase by improving the dispersion or reducing the aggregation of the CNTs. Liao^[69] and Kuan^[70] observed that the bulk or the surface electrical resistance decreased as the degree of crystallinity of CNTs/Polymer nanocomposite bipolar plates reduced at the same CNTs loading. This phenomenon may be attributed to the better dispersion of CNTs in low crystalline polymer matrix than high crystalline matrixes^[69,70]. The probability of self-aggregation in the conductive materials will be reduced,

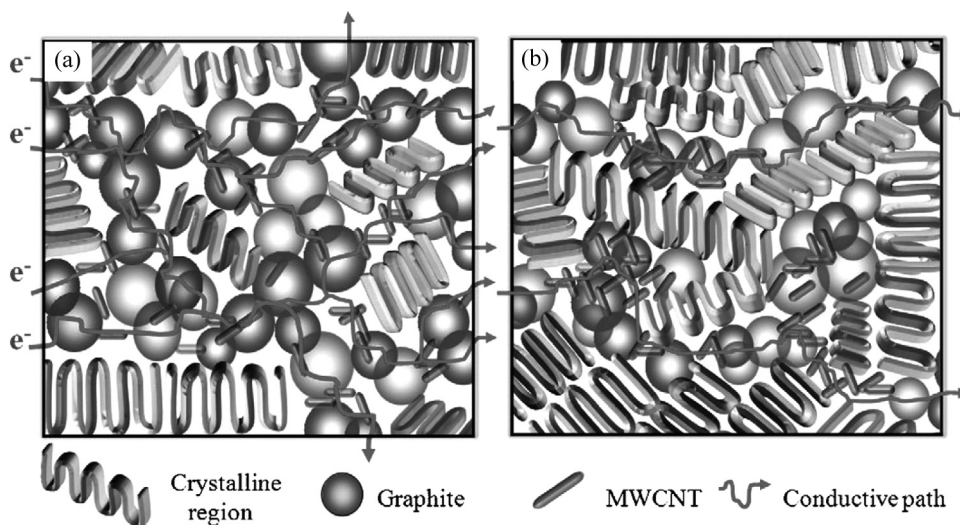


FIG. 2. The model of conductive paths in the nanocomposite bipolar plates with (a) better dispersion of MWCNTs in LC-PP matrix (b) MWCNTs aggregation in HC-PP matrix^[69].

and thus, more electrical conducting paths were built up by the binary conductive materials system (Fig. 2). Mazinani^[71] thought that good dispersion helps distribution of MWCNTs along the fiber axis and increase the conductivity ultimately when below the percolation thresholds, Grossiord^[40] and Xu^[72] pointed out that due to the increasing of the number of complete conductive paths, good dispersion can bring good electrical conductivity.

However, there are also some scholars hold the opposite opinions, they think that the aggregation bring good electrical conductivity with their own reasons: Bryning et al.^[35] found that SWNT aggregation helps because of the continuous nanotube chains that form in the semi-dilute concentration regime, Martin et al.^[66], Du^[55] and Hu^[68] considered that electrical conductivities or low percolation thresholds can be accomplished via heterogeneous distributions of SWNT in polymer matrices. Gojny et al.^[42] showed that the stable dispersion can be regarded as detrimental for the overall conductivity because it led to the formation of an isolating epoxy layer around the nanotubes, and Delozier et al.^[44] thought that more uniform distribution of SWNT bundles would result in the decreasing of the likelihood of bundles touching to form a conductive network and at last the electrical conductivity would fall.

Confront with these two contradictions, Li^[73] raised a model to summarize the effects of the dispersion, they thought that in practical nanocomposites, the percolation threshold of these nanocomposites can be expressed by

$$P_c = \frac{\xi \varepsilon \pi}{6} + \frac{(1 - \xi) 27 \pi d^2}{4 l^2} \quad (2)$$

where P_c , percolation thresholds; d , diameter of the CNTs; l , length of the CNTs; ε , the localized volume content of CNTs in an agglomerate; ξ , the volume fraction of agglomerated CNTs. From this model we can found that the dispersion has great effect to the percolation, but we cannot simply estimate how the value of the percolation would change when improve the dispersion.

Concerning about how to evaluate the dispersion, Peter^[74] has developed a method to quantitatively assess the dispersion of the CNTs in the composites by using the protocol of high kV imaging and 3-dimensional image analysis, it showed that nanocomposites can be sampled on a sufficiently large volume to differentiate the extent of dispersion.

Structure of the CNTs

After MWCNT were first time introduced by Iijima, SWCNT was then discovered successively also by him in 1993^[75]. The electronic properties of perfect MWCNT are rather similar to those of perfect SWCNT. However, due to weak coupling between the concentric cylinders of the MWCNT, only the outer shell contributes to the electron transport and thus to the final electronic properties^[40].

It was found by Gojny et al.^[42] that the increase in conductivity with weight percent loading of nanotubes was virtually independent of type of non-functionalized nanotubes. However, when volume fraction is considered, the nanoparticles with the highest densities show the lowest percolation thresholds. Consequently, MWCNTs exhibit the highest potential for an efficient enhancement of the electrical conductivity, due to the relatively low surface area and high aspect ratio that enables a good dispersibility.

It has been shown, both experimentally and theoretically, that the percolation threshold strongly depends on the aspect ratio (length-to-diameter ratio) of the filler particles^[73,76,77]. At similar states of CNTs orientation, the higher the aspect ratio, the lower the CNTs concentration necessary to reach the percolation threshold, and to get conductive films^[40,78], and Rosca^[79] even found that the composite conductivity increases almost 10 times as the aspect ratio increases 5.5 times.

Besides these, Sandler^[67] used the chemical vapour deposition process to get the aligned MWNTs, it is shown that sufficient conductivity for anti-static applications can be achieved at an average nanotube loading of approximately 0.005 wt%.

Preparation Methods

There are a lot of methods to prepare the nanocomposites, such as: solution-mixing method, melt blending, shear mixing, in-situ polymerization and so on. Because the fabrication process has a significantly effect on the formation of conducting network^[80], to make the electrical properties of nanocomposites better, we should use different methods in different circumstances.

In Yuen's work^[81], acid-modified CNTs were grafted with soluble polyimide then was added to the polyamic acid and heated to 300°C to form CNTs/polyimide composites via imidation, he found that compared with the acid-modified CNTs/polyimide composites, because of the show micro phase separation in the insoluble polyimide matrix, the CNTs connected each other and thus increase the electrical conducting effectively. Lanticse^[82] found that the doctor blade technique induced preferential alignment of the nanotubes in composite and a higher degree of alignment is achieved in composites with lower contents of nanotubes.

It was shown that this technique caused some nanotube concentrations at the central section of the composite thereby increasing the effective local concentration of nanotubes that consequently, increased the amount of nanotube contacts in the composite and effectively increased the electrical conductivity of the composite up to 6 orders of magnitude higher. In Rosca's work^[79], 3-roll milling was successfully used to form a homogeneous MWCNT-epoxy dispersions with nanotube loading form 0.015 up to 8 wt%.

Shear intensity and number of passes determine the nanotube dispersion and consequently the composite conductivity. The highest conductivity was observed at medium shear intensities and moderate number of passes, as insufficient dispersion at low shear intensity or over-dispersion at high shear intensity and increased number of passes affects the composite conductivity. Cipriano et al.^[83] showed that melt annealing at temperatures above the polymer's glass transition temperature is also an

efficient preparing method to make nanocomposites of high conductivity, the mechanism can be explained that during processing, the particles become aligned in the flow direction, leading to a decrease in the connectivity of the particle network, the same connections, however, are re-established during annealing.

Type of Polymers

Due to the different structures and different properties especially electrical conductivity of different kinds of polymers, when reinforced by CNTs, the nanocomposites will show different electrical percolation thresholds. The electrical percolation thresholds for CNT-filled nanocomposites with thermosets such as epoxy resin^[84] and polyimide^[54] usually lie in the range from 0.1 to 1 wt%, while the values for thermoplastics such as polyethylene^[85,86], polypropylene^[87] polyamide^[88], polycarbonate^[89], poly(ethylene terephthalate) and polystyrene^[65] lie between 0.2 and 15 wt%. Bin et al.^[86] prepared MWNT/high-density polyethylene by gelation/crystallization from solutions, and revealed that the percolation occurred between 5 and 15 wt% and it was also found by Meincke et al.^[88] that CNT-filled polyamide-6 showed an onset of electrical conductivity at CNT loadings of 4–6 wt%. However, in Kharchenko's work^[87], he made the polypropylene/MWNT nanocomposites by melt blending, and it was displayed that the percolation thresholds for conductivity and firmness were both at the concentration ranging from only 0.25 to 1 wt% at 200°C.

Nogales et al.^[90] prepared SWNT filled poly(butylene terephthalate) by in situ polymerization and reported achievement of low percolation threshold around 0.2 wt% of SWNT, Du^[51,91] used the coagulation method to produce SWNT/poly(methyl methacrylate) (PMMA) nanocomposites, and disclosed percolation threshold between 0.2 and 2 wt%.

The Electrical Anisotropy of the Nanocomposites

In Lanticse's work^[82], the nanocomposites were prepared by the doctor blade technique. It was shown that the anisotropy in conductivity is not limited to direction parallel and perpendicular to the blading direction of the composite film but is also exhibited at the thickness direction of both the film and bulk composites. Slight tilting of the nanotubes in the composite film could easily create additional contacts between nanotubes resulting into higher electrical conductivities in the film than in the bulk.

MEASURES TO IMPROVE THE ELECTRICAL PROPERTIES

There are 3 methods to improve the dispersion of the CNTs in the polymer; the first is direct mixing of the CNTs and the polymer, next is the modification of either the polymer matrix or the NT walls, and the last and the most

promised one is the addition of the third component^[40]. Delozier et al.^[44] thought that the most effective means for dispersing SWNTs is the use of non-covalent modification, a technique that does not alter the properties of SWNTs.

Direct mixing of the CNTs and the polymer appears to be the easiest and least laborious way to achieve the incorporation of CNTs into a polymer matrix and had proven its efficiency, on the other hand it brings the inhomogeneous, but these agglomerated can be a key factor in lowering considerably the value of the percolation threshold^[67]. Modifying either the CNTs walls^[92,93] by functionalization or the polymer matrix^[94], is found to improve the quality of the interface between the components of the nanocomposite, but unluckily, both approaches lead to the disturbance of the delocalization of the π -electrons of the CNTs walls, which results in a significant deterioration of its electrical properties. Finally, the third strategy is based on the use of a third component, which most frequently is a surfactant. The methods inspired by this strategy are mainly based on latex technology. The technique^[41,95] has been reported to be very successful to produce conductive nanocomposites with low percolation thresholds, as well as good conductivity levels.

Surfactant Treatment

The physical adsorption of surfactant on the CNT surface lowered the surface tension of CNTs, effectively preventing the formation of aggregates (Fig. 3). Furthermore, the surfactant-treated CNTs overcame van der Waals attraction by electrostatic/steric repulsive forces promoted by the surfactant treatment^[40,96]. The mechanical properties as well as the thermomechanical properties both showed significant improvements after the treatment; however, the electrical conductivities of the CNT nanocomposites with and without surfactant treatment were similar for the CNT contents studied, indicating little adverse effect due to wrapping of CNTs with surfactant^[39].

Sometimes, with further increase in CNT content beyond the percolation threshold, the electrical conductivities of the surfactant-treated CNT higher than the pristine CNT counterpart. It can be explained that the improved CNT dispersion due to surfactant helped better form the conductive networks within the polymer matrix, giving rise to an increase in the electrical conductivity at a low range of CNT content^[73]; however, in Mazinani's literature^[71], he had his opinions contract with this, he thought that when below the percolation thresholds, surfactant helped distribution of MWCNTs along fiber axis while beyond the percolation thresholds, addition of surfactant partially coats the MWCNTs and therefore in spite of forming networks; it could reduce the electrical conduction as has been previously observed^[97]. But when treated by silane, the electrical conductivity was significantly lower than the

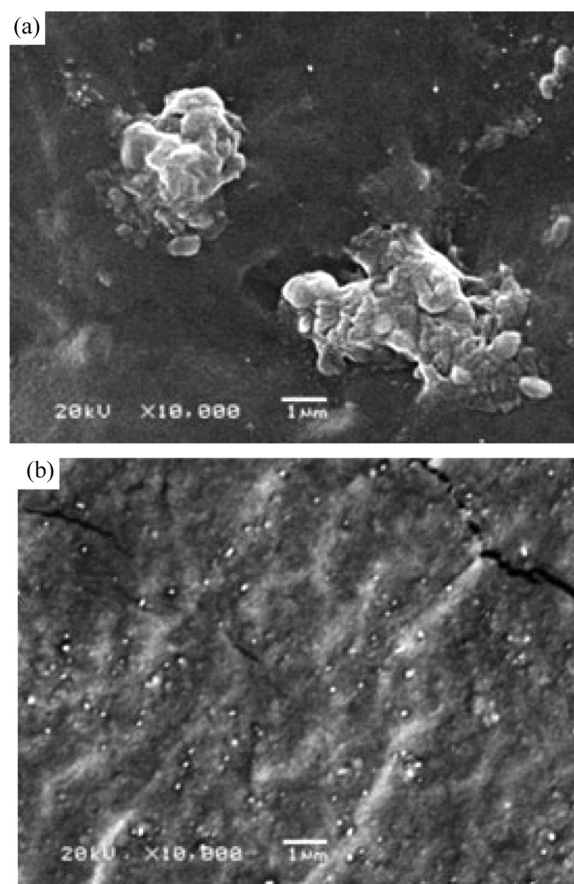


FIG. 3. SEM images of composites based on SBS and 4wt% of MWCNTs: (a) in the absence of the dispersant agent and (b) with 1 wt% of the dispersant^[36].

composites which were treated by another surfactant, because of the wrapping of silane-treated CNTs by covalently-bonded epoxy after curing, which in turn disturbed the π -electron system of CNT walls^[39,98].

Generally, ionic surfactants are preferable for CNT/water-soluble solutions, because that once spaces or gaps at the CNTs bundle ends are formed, they are propagated by the ionic surfactant adsorption, ultimately separating the individual nanotubes from the bundle. Alternatively, nonionic surfactants are proposed when organic solvents have to be used^[41,95].

Dealing with the CNTs

Functionalization of CNTs is also a viable means of enhancing dispersion, but this method changes the hybridization of the carbon atoms on the CNTs, and thus the properties can also be changed^[44].

Liu^[99] developed a facile route to modify CNTs with commercial-available engineering plastics such as PPO. The multi-bonded PPO chains in CNT-PPO significantly improve the compatibility and dispersion ability of CNT

in the PPO matrix, so as to increase the electrical conductivity of the resulting PPO/CNT nanocomposites.

Acid-Treated Methods

The increase in the electrical conductivity might have been due to the decrease in impure materials on the A-CNT surface by acid treatment up to 30 min, this indicates that the acid treatment leads to a reduced agglomeration of CNTs^[29] and Kwon^[30] also found that because of the smaller particle size and the good dispersion of the acid-treated MWCNTs in the water-borne polyurethane, the conductivity of the A-CNT/WBPU composites were higher than those of the WBPU/CNT composites, and this was also contributed to the high conductivity of pure A-CNT in comparison with that of pure CNT. However, there may be some decrease in electrical conductivity after 30 min of nitric acid treatment, which is presumably due to band gap enlargement and damage of the side wall of the CNTs.

Physical Methods

The use of mechanical means like high powered ultrasonication or shearing forces like mechanical stirring^[47] to disperse SWNTs is effective but may decrease the length and consequently alter the properties of the SWNTs.

When the composites were prepared by the melt-mixing methods, the screw speed and the mixing time showed both significantly effects to the electrical conductivity^[100]. The increase of the mixing time can improve the distribution of MWNT in the polymer matrix considerably. At concentrations near the percolation threshold, enhanced mixing time can transform the system from a non-percolated into a percolated one. The effect of mixing time is explained by the time necessary for diffusion of the polymer chains between the MWNT aggregates or bundles of the master batch.

The screw speed had also a significant effect on the dispersion of MWNT. At MWNT contents below percolation thresholds; increased screw speed induces better dispersion and then higher conductivity, whereas above percolation thresholds the efficient transfer of mechanical energy due to nanotube network formation can lead to enhanced breakage of the MWNT and decrease of DC conductivity and crossover frequency. Some literature articles^[80,101,102] have pointed out that the best dispersion attained, showing the lowest percolation threshold, did not correspond to the most energetic mixing conditions, at the medium shear mixing conditions. When the nanocomposites were treated by ultrasonication, the longer the sonication duration, the smaller the MWCNTs agglomerate (Fig. 4) and then the better conductivity^[47].

Beside the methods that mentioned above, it was shown by other researchers^[67,72,78,103,104] that aligning nanotubes to a certain direction through the use of electric^[78,104]

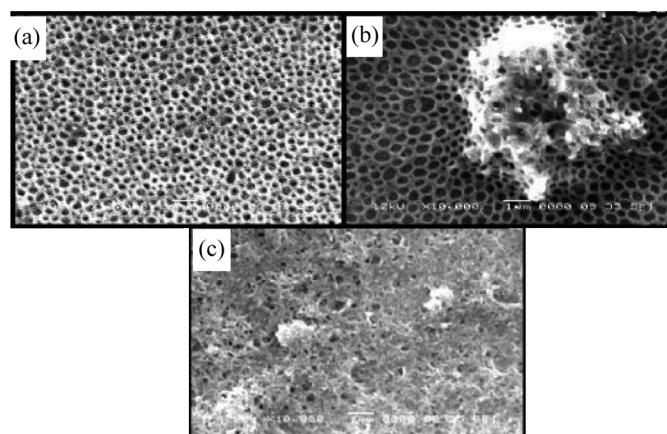


FIG. 4. SEM images of synthesized PMMA/MWCNTs nanocomposite thin films of (a) pure PMMA (b) 1.5% MWCNTs with 1 hr sonication and (c) 1.5% MWCNTs with 5 hrs sonication^[47].

and magnetic^[72] forces as well as flow-field mechanics^[103] could increase electrical conductivity. Some researchers^[66,72,79,80] also found that at low concentrations, change the using environment such as the temperature is very important as temperature determines the viscosity of the medium and mobility of the nanotubes, higher temperatures facilitate the re-aggregation of the MWCNTs into a more conductive network. On the contrary, at high nanotube loadings the temperature has only marginal effect on the system viscosity and nanotube mobility, that translates to only a small change in the conductivity.

THE APPLICATION OF CNTs/POLYMER COMPOSITES IN ELECTRICAL FIELD

Nanocomposites have widely applications in the electrical field. For example, polymer/CNT nanocomposites are being actively used in aerospace applications requiring electrical conductivity for dissipating electrostatic charges and electromagnetic interference shielding^[105]. Using surface modified CNTs (f-CNTs), Abraham^[106] fabricated chemiresistor thin films on a printed circuit board integrated to a wireless system using signal conditioning circuits. The gas detection principle is based on the change in electrical resistance of the thin films due to the presence of organic vapors. Beside these, the Polymer/CNTs nanocomposites also have advantages for device applications such as solar cells^[107], sensors^[108] and protective applications^[109].

REMARKS

The exceptional mechanical and physical properties demonstrated for carbon nanotubes, combined with their low density, make this new form of carbon an excellent candidate for nanocomposite reinforcement. And there is currently a growing interest in processing CNTs polymer electrical conducting nanocomposites from the point of

view that small quantity of addition can modify considerably the electrical behavior of a polymer matrix. It should be also emphasized that the importance of factors which affect the electrical properties. To make the most extraordinary electrical properties at the lowest percentage of the CNTs, some strategies such as surfactant treating, acid-treated and dealing with CNTs are needed. We did not have the pretension to give an exhaustive overview, but the aim was rather to illustrate the main strategies with some appealing examples.

Preliminary research in nanotube-based conductive composites has indicated that there is potential in carbon nanotubes for reinforcement, but, most importantly, it has illustrated the significant challenges that must be overcome before the potential is realized. First, some preparation technologies adopted currently are suitable to manufacture micrometer composites while insufficient to make the nanometer composites. There is a need for development of nanotube production techniques at the scale needed for producing macroscopic conductive composites that are cost-effective. Second, for the purpose of bulk production and cost reduction of the conductive nanocomposites, construction of the assembly line is necessary and imperative.

Third, it is very important to take intensive study about the compatibility of the interface because excellent interfacial compatibility will strengthen the interface action, promote dispersion of CNTs in the polymer and bring the extraordinary electrical conductivity ultimately. Finally, the extensive application of conductive nanocomposites has not been successfully developed. These issues are not only challenges but also opportunities to us. It is hoped and also believed that in the near future, the conductive nanocomposites will be widely used, such as the electrical wire, the antistatic instruments and so on.

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