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Preparation, Solubility, and Electrical Properties of Multiwalled Carbon Nanotubes/Poly(1-vinyl-1,2,4-triazole) Composites via in situ Functionalization

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The chemically modified multiwalled carbon nanotubes (MWCNT)/poly(1-vinyl-1,2,4-triazole) (PVTri) composites were prepared via in situ emulsion polymerization. The morphological and structural properties of the surface-modified MWCNT and the synthesized MWCNT/PVTri composites were characterized by Fourier transform infrared spectroscopy, X-ray diffraction, scanning electron microscopy, and transmission electron microscopy. The electrical properties of the composite material were analyzed by conductivity measurements. The thermal analysis and the Tg value of the polymer were determined by differential scanning calorimetry technique. The obtained novel MWCNT/PVTri composites exhibited excellent solubility and possessed a slightly higher glass transition temperature than that of the pure polymer.

Keywords Electrical properties; Multiwalled carbon nanotubes; Poly(1-vinyl-1,2,4-triazole); Polymeric nanocomposites; Surface properties

INTRODUCTION

Since their discovery in 1991, carbon nanotubes (CNTs) have generated interest in most areas of science and engineering due to their unprecedented physical and chemical properties[1]. Due to their high mechanical strength, aspect ratio, thermal conductivity, and electric conductivity, CNTs are attractive for polymeric composites as ideal reinforcement filling materials[2-7]. Polymer/CNT composites are expected to possess the good processability characteristics of the polymer and the excellent functional properties of the CNT[8-10]. However, the preparation of such materials is challenging due to ensuring the uniform dispersion of CNTs in the polymer matrices without agglomerates and adequate interfacial adhesion between the CNTs and polymers.

The failure of CNTs to disperse leads to a phase separation due to the strong surface interactions between the tubes. It is likely that chemical functionalization would not only serve to facilitate dispersion but would also stabilize the CNT and prevent agglomerations that could lead to defects[11]. To overcome this problem, chemical modification of the CNT surface is expected to play a key role in the disruption of the rope structure and the addition of reactive species to the nanotube to improve interfacial bonding and charge transfer in the resulting composites[10].

Carbon nanotubes can be divided into two categories as follows: single-wall carbon nanotubes (SWCNTs) and multiwall carbon nanotubes (MWCNTs). This classification depends on the folding angle and the diameter. SWCNTs can be metallic, insulating or semiconducting, while MWCNTs are always conductive[12]. SWCNTs and MWCNTs have been extensively studied in both fundamental and applied research fields[13-16]. MWCNTs are commonly used in composite research because their production is fast and inexpensive. In addition, MWCNTs are much cheaper than SWCNTs, and better suited to large scale industrial applications[17]. One drawback of MWCNTs is that their conductivity is typically one or two orders lower than that of SWCNTs[18].

Investigation of polymers containing nitrogen donor atoms is of interest due to a variety of applications, such as destabilizing negative colloids in effluents and water clarification, electrophoresis deposition, curing agents, activating electrode processes, recovery of trace metal ions, and anticorrosion coatings[19]. Poly(1-vinyl-1,2,4-triazole) (PVTri) is of great interest because it is relatively simple to synthesize and can generate permanent positive charge...
dyes [20]. PVTri is nontoxic, biocompatible, and easily soluble in water or polar organic solvents [21]. Recently, acid doped PVTri was studied as a promising proton conducting polymer electrolyte in proton exchange membrane fuel cells [22]. In addition, PVTri is thermally and electrochemically stable polymer and it can be used for high temperature applications [23].

In a previous study [24], we investigated in situ polymerization synthesis and characterization of single wall nanotube/poly(1-vinyl-1,2,4-triazole) nanocomposites. The structural and morphologic characterizations indicated that the SWCNT/PVTri composite was obtained. The DSC analysis demonstrated that the high thermal stability of the resulting composite material. In addition, the electrical and dielectric properties of SWCNT/PVTri composites were investigated. Due to the formation of a nanotube network in the polymer media, the composite material exhibited high electrical conductivity.

The primary purpose of this study is to synthesize and characterize the MWCNT/PVTri composites. In addition, the MWCNT/PVTri composite obtained in this study is compared to the SWCNT/PVTri composite obtained in a previous study. Many research studies have addressed the attachment of synthetic polymers onto multiwalled carbon nanotubes (MWCNTs), which can be used in the development of materials with new optical, magnetic, and electrical properties [16, 24, 25]. To the best of our knowledge, this study is the first to report the fabrication and evaluation of MWCNT/PVTri composites. In this study, chemically functionalized MWCNT/PVTri composites are produced using an in situ process. The functionalized MWCNT and the final composites have been characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), and Fourier transform infrared spectroscopy (FTIR). Thermal analysis was performed with differential scanning calorimetry (DSC). In addition, the electric conductivity of the MWCNTs and MWCNT/PVTri composites were measured and are discussed in this article.

EXPERIMENTAL

MWCNT Synthesis

MWCNTs was synthesized by the catalytic decomposition of acetylene (5% C2H2/N2) over a nickel catalyst. A calculated amount of metal salt [Ni(NO3)2·6H2O] was dissolved in distilled water, and the MgO support was added to the solution for catalyst preparation. The resulting mixture was placed in a beaker on a hot plate, and the water was evaporated while stirring. The as-prepared sample was dried overnight at 130°C resulting in a dry powder. The decomposition of acetylene was carried out in a vertical fixed-bed flow reactor at 600°C, and the supported catalyst was performed in a quartz boat that was inserted in a quartz tube.

After the reduction of the catalyst under a hydrogen flow, a 112 ml/min flow of an acetylene/nitrogen gas mixture was introduced into the chamber at 600°C. The temperature was increased to 700°C in 10 min and maintained for 5 min. Finally the temperature was set at 600°C for 3 h. After the growth of the CNTs, the furnace was cooled to room temperature under a flow of nitrogen gas. The CVD reactor scheme that was used to synthesize the MWCNTs is shown in Fig. 1.

Chemical Modification of MWCNT

MWCNTs were sonicated in a mixture of concentrated sulfuric and nitric acid (3:1 by volume) for 24 h at 200 rpm and 40°C. Then, the obtained carboxyl terminated MWCNTs (0.5 g) were dispersed in 25 mL of anhydrous ethanol γ-Methacryloxypropyl trimethoxysilane (KH-570) (0.5 mmol), which was diluted in 25 mL of anhydrous ethanol, was added dropwise to the aforementioned solution for 12 h. The resulting mixture was maintained for 36 h at the same temperature and mixing speed, and the chemical modifications of the MWCNTs were achieved. After being centrifuged at 5000 rpm for 60 min, the prepared material was dispersed in anhydrous ethanol, and the prepared material was dispersed process was repeated 5 times. After being dried for 48 h at 40°C, the MWCNTs were obtained with the desired chemical modifications [24, 25].

In situ Polymerization of PVTri with MWCNT

The chemically modified MWCNTs that were previously synthesized (0.12 g, 10 mmol), lauryl sodium sulfate (0.05 g, 1.4 mmol), and sodium bicarbonate (0.25 g, 3.0 mmol) were added to 150 mL of water under stirring at 40°C. Then, 1-vinyl-1,2,4-triazoles (2.85 g, 30 mmol) was added to the solution under a nitrogen atmosphere and was preemulsified for 14 hours. The explosive compound ammonium persulfate (0.15 g, 0.7 mmol), which was dissolved in 20 mL of water, was added dropwise to the abovementioned flask and maintained for 12 h at 80°C. Then, the monomers and surfactants in the suspension were removed under centrifugation at 3000 rpm to yield the stable MWCNT/PVTri composite. The obtained MWCNT/PVTri composite was centrifuged several times and washed with 50 mL of methanol to completely remove the excess poly(vinyl)triazoles, monomer, and surfactants which were annealed for 24 hours at 65°C under vacuum [24, 25].

Characterization Techniques

The polymer and composite structures were characterized using infrared spectroscopy. The infrared spectra were recorded on a Bruker Tensor 27 spectrophotometer in the range of 4000-400 cm−1, with a 2 cm−1 resolution over 30 scans, using Ge ATR. X-ray diffraction (XRD) patterns were recorded on a Bruker AXS Diffractometer D8 and a rotating anode X-ray generator working at 40 kV and
40 mA with a CuKα (λ = 1.54 Å) monochromatic radiation device. The morphology of the MWCNT/PVTri composite was characterized using a Philips XL30 SFEG SEM and a Philips 200 kV Field Emission TEM. DSC thermograms for the PVTri and MWCNT/PVTri composites were obtained with a NETZSCH DSC 404C apparatus.

To determine the glass transition temperature of the polymer and composite, the heat flow was measured under an Ar flow (50 ml/min) by increasing the sample temperature at a rate of 10 K/min to 500°C. The sample size of the PVTri polymer was 5.7 mg, and the sample size of the MWCNT/PVTri composite was 113.0 mg. The electric measurements of the MWCNTs, PVTri, and MWCNT/PVTri composites were performed using a picoamperometer (Keithley 6487). The dielectric measurements of the MWCNT/PVTri sample were performed between 20 Hz and 1 MHz at room temperature with a LCR Meter (Agilent 4284 A). The samples were cold-pressed into disks that were 6 mm in diameter and 1 mm in thickness.

RESULTS AND DISCUSSION
Preparation and Characterization of MWCNT/PVTri Composite

The chemical functionalization of CNTs is one of the few methods that enhance the interfacial adhesion between the nanotubes and the matrix[2]. The process generates functional groups at the surfaces of the CNTs and these functional groups can react with other chemicals and polymers resulting in an enhancement in the interfacial bond between the matrix and the tubes for their application in polymer composites. The silanization of functionalized nanotubes is used to enhance the interfacial adhesion between the nanotubes and the matrix, and the R functional group of the silane coupling agent readily reacts with the hydroxyl groups on the CNT surface, which are produced via oxidation[26].

In our previous studies[16,24], we have shown that silane modification of CNTs with 3-(trimethoxy)-propylmethacrylate-silane is an effective method for producing functional groups on the surface of CNTs, which promotes better dispersion in the polymer matrix. In this study, 3-(trimethoxy)-propylmethacrylate-silane was used as the silane agent to treat MWCNTs, which were functionalized by oxidation with a mixture of concentrated HNO3 and H2SO4. The functionalized MWCNTs were chemically modified by covalent bonding of the 3-(trimethoxy)-propylmethacrylate-silane with oxidized CNTs.

This report examines the reactions for the preparation of MWCNT/PVTri, and the chemical modification and emulsion polymerization were performed as shown in Figure 2. The methoxy groups of KH-570 were hydrolyzed with the hydroxyl groups on the surface of MWCNT under the influence of a base. A chemical-bond linkage was formed between KH-570 and the MWCNT. The methacrylate group of the KH-570 reacts with the C\(\equiv\)C group of 1-vinyl-1,2,4-triazoles monomer via free radical polymerization. Finally, the MWCNT/PVTri composite is formed. We found that the previously reported synthesis method was also suitable for preparation of the MWCNT/PVTri composite. The separation of the free monomer, poly (1-vinyl-1,2,4-triazole), and surfactant from the MWCNT/PVTri composite was achieved by repeated centrifugations to remove the unreacted compounds.

The chemical groups on the surface of the MWCNTs were investigated using FTIR spectroscopy, as shown in Figure 3. After treatment with H2SO4/HNO3, the MWCNTs exhibited an IR peak at 1722 cm\(^{-1}\) (Fig. 3a), which is indicative of the C=O stretching vibration of carboxylic acid groups. The broad peak at 3399 cm\(^{-1}\) is
assigned to the O-H stretches, while the peak at 1587 cm⁻¹ is attributed to the MWCNT C=C stretching mode associated with sidewall attachment⁵,¹³. The MWCNT/PVTri composite exhibited a new absorption band at 1562 cm⁻¹ (Fig. 3b), which is associated with the C=N stretching band. The pristine PVTri²⁷ exhibited one absorption weak at 1508 cm⁻¹ (Fig. 3c). Two additional bands at 1340 and 921 cm⁻¹ were assigned to the ring vibration and the nitrogenous penta-heterocyclic compound stretching bands, respectively, in the MWCNT/PVTri composite (Fig. 3b). However, the presence of MWCNTs in the polymer matrix resulted in very little change in the FTIR spectrum, which is most likely due to the low MWCNT composition and the weak vibration signals of the MWCNTs. However, the FTIR spectrum of SWCNT/PVTri exhibited significant differences from the spectrum of modified SWCNT and pure PVTri²⁴. Therefore, the results indicate that interfacial interaction between the functionalized SWCNTs and the PVTri matrix and the dispersion of the functionalized SWCNTs in the PVTri matrix are better than the interfacial interaction and dispersion of the MWCNTs and PVTri.

The XRD patterns of the MWCNT/PVTri composite PVTri, and acid (HNO₃) treated MWCNT samples are shown in Figure 4. The MWCNTs (Fig. 4a) exhibit two intense peaks at 26.019° and 43.023° corresponding to the graphite reflection. The peaks at 35.055° correspond to NO₂-C formed after acid treatment. The PVTri sample (Fig. 4b) exhibited only one intense peak indicating that the material is not crystalline. A comparison between the XRD powder pattern of the MWCNT/PVTri (Fig. 4c) composite and the MWCNTs indicates that some peaks, such as 35.055°, 61.500°, and 85.159°, disappeared. The peaks of the MWCNT sample that appeared at 26.019°, 22.435° and 43.023° are shifted to 21.401°, 26.020° and 42.840°, respectively, in the MWCNT/PVTri composite XRD spectra. The crystallization degree of the polymer changes upon introduction of the MWCNTs into the PVTri matrix. A combination of structural characterization and SEM analysis confirmed the formation of the MWCNT/PVTri composite.

**Morphological Properties**

The surface morphology of the materials was analyzed by HRTEM and SEM under high magnification. The HRTEM images of the surface functionalized MWCNTs are shown in Figure 5. Figure 5a shows a general view of the MWCNT sample after acid treatment and air oxidation. The unreacted metallic catalyst particles have disappeared. The MWCNT sample that appeared at 26.019°, 22.435° and 43.023° are shifted to 21.401°, 26.020° and 42.840°, respectively, in the MWCNT/PVTri composite XRD spectra. The crystallization degree of the polymer changes upon introduction of the MWCNTs into the PVTri matrix. A combination of structural characterization and SEM analysis confirmed the formation of the MWCNT/PVTri composite.
from another one by a distance of 0.37 nm. At this magnification, on the external planes, damage created by the acid treatment and air oxidation can be observed.

The surface functionalized MWCNT is shown in Figure 6. Figures 6a and 6b show the fibrous structures (MWCNT) at the edge or borders of the polymer. Some MWCNTs formed bridges between the polymer/MWCNT aggregates. These bundles are fully encapsulated by PVTri.

**Solubility Properties**

Figure 7 shows the solubility of MWCNT and MWCNT/PVTri in ethanol and dimethylformamide (DMF). The stability of the dispersion 2 weeks after ultrasonication of the samples is compared. The MWCNT exhibited poor dispersion in ethanol (Fig. 7a) and DMF (Fig. 7c). The MWCNT/PVTri composites dissolved in ethanol (Fig. 7b) and DMF (Fig. 7d) resulting in homogenous solution, which was very stable even after prolonged standing for 2 weeks. Similar to the solubility results reported in a previous study\[24\], neither sedimentation nor aggregation of the nanotube bundles was observed. Because
PVTri was covalently bonded to MWCNT, its surface was altered resulting in a hydrophobic surface, which is responsible for the enhanced stability of the MWCNTs in common organic solvents.

Thermal Analysis

The glass transition temperature ($T_g$) of the pure PVTri was measured at the Tubitak center (Turkey) at 165 °C. The incorporation of CNTs into this polymer matrix should increase the $T_g$ of the polymer resulting in the formation of a strong bond between the polymer matrix and the CNTs. DSC thermal analysis was performed on the PVTri and MWCNT/PVTri composite. The DSC thermogram of PVTri and MWCNT/PVTri are shown in Figures 8a and 8b, respectively. Although the glass transition temperature of PVTri is 165 °C, the $T_g$ value of MWCNT/PVTri, which is measured to be approximately 220 °C, is higher than that of pure PVTri.

The increase in the $T_g$ of the silanized composite is due to the silane molecules forming covalent bonds with the CNTs producing a higher cross-linked network compared to that in the unmodified and oxidized composites [28,29].

The silane modification of the CNTs induces improved interfacial strength between the CNTs and polymer matrix, which reduces the mobility of the polymer matrix around the CNTs. The attached silane groups are expected to improve the interaction between the modified MWCNTs and the polymer molecule chains [26].

However, in a previous study [24], the $T_g$ of SWCNT/PVTri was determined to be approximately 300 °C. The $T_g$ value for SWCNT/PVTri is larger than that of MWCNT/PVTri. This difference can be explained in several ways. One explanation is that the chemically modified MWCNT was not homogeneously dispersed in the polymer matrix as well as the modified SWCNT. The second explanation is that the interaction between the modified MWCNTs and the polymer chains is less than that for the modified SWCNTs and polymer chains. An improved interaction between the nanotubes and the polymer matrix should lead to a stronger shift in the glass transition temperature [30].

Electric Conductivity

In electronic industries, polymeric materials are being widely used to produce numerous parts such as circuit boards, encapsulants for electronic device, monitor frames, keyboards, personal computer housings, and mice. Many of these applications take advantage of the good insulating property of polymer materials but some applications require polymer materials that are electrically conductive. However, most polymer materials possess a very high electrical resistance [31]. A common approach for the development of conductive properties in these polymers involves adding conductive fillers to a non-conductive polymer matrix. One of the commonly used conductive fillers is carbon nanotubes (CNTs) [32,33]. Pure PVTri is an excellent insulating material and has an electrical conductivity of $10^{-11}$ S/cm at 10 Hz and 20 °C [34]. To make PVTri electrically conductive, electrically conductive fillers must be added.

In this study, two disk-shaped samples that are 8 mm in diameter and 1.73 and 0.78 mm in thickness were prepared via cold-pressing. I-V measurements were performed by pressing the bottom and the top surface of the samples.
between two metal electrodes at room temperature using a picoamperometer (Keithley 6487). Figure 9 shows the current density as a function of the electrical field for the MWCNT/PVTri sample at room temperature.

The J-E curve exhibits nearly ohmic behavior. The conductivity of the MWCNT/PVTri composite is calculated to be 9.35 mS/m. Using an in situ polymerization technique, the attachment of PVTri to the MWCNTs significantly increases the electric conductivity of the synthesized nanocomposites. This growth results from the formation of a conducting network by the nanotubes in the polymer matrix\[^{35-37}\], as shown in Figure 10.

Numerous studies have shown that the electrical conductivity strongly depends on the polymer type, synthesis method, aspect ratio of the CNTs, disentanglement of the CNT agglomerates, uniform spatial distribution of individual CNTs and the degree of alignment\[^{38,39}\]. The electrical conductivity of either the SWCNT/PVTri or MWCNT/PVTri composites is primarily determined by the state of the fillers (i.e., dispersion and distribution) in the PVTri matrix. In comparison to MWCNT/PVTri, SWCNT/PVTri\[^{24}\] exhibited a much higher conductivity due to the stronger interaction of PVTri with the chemically modified SWCNTs compared to the chemically modified MWCNTs. In addition, because silane coupling agents are able couple the modified SWCNTs to PVTri, the strong interfacial adhesion between the SWCNTs and PVTri may stabilize the dispersion of the SWCNTs in the PVTri matrix preventing the CNTs from agglomerating. The improved dispersion means that the chemically modified SWCNTs are

![FIG. 8. DSC thermograms of a) PVTri and b) MWCNT/PVTri composite as a function of temperature.](image)

![FIG. 9. J-E curve for the MWCNT/PVTri sample.](image)

![FIG. 10. Explanation of the MWCNT/PVTri structure formed by nanotubes and polymer chains.](image)
able to form more connections than the modified MWCNTs, which leads to a higher electrical conductivity for the composites. In addition, several studies have reported that acid-modified MWCNTs exhibited less improvement in the electrical conductivity of MWCNT/polymer composites [40].

Dielectric Conductivity

The dielectric measurements of the MWCNT/PVTri and pure PVTri samples were performed in the frequency range of 20 Hz to 1 MHz at room temperature using a LCR Meter (A LCR (Inductance, Capacitance, and Resistance) meter (AGILENT 4284A). Two samples were cold-pressed into a disk-shape sample that was 8 mm in diameter and 0.88 and 0.99 mm in thickness. Figure 11 shows the dependence of the capacitance of the MWCNT/PVTri and pure PVTri samples on the frequency at room temperature. As expected, the capacitance of the structure formed as a metal-insulator-metal decreased as a function of the frequency. This behavior is due to the orientation with the electric field of the dipoles that exist or appear with an external electrical field in the insulator depending on the frequency [41].

The high conductivity of the MWCNTs does not directly increase the dielectric constant of the medium. However, MWCNT/PVTri behaves as a capacitor with high capacitance when it is placed between two metals. As shown on Figure 11, the capacitance value of the capacitor that consists of MWCNT/PVTri reaches a higher value compared to the capacitor that consists of PVTri. Because the capacitor consists of a system, there are several minicapacitors connected in parallel between them in the structure [42,43]. In the MWCNT/PVTri structure, the PVTri molecules that are connected to the MWCNTs generate an insulator medium between two MWCNTs and act as small capacitors (Figs. 12a and 12b). These capacitors, which are called

![FIG. 11. Capacitance as a function of frequency for MWCNT/PVTri and pure PVTri.](image1)

![FIG. 12. Schematization of MWCNTs and PVTri behavior in the composite: a) a typical capacitor figure, b) a representative minicapacitor figure, and c) a representative capacitor figure consisting of MWCNT/PVTri.](image2)

![FIG. 13. Dependence of the dielectric constants of a) pure PVTri and b) MWCNT/PVTri on the frequency.](image3)
minicapacitors, comprise the whole capacitance because they are connected in serial and parallel depending on their contact conditions in the blend (Fig. 12c). This result indicates that the amount of parallel connections is considerably higher than the amount of serial connections, because the capacitance of MWCNT/PVTri is higher than pure PVTri (Fig. 12c).

The overall capacitor, which has a very high capacitance, is similar to an insulator with the having same geometry and a high dielectric. The dielectric constant of this type of insulator is 1218041,491 at 1 kHz (Fig. 13). However, such an insulator with a high dielectric constant does not exist.

To obtain this high capacitance with PVTri, the geometry factor is $4.5 \times 10^{-12}$. However, for MWCNT/PVTri, the same capacitance is reached with a $0.15 \times 10^{-17}$ geometry factor. In other words, a higher capacitance at the same geometry can be reached with MWCNT/PVTri compared to the one with PVTri. Therefore, the capacitors, which were prepared with MWCNT/PVTri, exhibit a very high capacitance at a small geometry and have the potential for use in the electronics industry$^{[44,45]}$.

**CONCLUSIONS**

In this study, MWCNTs were effectively functionalized by a mixture of concentrated sulfuric and nitric acid (3:1 by volume) at room temperature. The carboxyl and hydroxyl functionalized MWCNTs were reacted with a silane-coupling agent, 3-(trimethoxy)-propylmethacrylate-silane (KH-570). The chemically modified MWCNT/Poly(1-vinyl-1,2,4-triazole) nanocomposites have been prepared by in situ solution polymerization. The synthesized MWCNT/PVTri composites were characterized using Fourier transform infrared spectroscopy, X-ray diffraction, scanning electron microscopy, and transmission electron microscopy. In addition, the results obtained from previous research examining SWCNT/PVTri composites have been compared with these results. Based on the results from this, we can make the following conclusions:

1. Silane modification of MWCNTs with 3-(trimethoxy)-propylmethacrylate-silane is an effective method for producing functional groups on the surface of MWCNTs and promoting better dispersion in the polymer matrix.

2. The presence of MWCNTs in the polymer matrix resulted in very little changes in the FTIR spectrum, which is most likely due to the low MWCNT composition and the weak vibrational signals of the MWCNTs. However, the FTIR spectrum of SWCNT/PVTri exhibited significant differences from the spectrum of modified SWCNTs and pure PVTri$^{[24]}$. This result indicates that the interfacial interactions between the functionalized SWCNTs and the PVTri matrix as well as the dispersion of the functionalized SWCNTs in the PVTri matrix are better than those of the MWCNTs.

3. The MWCNT/PVTri composites were dissolved in ethanol and DMF resulting in the formation of a homogenous solution, which was very stable even after prolonged standing for 2 weeks. Similar to the solubility results reported in a previous study$^{[24]}$, neither sedimentation nor aggregation of the nanotube bundles was observed.

4. A general increase in the thermal stability of MWCNT/PVTri was observed due to the addition of the MWCNTs. However, the increase in $T_g$ value of SWCNT/PVTri (in a previous study$^{[24]}$) is larger than the value of MWCNT/PVTri due to the modified SWCNTs being dispersed more homogeneously in the polymer matrix compared to the chemically modified MWCNTs. In addition, the interaction between the modified MWCNTs and the polymer chains is weaker than that between the modified SWCNTs and the polymer chains.

5. Using an in situ polymerization technique, the attachment of PVTri to MWCNT significantly increased the electric conductivity of the synthesized nanocomposites. In comparison to MWCNT/PVTri, SWCNT/PVTri$^{[24]}$ exhibited a much higher conductivity due to the stronger interaction of PVTri with the chemically modified SWCNTs compared to the chemically modified MWCNTs.

The high electrical conductivity and chemical stability may lead to the extensive application of carbon materials with precious metal catalyst supports in fuel cell membranes and bipolar plate fabrication. This work could provide a basis for future investigations into CNT-based bipolar plates for fuel cell applications.

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