

New strategy for chemically attachment of Amide group on Multi-walled Carbon Nanotubes surfaces: synthesis, characterization and study of DC electrical conductivity

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ABSTRACT

A new method of amidation of Carboxy Multi Walled Carbon Nanotubes (MWCNT-COOH) with diamine monomer such as ethylene diamine (EDA) and O-Phenylenediamine (OPDA) was applied by using a solution blending technique. The structure and properties of these composites have been investigated by FTIR, SEM, TEM, XRD, UV, DSC and TGA. The formation of Poly [MWCNT/ Amide] composites was confirmed and the DC electrical conductivity of poly-composites was in the range 4.5×10^{-6} - 5.3×10^{-6} S/cm due to the interaction between the nanotubes.

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1. Introduction

The applications of nanotechnologies can cover materials manufacturing, nano-electronics and computer technology, medicine, health, aeronautics, space exploration, environment devices, information storage, biotechnology and polymer technology¹⁻¹⁵.

The preparation of polymer nanocomposites filled with carbon nanotubes generally requires the nanotubes to be homogeneously dispersed and compatible with the polymer matrix¹⁶. The preparation of poly Amide-functionalized multi-walled carbon nanotube is highly relevant and useful for fabricating nanocomposites. Recently, Chen et al.,¹⁷ treated oxidized nanotubes with long-chain alkyl

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amines via acylation and, for the first time, made the functionalized material soluble in organic solvents. Qu et al.¹⁸ synthesized soluble nylon functionalized carbon nanotubes using a grafting-form strategy by attaching caprolactam molecules onto the nanotubes, followed by anionic ring-opening polymerization of these bound caprolactam species with the same monomers in bulk. Gao et al.¹⁹ reported a method for grafting Poly Amide 6, (PA6) chains to single-walled carbon nanotubes (SWNTs) through condensation reactions between the carboxylic groups of functionalized SWNTs and the terminal amine groups of PA6 by in situ polymerization. In addition, Yang et al.²⁰ prepared multi-walled carbon nanotubes (MWNTs) functionalized with PA6 by anionic ring-opening polymerization. However, this method is inefficient because the reaction time often lasts several hours, and the grafting ratio of PA6 is relatively low. In a different approach, Yan and Yang,²¹ have used Oxidized MWNTs, which were previously modified with isocyanate groups to prepare Poly Amide composites by in situ anionic ring-opening polymerization (AROP). In this study, a new method of amidation of MWCNT-COOH with diamine monomer such as ethylene diamine and *O*-Phenylenediamine will be applied by using the technique of solution blending. Furthermore, the structure and properties of these composites will also be investigated by FTIR, SEM, TEM, XRD, UV, DSC and TGA. The most important part of this work is the study of the DC electrical conductivity of the composite.

2. Results and Discussion

2.1. Characterization

Poly[MWCNT/Amide] composites were prepared by reacting MWCNT-COOH with (EDA and OPDA) in a refluxing solvent (DMF) to give poly-composites products. The method employed to prepare the Poly[MWCNT/Amide] composites was solution blending. The Chemical reaction of Poly[MWCNT/Amide] composites is shown in **Figure 1**. **Table 1** also summarizes the physical properties (melting point, color, percentage yield and solubility) of MWCNT-COOH and Poly[MWCNT/Amide] composites. Generally, these compounds showed good solubility mainly in DMF and DMSO; and they were either partially soluble or insoluble in other common organic solvents.

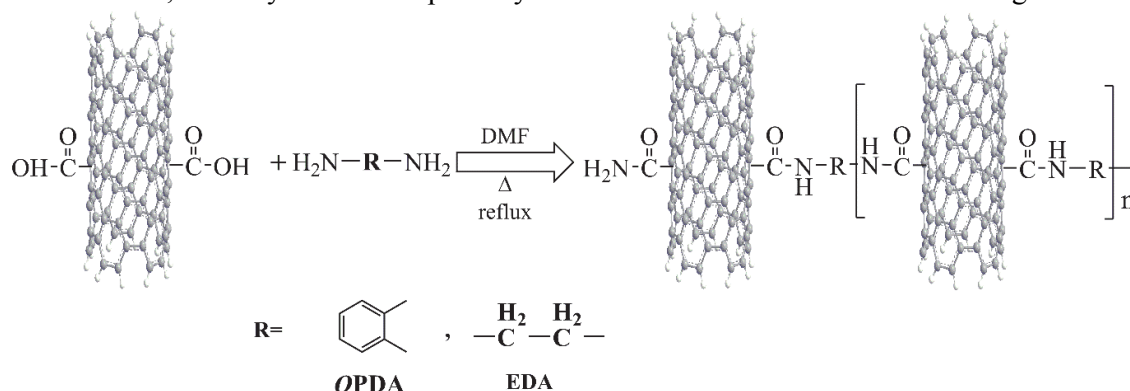


Fig. 1. Synthesis of Poly[MWCNT/Amide] composites

Table 1. Physical properties of MWCNT-COOH and of Poly[MWCNT/Amide] composites

No	Symbol	%Yield	Color	M.P.	Solubility		
					DMSO	DMF	EtOH
1	MWCNT-COOH	///	Black	> 350°C	++	+	-
2	Poly[MWCNT/OPDA]	87%	Black	> 350°C	++	+	-
3	Poly[MWCNT/EDA]	70%	Black	> 350°C	++	+	-

++;Soluble, +;Partially Soluble,-; Not Soluble.

2.2. Fourier Transform Infrared Spectroscopy (FTIR)

In **Fig. 2**, the IR spectra of MWCNT-COOH show a broad peak at 3434 cm⁻¹ that can be assigned to the O-H stretching of carboxyl groups (COOH). The peak at 1542 cm⁻¹ can also be associated with

the C=C stretching vibration of the MWCNT backbone, whereas the peak at 1637 cm^{-1} is related to the C=O stretching vibration of the carbonyl group acid,²².

The IR spectrum of the Amide-functionalized MWCNT: (Poly[MWCNT/EDA] and Poly[MWCNT/OPDA] in Fig. 2, shows the shift to the lower wave length corresponding to carbonyl of Amide (C=O) stretch from 1637 cm^{-1} to 1628 cm^{-1} . The presence of new bands at (1546 cm^{-1} , 1560 cm^{-1}) and (1117 , 1033 cm^{-1}) corresponds to N-H and C-N bond stretching, respectively; and broad diffuse peaks appear at (3431 , 3433 cm^{-1}) due to N-H stretching vibrations,²³.

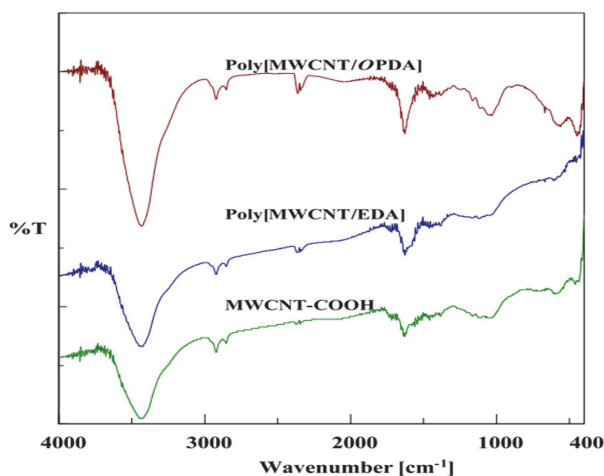


Fig. 2. FTIR spectra of MWCNT-COOH and Poly[MWCNT/Amide] composites

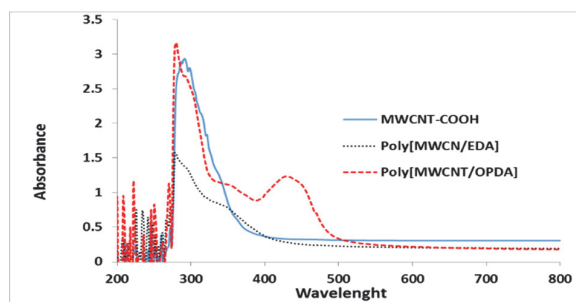


Fig. 3. UV-Vis spectra of MWCNT-COOH and Poly[MWCNT/Amide] composites

2.3. UV/Vis Spectroscopy

Fig. 3 shows the electronic spectra of MWCNT-COOH three bands π - π^* transition at λ_{max} 286, 290 and 298 nm, and the other two bands n - π^* transition at λ_{max} 320 and 338 nm. Meanwhile, the spectra of Amides show a lower shift of π - π^* transition λ_{max} at (276,292) and (280,293) nm for Poly[MWCNT/EDA] and Poly[MWCNT/OPDA], respectively. Also, for n - π^* transition, it shows a red shift and a new band λ_{max} at (346) and (352,432) nm for Poly[MWCNT/EDA] and Poly[MWCNT/OPDA] appears respectively, which confirms that the Amide functional group was formed.

2.4. Microscopy Characterization (TEM, SEM)

2.4.1. Transmission Electron Microscopy (TEM)

Transmission electron microscopy (TEM) is often used to observe the length and diameter of carbon nanotubes. Thus, **Fig. 4(a-f)** presents TEM microphotographs of the MWCNT-COOH and Poly[MWCNT/ester] composites at different magnifications. Also, **Fig. 4(a and b)** shows TEM images of MWCNT-COOH, which formed an entangled structure with an average diameter of 8-15 nm and their average length is approximately equal to $50\mu\text{m}$, which was provided by the supplier (Timesnano). In addition, a small spot shape was observed which might be ascribed to $-\text{COOH}$ group.

As shown in **Fig. 4(c-f)**, the MWCNT-COOH, after polymerizing them with OPDA and EDA, the Poly[MWCNT/Amide] composites display a relatively good dispersion and appear less entangled. The most twisted structures of Poly[MWCNT/OPDA] may be due to the Tensile angle on OPDA. The images also clearly show that the spot's shape for the COOH groups that disappeared in Poly[MWCNT/Amide] composites.

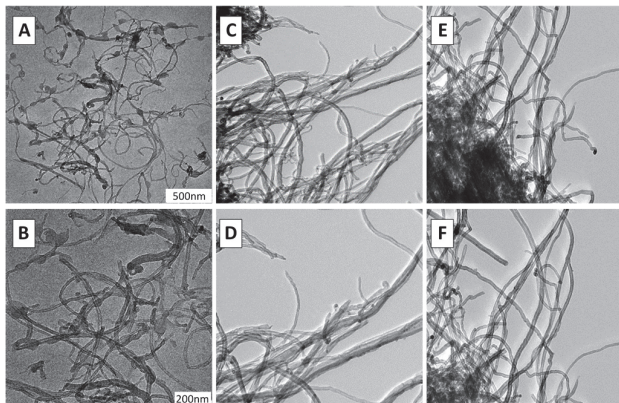


Fig. 4. TEM microphotograph of MWCNT-COOH and Poly[MWCNT/Amide] composites: MWCNT-COOH (a) $\times 50\,000$; (b) $\times 100\,000$, Poly[MWCNT/OPDA] (c) low magnification; (d) high magnification and Poly[MWCNT/EDA] (e) low magnification, (f) high magnification

2.4.2. Scanning Electron Microscopy (SEM)

Scanning electron microscopy was also used to confirm the possible morphological changes on functionalized MWCNT. **Fig. 5(a-l)** shows SEM microphotographs of the surface morphology and the dispersion of the MWCNT-COOH and Poly[MWCNT/Amide] composites at different magnifications. **Fig. 5(a-d)** also shows that the MWCNT-COOH forms large agglomeration, random and curled structure, and possesses high aspect ratio; this may be because of the hydrogen bonds between the nanotubes. Considering **Fig. 5(e-l)**, it displays Poly[MWCNT-OPDA] and Poly[MWCNT-EDA] composites; many walls were broken and appeared to be thicker compared to the MWCNT-COOH. In addition, it is noticed that masses have become smaller, which reduced the hydrogen bonding; the Amide bonds were also observed between MWCNT and (OPDA or EDA). The images clearly show that the surface morphology of Poly[MWCNT/Amide] composites is significantly different, compared to the MWCNT-COOH.

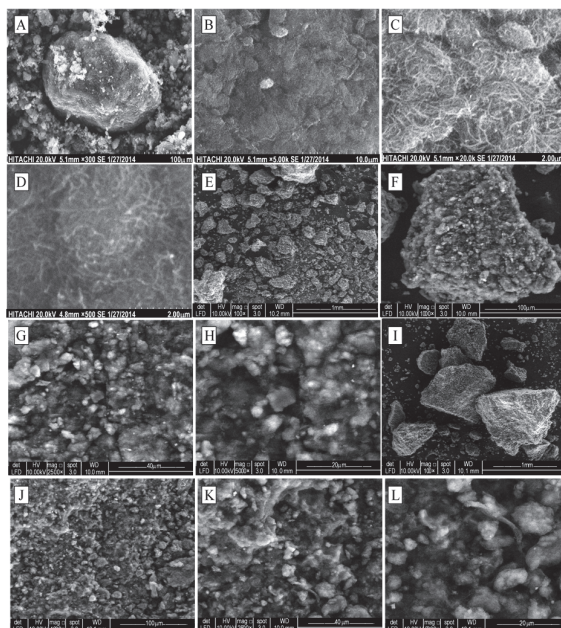


Fig. 5. SEM microphotograph of MWCNT-COOH and Poly[MWCNT/Amide] composites: (a) MWCNT-COOH ($\times 300$), (b) MWCNT-COOH ($\times 5000$), (c) MWCNT-COOH ($\times 20\,000$), (d) MWCNT-COOH ($\times 40\,000$) (e) Poly[MWCNT/OPDA] ($\times 100$) (f) Poly[MWCNT/OPDA] ($\times 1000$), (g) Poly[MWCNT/OPDA] ($\times 2500$), (h) Poly[MWCNT/OPDA] ($\times 5000$), (i) Poly[MWCNT/EDA] ($\times 100$), (j) Poly[MWCNT/EDA] ($\times 1000$), (k) Poly[MWCNT/EDA] ($\times 2500$), (l) Poly[MWCNT/EDA] ($\times 5000$)

2.5. X-ray Diffraction

X-ray diffractions of the MWCNT-COOH and Poly[MWCNT/Amide] composites are shown in **Fig. 6**, where the sharp diffraction patterns at $2\theta=26.6^\circ$ and 45.45° correspond to the graphite structure of MWCNT-COOH. After functionalizing MWCNT-COOH with EDA and OPDA the crystallinity increases, and the intensity of the Poly[MWCNT/OPDA] becomes sharper than that of the Poly[MWCNT/EDA].

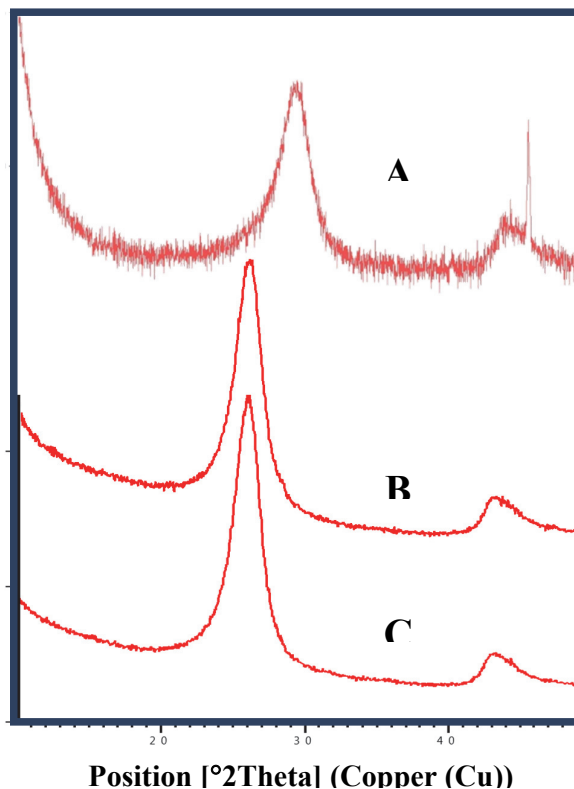


Fig. 6. X-ray diffraction of MWCNT-COOH and Poly[MWCNT/Amide] composites: (a) MWCNT-COOH, (b) Poly[MWCNT/EDA] and (c) Poly[MWCNT/OPDA]

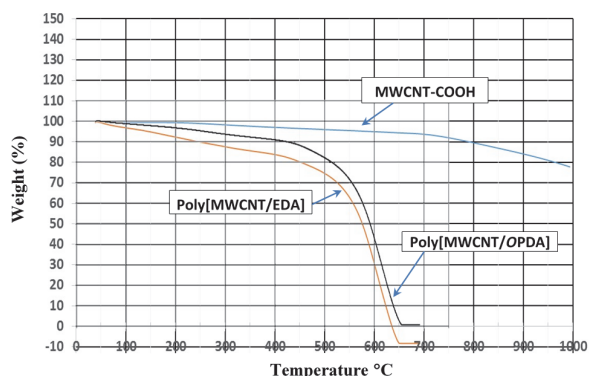
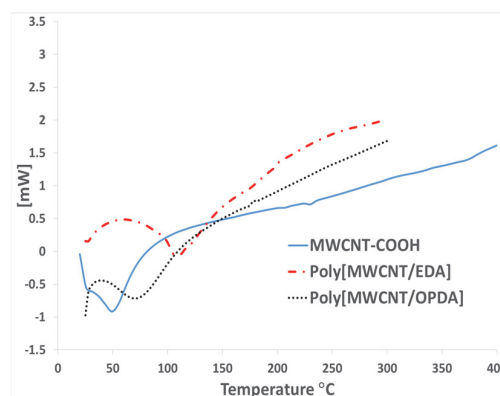
2.6. Thermal properties (Thermogravimetric analysis (TGA) and Differential Scanning Calorimetry [DSC])

The TGA and DSC of MWCNT-COOH and Poly [MWCNT/Amide] composites are presented in **Fig. 7 and Fig. 8**, respectively, and summarized in **Table 2**. The curves show that the MWCNT-COOH is more stable than their poly-composites; the order of thermal stability is MWCNT-COOH > Poly[MWCNT/EDA] > Poly[MWCNT/OPDA], and the highest thermal stability of MWCNT-COOH is related to the hydrogen bonds between carboxylic groups.

The degradation process of Poly[MWNT-EDA] exhibits four steps: the first step is at $\sim 60^\circ\text{C}$, assigned probably to moisture, the second step is at $\sim 200^\circ\text{C}$, because the amidic groups which were broken into pieces, and the third and fourth steps are at ~ 350 and 605°C , respectively, which are normally attributed to the degradation of graphite structures. The degradation process of Poly[MWNT-OPDA] can be shown in three steps: i) ($39\text{-}400^\circ\text{C}$) which is assigned to the breaking of amidic groups, ii) and iii) at ~ 460 and 613°C , respectively, which are normally attributed to the degradation of graphite structures²³. An increase in the mass loss of the Poly[MWCNT/Amide] composites was also observed, that is probably due to the deformed hydrogen bond of carboxyl group in MWCNT-COOH, which confirmed the amidic groups was obtained and disappeared the $-\text{COOH}$ group. The thermal stability of Poly[MWCNT/OPDA] which is more than Poly[MWCNT/EDA] refers to the conjugation and size of the aromatic ring of OPDA, shown in **Table 4** below.

Table 2. TGA and DSC results of the MWCNT-COOH and Poly[MWCNT/Amide] composites

Compound	Step	TGA			DSC		Res. %	
		Wt. Loss %	T _i /°C	T _f /°C	T _{DTG}	T _{DSC}		
MWCNT-COOH	1 st	0.78	33.46	149.99	52.54	31, 55	endo	99.22
	2 nd	5.30	148.89	673.29	298.22	-	-	93.92
	3 rd	8.19	673.23	880.34	753.55	*	*	85.73
	4 th	8.30	880.17	1015.74	948.50	*	*	77.43
Poly[MWCNT/EDA]	1 st	2.97	41	100	60.5	73	endo	97.5
	2 nd	12.97	100	400	200	''''	-	84.97
	3 rd	9.55	400	502	350	''''	-	74.98
	4 th	66.37	502	648	605	''''	-	8.61
Poly[MWCNT/OPDA]	1 st	9	39	400	-	107	endo	91
	2 nd	9	401	500	460	''''	-	82
	3 rd	501	660	613	''''	-	1	

**Fig. 7.** TGA curves of MWCNT-COOH and Poly[MWCNT/Amide] composites**Fig. 8.** DSC curves of MWCNT-COOH and Poly[MWCNT/Amide] composites

DC Electrical conductivity

It is generally agreed that the mechanism of conductivity in the π -conjugated polymeric materials is based on the motion of charge defects within the conjugated framework. The charge carriers, either positive p-type or negative n-type, are the products of oxidizing or reducing the material, respectively. The following overview describes these processes in the context of p-type carriers although the concepts are equally applicable to n-type carriers,^{24,25}.

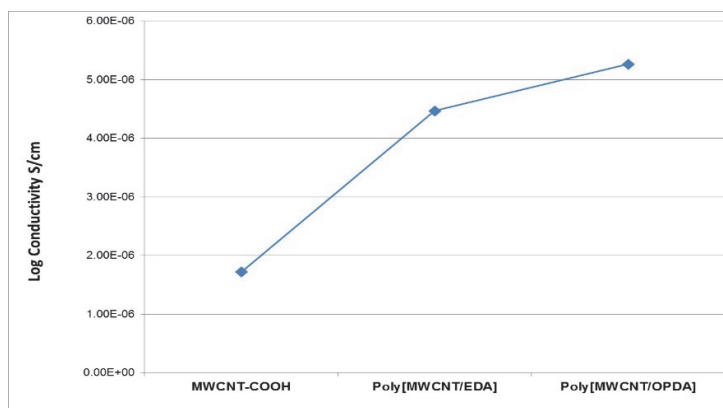
**Fig. 9.** DC electrical conductivity of MWCNT-COOH and Poly[MWCNT/Amide] composites at room temperature

Fig. 9 shows the DC electrical conductivity of MWCNT-COOH and Poly[MWCNT/Amide] composites at room temperature. The order of DC electrical conductivity is Poly[MWCNT /OPDA] > Poly[MWCNT /EDA] > MWCNT-COOH (5.2671E-06, 4.4670E-06 and 1.7181E-06 S/cm), respectively. There is a slight increase in the DC electrical conductivity of Poly[MWCNT/OPDA] and Poly[MWCNT/EDA] due to the linking of MWCNT with diamines. However, the DC electrical conductivity value of poly[MWCNT/OPDA] that is higher than Poly[MWCNT/EDA] may be due to the conjugation of the aromatic ring.

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4. Experimental

4.1. Materials

Carboxy Multi Walled Carbon Nanotubes (MWCNT-COOH) were purchased from Timesnano (Chengdu Organic Chemicals Co. Ltd., Chinese Academy of Sciences) China. The diameter and length of MWCNT ranged between 8-15 nm and 50 μm respectively. Purity was over >95%, and the carboxyl group coverage over the nanotube surface was (2.56 %wt.). Ethylenediamine (EDA), O-Phenylenediamine (OPDA) were obtained from Sigma Aldrich, whereas dimethylsulfoxide (DMSO) was purchased from Scharlau. N,N-Dimethylformamide (DMF 99%), Tetrahydrofuran (THF 99.9%) and Ethanol (96%) were purchased from Fluka and used as received without any further treatment in this study.

4.2. Instrumentation

The FTIR spectra were recorded using the KBr disc technique on a JASCO 410 FTIR Spectrophotometer (at Sana'a University, Sana'a, Yemen). The melting points were measured with an electrothermal melting point apparatus (at Sana'a University, Sana'a, Yemen). The thermal analyses (TGA and DSC) were carried out on a Mettler Toledo TGA/SDTA851e analyzer, and Mettler Toledo DSC823e analyzer, respectively, at 23 to 1000 $^{\circ}\text{C}$, under 20 ml of nitrogen per minute and a heating rate of 10 $^{\circ}\text{C}$ per minute (at UPM AND UM Universities, Kuala Lumpur, Malaysia). UV-vis absorption spectra were measured using a Specord 200, Analytik Jana, Germany in DMF ($\sim 10^{-4}$ mol/dm³) (at Sana'a University, Sana'a, Yemen). The X-Ray diffraction was carried out on a BrukerAxs Da Advance, Germany (at UPM AND UM Universities, Kuala Lumpur, Malaysia). The electrical conductivity measurements were taken on a Keithley Picoammeter/Voltage Source Model 6487 and using a double probe conductivity cell that is locally fabricated (at Sana'a University, Sana'a, Yemen). The Scanning Electron Microscope (SEM) was carried out on a (SEM HITACHI S-3400N) (at UPM AND UM Universities, Kuala Lumpur, Malaysia). The Transmission Electron Microscope (TEM) was carried out on a Phillips CM-12, USA, and the samples were prepared by Leica ultracut UTC ultramicrotome (JEOL, Japan) with an accelerating voltage of 100 kV.

4.3. Preparation of Poly[MWCNT/Amide] composites

4.3.1. Preparation of poly-composites of MWCNT-COOH with EDA

0.3 g sample of MWCNT-COOH was dispersed in 15 ml DMF, and 1 ml of EDA was added to the MWCNT dispersion in DMF. The mixture was then stirred for 24 hours at 90 °C under reflux. After cooling to room temperature, the mixture was vacuum-filtered through a 0.22 µm membrane and was thoroughly washed several times with DMF. The filtered solid was then dried in a vacuum oven at 90 °C for 24 hours,²⁶.

4.3.2. Preparation of poly-composites of MWCNT-COOH with OPDA

0.3 g sample of MWCNT-COOH was dispersed in 15 ml DMF and 0.2 g of OPDA was dissolved in 10 ml DMF and added to the MWCNT dispersion in DMF. The mixture was then stirred for 24 hours at 90 °C under reflux. After cooling to room temperature, the mixture was vacuum-filtered through a 0.22 µm membrane and was thoroughly washed several times with DMF. The filtered solid was then dried in a vacuum oven at 90 °C for 24 hours,^{27,28}.

5. Conclusions

In summary, Poly[MWCNT/Amide] composites were prepared by the reaction of MWCNT-COOH with (EDA and OPDA) by solution blending techniques. The obtained poly-composites were characterized by FT-IR, UV-Vis, XRD, TEM, SEM, TGA, DSC, and DC electrical conductivity. X-ray diffraction confirms an increase in the crystallinity of the Poly[MWCNT/Amide] composites. The high thermal stability of MWCNT-COOH was linked to the hydrogen bond between carboxylic groups. The increase in the thermal stability of the Poly[MWCNT/Amide] composites is due to a hydrogen bond in poly-composites and the H-bonded of the un-reacted carboxyl group. There is a slight increase in the DC electrical conductivity of Poly [MWCNT/OPDA] and Poly[MWCNT/EDA] due to the linking of MWCNT with diamines. Nevertheless, the DC electrical conductivity value of Poly [MWCNT/OPDA] that is higher than Poly[MWCNT/EDA] may be assigned to the conjugation of the aromatic ring.

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