Effect of Different Multi-Walled Carbon Nanotubes MWCNTs on Mechanical and Physical Properties of Epoxy Nanocomposites

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Abstract. Different types of long and short multiwalled carbon nanotubes (MWCNTs) were used as structural support in epoxy resin with a percentage of weight (0.1,0.2,0.5,1.0,1.5,2.0,2.5,3.0,3.5,4.0,4.5,&5) wt. percent. Utilization of direct mixing processes to prepare Nano-Composites (Epoxy / MWCNTs). The ultrasonic mixing method has been used to disperse the nanotubes into the epoxy resin system. SEM was used to demonstrate the dispersion in the epoxy polymer matrix of different MWCNT concentrations. The findings show that electrical conductivity properties boost effect, thermal conductivity coefficient, and A.C with percentage enhancement of less than 2 percent, and then raising with further raise in the content of MWCNTs.

Keywords. Multi-Walled Carbon-Nanotubes (MWCNTs), Epoxy, Nano-composites.

1. Introduction

Owing to the comparatively poor mechanical properties of polymer resin, its use in the components has been inhibited. The physical characteristics of curing epoxy resins depend on the composition, the degree of curing, and the time and temperature of the curing method. Due to its unusual mechanical, thermal, and electrical properties, nanotubes (CNT) as the filler have attracted substantial interest in the polymer matrix. CNTs have a strong propensity to agglomerate due to their high aspect ratio and immense surface area, leading to the polymer matrix, and inhomogeneous dispersion. To allow the development of high-performance CNT/polymer composites, various techniques for the dispersion of CNTs[1-6] have been extensively studied and compared. Two major challenges must be addressed: (i) the homogeneous dispersion of CNTs in the matrix and (ii) the strong interfacial interaction to allow the load transfer from the matrix to the CNTs to be efficient [7,8]. Some early research findings showed that carbon nanotube-epoxy nanocomposites were barely stronger or weaker than net epoxy. In fact, for nanotube-reinforced polymers, another key problem to be solved is the dispersion of CNTs in the matrix method. There is an enormous surface area of nano-scaled particles (1000m²/g and more). Several magnitudes are greater than that of the surface of conventional fillers. This surface area serves as a stress transfer interface but it is also accountable for the CNTs. Strong propensity to form agglomerations, [9].

Higher carbon nanotubes loading in composites results in higher levels of carbon nanotubes loading of carbon nanotubes anf higher thermal conductivity, but its thermal conductivity value is well below its theoretical expectations, based on the simple mixture law, due to its broad interface dispersion and
contact resistance. Because of decreased interface number and contact resistance, the alignment of CNTs in the composite also increases thermal conductivity along the alignment direction.[10,11]. To achieve greater electrical resistance, we need to focus on: (1) by using longer CNTs, the number of inter-tube contacts. in the carbon nanotube networks and their composites is reduced (2) enhancing the alignment, and (3) maintaining a solid electrical/thermal interface with CNT-CNT and CNT-epoxy. To observe the effects of CNT, the researchers used long-MWCNT and its networks. For this purpose duration was expanded for these long-MWCNTTs mechanically. Sheets were used to achieve a greater degree of alignment to decrease the number of contacts in the composite coating. It was placed between tubes. Long-MWCNT composites have already shown great performance gains compared with previous CNT composites. However, due to greater interfacial bonding between the nanotubes and the matrix of the epoxy, the mechanical properties of functionalized CNT composite were significantly improved [12,13]. The majority of CNT composite thermal conductivity measurements have been performed at room temperature and temperature-dependency is restricted to the MWCNT or SWCNT network, [14-17]. The aim of the effects of CNT duration and MWCNT were further explored in this analysis concentration on epoxy polymer tensile and thermal conductivity properties.

2. Experimental work

2.1. Products and preparing samples

To prepare the samples, raw materials were used; epoxy was used as a matrix (Nitofill, EPLV with Ni to fill Fosroc Company EPLV hardener). The mixing ratio of resin and hardener is 3:1 and 40 minutes at (35) °C gelling time, 1.0 Poise at (35) °C mixed viscosity, and MWCNTs provided by (35) °C and MWCNTs manufactured by NANOSHEL LLC USA are the nanoparticle fillers. To prepare samples that are cost-effective with simple production methods, the hand layup technique was used in different weight percentages of MWCNTS (1,0.2,0.5,1.0,5,2,0,2,5,3,0,3,5,4,0,4,5,&5wt. percent). By applying MWCNT to the epoxy resin, the amount of epoxy was blended with epoxy using direct mixing. The mixture was demanded for 30 minutes in an ultrasonic bath) and then agitated (2 hours) to disperse the MWCNTs in the polymer matrix. After the hardener has been added to the mixture and stirred for (4 minutes), the vacuum machine (10-2 bar) was used to extract a bubble before molding. They cast the mixture into a mould. The samples remained for 72 hours before the samples were extracted and left in the vacuum chamber for (14) days before any test to achieve better curing conditions, Table 1.

| Materials        | Length (µm) | Diameter (nm) | Surface area (m²/g) | Purity % |
|------------------|-------------|---------------|---------------------|---------|
| Epoxy (EPLV)     | ------------|---------------|---------------------|---------|
| MWCNT LONG       | ~50         | 5-15          | 233                 | >97     |
| MWCNT SHORT      | 3-8         | 20-30         | 90-350              | 99.8    |

2.2. Characterization of MWCNTs and their Epoxy Composites Testing.

2.2.1. FTIR spectra. FTIR spectra aggregated by a spectrum one FTIR spectrometer. CNTs were mixed with KBr powder and strangled as pellets for data collection. The spectrum was collected against KBr background. The nanocomposites were factored as films and the spectra were collected beside the background. The FTIR spectra were for pure epoxy and epoxy / MWCNTs nanocomposites.

2.2.2. Scanning electron microscope (SEM). A scanning electron microscope (VEGA\Easy prope) was used to gauge the morphology of MWCNTs and EP/MWCNT as revealed in results. After observing the specimens with the tensile test, the Scanning Electron Microscopy (SEM) revealed well-dispersed nanotubes in the matrix.

2.2.3. Tensile test. Samples were arranged according to the ASTM D638 standard. The machine used for testing the tensile strength was microcomputer-controlled universal testing of electronic Machine
(model WDW-50) maximum load (50KN). The test was directed at speed (10 mm/min) and temperature (25°C). The tensile load was performed until the sample failed and the stress-strain curve was obtained. Each sample was tested 2 times and the average results have been reported. Equations (1.4) and (1.3) were used to determine the modulus of elasticity, tensile strength respectively, and toughness from the stress-strain curves to ensure that affiliations are as complete as possible and include the country.

\[
\sigma = Y\varepsilon 
\]  
\[
\varepsilon = \frac{\Delta L}{L_o} = \frac{L - L_o}{L_o} 
\]  
\[
Y = \sigma/\varepsilon 
\]  
\[
\sigma = F/A 
\]

Where; F is the force, A is the area of a plane, the elongation ∆L = L - Lo and L, Lo the instantaneous and original length[10].

2.2.4. Impact test. The researchers used standard impact fracture energy calculation MODEL(XJJ series), ASTM standard(6110), Laryee, China Origin[18], and the Charpy impact value slows to calculate the dynamic power. The Charpy effect's impact value is expressed by the following equation.

\[
E = M \times (\cos \beta - \cos \alpha) - (\cos \alpha - \cos \alpha) \times \frac{\alpha + \beta}{\alpha + \alpha} 
\]  
\[
G_s = \frac{e}{A} 
\]

E: Impact fracture energy (kJ), Gs: Impact strength (kJ/m²), M: Impact force torque (N.m), β: Pendulum raising angle after samples breaking, α: Actual measerment value of pendulum rising angle, ā: Avoiding angle, and A: Area of the sample (m²)

2.2.5. Thermal conductivity. The coefficient of the thermal conductivity was calculated from the data measured by using the lee's disk method. The emitted energy was exposed to the area of the surface and thermal conductivity calculated from the sample by using equations (1.7) and (1.8) respectively.

\[
e = \frac{VI}{aATA+aS(TA+TB)+aB TB+aCTC} 
\]  
\[
K = \left[\frac{ed}{2\pi r^2 (TB-TA)} \right] \left[\frac{as(TA+TB)}{2} + 2 aA TA \right] 
\]

K: Thermal conductivity Coefficient (W/m.K), e: Energy be emitted from the exposed area of the surface ( joules), aA ,aB,aC , aS : The specimen and heater exposed surface area of A,B,C,S respectively (m²),TA ,TB and TC: are the temperature of the disks A, B and C (K) [19].

3. Results and discussion

FTIR spectra can provide additional indications for the difference in mechanical properties and thermal properties of different samples concerning their molecular structures. Figures (1, 2) show the neat epoxy resin and MWCNT/epoxy nano-composite FTIR spectrum. The FTIR spectrum of commercial epoxy resin is presented in Figure 1. The epoxy group is denoted by bands at 2966.52 cm⁻¹ and 1246.02 cm⁻¹ (830 cm⁻¹). Fig.2 shows the elimination of the peak (2966.52 cm⁻¹). The peak band area at 914.26 cm⁻¹ increased to 937.40 cm⁻¹ and decreased in the peak of the -OH groups by the FTIR of (epoxy/MWCNTs) as in Fig.2. The least extreme peak may be observed at 3506.59 cm⁻¹ to 3417.86 cm⁻¹. The interaction between epoxy resin and MWCNTs is a result of the good dispersion of MWCNT which leads to a shift in the peaks of FTIR epoxy groups. To evaluate the morphology and dispersion of MWCNTs in the epoxy matrix, SEM was used. The scanning electron
micrographs of the samples observed under the microscope after the tensile tests are illustrated in figures (3,4 and 5). The most common failure observed was in MWCNT-reinforced polymer nanocomposites. Figure (3) indicates that the tidy epoxy samples have a clean and smooth base. The integration into the matrix of pristine MWCNTs raises the roughness, as shown in Figures (4) and (5). As the dispersion level increased (1.5 percent wt NWCNTs) nano-composites Figure (4), better dispersion of the MWCNTs and greater adhesion between the MWCNTs and the epoxy matrix make the smoother epoxy fracture surface becomes rougher. Better MWCNT dispersion and greater adhesion between the MWCNTs and the epoxy matrix are ideal for mechanical and thermal conductivity. Increasing MWCNTs concentration above (1.5%wt MWCNTs) leads to increasing the agglomeration size Figure (5), which leads to reducing the reinforcing effect of MWCNTs and decreasing the mechanical and thermal properties of Epoxy/MWCNTs nanocomposites. The stress-strain curves of epoxy and MWCNT/epoxy composites are analyzed by tensile testing at room temperature. Figures (6) and (7) provide a comparison between ultimate tensile strength and Young's modulus results (MPa) of epoxy resin with differing quantities of MWCNT reinforcing agent. After the tensile stress reached the highest stage, all specimens broke instantly. Also, no yield point was observed for any of the samples, since there is a brittle property in the epoxy resin used in our research. Basically, the mechanical strength depends on the interface bond between the filler and the epoxy. Mechanical properties are enhanced in the case of MWCNTs/epoxy nano-composites, but not as much as anticipated. The following two explanations can clarify this. The weak bond between the CNTs and the surrounding matrix is the first explanation. Interfacial bonding between the MWCNTs and the polymer resin has been stated to be weak and the transfer of load from the polymer to the MWCNTs is not sufficient enough to crack the MWCNTs under tensile load. The second explanation is related to the structural issue of the MWCNTs' outer shells, since there would be weak van der Waals forces acting between the MWCNTs' individual graphene shells, slipping between the shells. The epoxy composite with LMWCNTs of 2 percent wt content showed the highest ultimate tensile strength and Young's modulus, (45.20 MPa), (1140 MPa) respectively, and for 2 wt% SMWCNTs (40.81 MPa), (1100 MPa), compared with the neat epoxy (35.26 MPa), (840 MPa). Above (2%) MWCNTs both of ultimate tensile strength and Young's modulus decreases, this is because both the dispersion of MWCNTs and the volume of MWCNTs in the epoxy resin are highly dependent on the mechanical strength of the epoxy composites. The decrease in strength with a higher MWCNT content disrupts the epoxy permeation on the surface of the MWCNT. There is decreased interfacial adhesion between the MWCNTs and the matrix, and hence the effective interfacial surface for the epoxy.[15]. These phenomena induce the power of the reinforcing gent's embed. The findings of the current study indicate that the agglomeration size increases by increasing the content of multi walled carbon nanotubes. It is obvious that because of agglomeration, the reinforcing impact of the MWCNTs decreases. The charpy impact strength values for the net epoxy and Epoxy/MWCNTs nano-composites are shown in Fig. (8). Based on the values, the impact strength of the neat epoxy increased with an increase in the content of LMWCNTs to (1.5 wt. percent). Based on values, the impact strength of the neat epoxy increased to (1.5 wt. percent) [Repetition?]. The strong dispersions of the nano-tubes in the matrix and the good reaction between the epoxy and the grafted nano-tubes have been due to this. The long MWCNTs given high values of impact strength compare with Short MWCNTs. All decreases in the flexural module, flexural strength, hardness, and impact strength value of MWCNT/epoxy nanocomposites are due to the formation of nano-tubes agglomerates for both types within the polymer matrix, which decreased the CNTs' reinforcing effects by acting as resin flaws. Figure (9) shows the thermal conductivity (W/m. K ) as a function of CNT concentration for short- and long-MWCNT/epoxy composites. The findings show that both forms of MWCNTs enhance epoxy polymer thermal conductivity. The thermal conductivity of epoxy is determined by the option of the filler, which is the percentage of the load of the filler. The thermal energy transfer process depends on the structural nature of the material and the style of the transformation process differs depending on the material, and lattice waves and free electrons are two ways to convert thermal energy. Better thermal conductivity values lead to (2 wt% LMWCNTs ) up to(1.3 W/m.K ), and for SMWCNTs at the same percentages, it reaches (0.785 W/m.K ) compared with the values of neat epoxy polymer
Above 2%wt, a decrease is observed in thermal conductivity that leads to agglomerate and poor dispersion when increasing the percentage weight of MWCNTs as in Figure (5) for SEM.

Figure 1. Fourier transforms infrared spectra for the resin of epoxy.

Figure 2. Fourier transforms infrared spectra of epoxy /MWCNTs Nano-composites.

Figure 3. SEM for epoxy resin.

Figure 4. (1.5 wt%)MWCNTs dispersion in epoxy resin.
Figure 5. (3.5 wt%) MWCNTs dispersion in epoxy resin.

Figure 6. Ultimate tensile strength vs MWCNT wt% of Epoxy MWCNTs composites.

Figure 7. Young’s modulus vs MWCNT wt% of Epoxy/ MWCNTs composites.
4. Conclusion
In this study, two MWCNTs types (long and short/MWCNTs) were used to prepare epoxy/ nanocomposites. In terms of epoxy resin mechanical and thermal conductivity properties, the reinforcement of the epoxy resin by MWCNTs was examined. The nano-composites with (1.5 wt%) MWCNTs exhibits good dispersion. The highest ultimate tensile strength and Young's modulus reaches (45.20 MPa), (1140 MPa) for LMWCNTs, and (40.81 MPa), (1080 MPa) for SMWCNTs compared with the the value of pure epoxy of (35.26 MPa), (840 MPa). Maximum thermal conductivity for both types is also observed in the nano-composite at (2wt% MWCNTs), which is about 4 times for LMWCNTs and 2 times for SMWCNTs higher than that of the pure epoxy. Above (2 wt % MWCNTs) a decrease is observed in tensile properties and thermal conductivity values due to agglomeration of MWCNT. The findings show that LMWCNTs are better than SMWCNTs for improving mechanical and thermal properties.

Figure 8. Charpy impact vs MWCNT wt% of Epoxy MWCNTs composite.

Figure 9. Thermal conductivity vs MWCNT wt% of Epoxy / MWCNTs composites.
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