

Research Article

Shailesh I. Kundalwal* and Ankit Rathi

Improved mechanical and viscoelastic properties of CNT-composites fabricated using an innovative ultrasonic dual mixing technique

<https://doi.org/10.1515/jmbm-2020-0008>

Received Jun 13, 2020; accepted Aug 07, 2020

Abstract: Carbon nanotube (CNT) acts as the most promising nanofiller due to its high aspect ratio and exceptional nanoscale-level properties. However, the dispersibility of CNTs in the conventional polymer matrices is a very critical issue in developing the high-strength and light-weight polymer-based nanocomposites. In this study, an attempt was made to develop cluster-free and uniform dispersion of multiwalled carbon nanotubes (MWCNTs) in the epoxy matrix using an innovative ultrasonic dual mixing technique. The effect of dispersion of MWCNTs on the mechanical and viscoelastic properties of MWCNT-epoxy nanocomposites was comprehensively studied. Our results reveal that the tensile strength and toughness of epoxy nanocomposites with 0.50 wt.% of MWCNTs improved by 21% and 46%, respectively, as compared to neat epoxy. The nanocomposite samples with the same CNT loading show maximum enhancements of 22% and 26% in the lap shear strength and storage modulus, respectively. The tensile fracture surface examination of MWCNT-epoxy nanocomposites using field emission scanning electron microscopy indicated the cluster-free and uniform dispersion of MWCNTs in the epoxy matrix.

Keywords: carbon nanotubes; nanocomposites; mechanical and viscoelastic properties; characterization

1 Introduction

In recent years, the high-strength and light-weight polymer nanocomposites gained much attention in multifarious applications because of their multifunctional behaviour, versatility and low cost. The polymer nanocomposites possess high thermo-mechanical properties that fulfil the potential applications of automotive and aerospace, coating and paint, and electrical and electronics industries [1–3]. The epoxy resin is the most versatile polymer matrix material that offers excellent structural and adhesive properties, high thermal stability and good corrosive resistance [4–7]. Despite these advantages, the high cross-linking density and poor resistance to crack propagation of neat epoxy possess low strength as well as fracture toughness, which limits its structural applications [8].

Some recent studies in nanotechnology demonstrate that the addition of carbon-based nanofillers such as carbon nanotubes (CNTs) and graphene improve the mechanical, thermal as well as structural properties of polymer composites [9–13]. CNTs offer incomparable high-strength and large-surface area with unique advantage of 1D physical structure that make them ideal nanofillers for conventional polymer composites [14–16]. The incorporation of CNTs in the polymer matrix drastically improves the load bearing capacity of resulting nanocomposite and thus, its overall mechanical properties increase. However, high specific surface area of CNTs create strong van der Waals attractive forces between them that results in the agglomeration of CNTs [17–19]. For instance, Song and Youn [20] investigated the effect of CNTs on the thermo-mechanical and electrical properties of epoxy nanocomposites. They found that the dispersion state of CNTs alter the physical properties of nanocomposites. Poorly dispersed CNTs improved the storage modulus, loss modulus and viscosity due to their rheological point of view. Whereas the good dispersion of CNTs improved the mechanical properties such as strength and toughness of polymer nanocomposites. Allaoui *et al.* [21] studied the mechanical and electri-

*Corresponding Author: Shailesh I. Kundalwal: Applied and Theoretical Mechanics (ATOM) Laboratory, Discipline of Mechanical Engineering, Indian Institute of Technology Indore, Simrol, Indore 453 552, India; Email: kundalwal@iiti.ac.in

Ankit Rathi: Applied and Theoretical Mechanics (ATOM) Laboratory, Discipline of Mechanical Engineering, Indian Institute of Technology Indore, Simrol, Indore 453 552, India

cal properties of multiwalled carbon nanotube (MWCNT)-based polymer nanocomposites. Results revealed that the reinforcement of 1 and 4 wt.% of MWCNTs in the epoxy matrix drastically improved the mechanical properties of polymer nanocomposites. Kumar *et al.* [22] studied the characterization of CNT-epoxy adhesive joint on mild steel substrate. They found that the lap shear strength of substrates depends on the surface morphology of substrates, bond line thickness and wettability of adhesive mixture. Montazeri and Chitsazzadeh [23] studied the mechanical properties of MWCNT/epoxy nanocomposites by varying the sonication process parameters. They found that the increase in sonication time and power output increases Young's modulus up to a certain limit, and further increase in sonication time decreases Young's modulus of polymer nanocomposites. Gojny *et al.* [24] investigated the effect of different types of CNTs (*i.e.*, single-, double- and multi-walled CNTs) on the mechanical properties of epoxy nanocomposites, produced by calendaring process. Results revealed that the nanocomposites having 0.5 wt.% loading of double-walled CNTs provide the most significant improvement in the mechanical properties compared to all other loading contents. However, the above studies also reveal that the uniform dispersion and strong bonding of CNTs with the surrounding polymer matrix are important and critical issues, which restrict the applications of CNTs for obtaining high-performance polymer nanocomposites. To overcome these issues, various techniques such as solution mixing [25], high energy ball milling [26], In situ polymerization [27], CNT functionalization [28], and ultrasonic mixing process [29] were used previously. Among them, the combination of ultrasonic mixing process with magnetic stirrer known as ultrasonic dual mixing (UDM) technique is expected to be the most promising method to obtain the uniform dispersion and strong interactions of nanofillers with the surrounding polymer matrix. During this process, the probe inserted in nanocomposite mixture solution vibrates with ultrasonic waves in KHz range. The particles closed to the probe get blasted and release massive amount of energy. The bubbles are created, and they collapse with the surrounding solution by disrupting the particles interactions inside the solution. Subsequently, the magnetic stirrer provides a rotating magnetic field to the solution through stirring bar. Therefore, this technique is expected to provide the homogenous distribution of nanoparticles by breaking their clusters inside the polymer matrix [30, 31].

Existing studies proved that the functionalization of surfaces of CNTs improves the mechanical as well as physical properties of polymer nanocomposites [28]. However, the complex surface modification process of CNTs

deconstructs their atomic structures that results in decreasing the bulk properties of polymer nanocomposites. Therefore, the surface modification of CNTs is not always favourable [32]. From the literature review, we can conclude that the mechanical and adhesive properties of CNT-based nanocomposites primarily influenced by the dispersion of CNTs and their interaction with the matrix as well as the processing technique used for synthesis of polymer nanocomposites. In the present study, MWCNTs were reinforced with epoxy matrix using an innovative UDM technique. An effort was made to obtain the uniform stable dispersion and good interactions of MWCNTs with the surrounding epoxy matrix using an optimised UDM process parameters. The MWCNT-epoxy nanocomposite samples were characterized by performing tensile test, lap shear strength test and dynamic mechanical analysis (DMA). The structural morphology of MWCNT nanofillers was studied using TEM, and finally, the dispersion of CNTs and fracture mechanisms of nanocomposite samples were studied by FESEM.

2 Experimental methods

2.1 Materials used

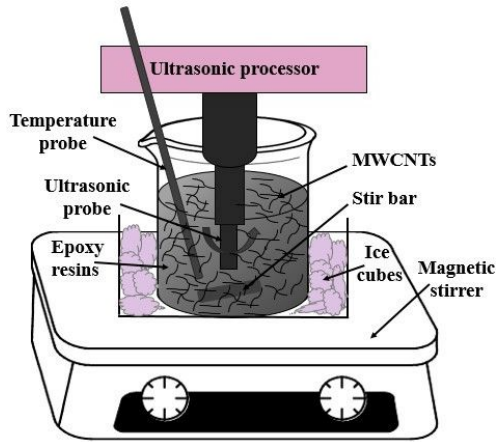
The epoxy resin (EPOFINE - 1564) of viscosity range 1000-14000 mPas (density ~ 1.10-1.20 g/cc) at room temperature and aliphatic hardener (FINEHARD - 3486) of viscosity < 50 mPas (density ~ 0.94 - 0.95 g/cc) at room temperature purchased from Fine Finish Organics Pvt. Ltd, India. Polymer matrix and curing agent with a weight ratio of 100:33 were used for the fabrication of polymer nanocomposites. The commercially available MWCNTs of average diameter 20 nm (length ~15 μ m and purity > 99%) was obtained from a certified source of M/s Nano Shell LLC, USA.

2.2 Nanocomposites synthesis

The MWCNTs (0.25 wt.%) were introduced into a semi-viscous epoxy resins with 10% acetone in a vessel and thoroughly mixed by a glass rod stirrer. In order to achieve cluster-free uniform dispersion of MWCNTs, a titanium alloy tip of 20 mm diameter of an ultrasonic processor was introduced in MWCNT-epoxy adhesive mixture with power output 750 W that generated ultrasonic waves of amplitude 70% at 20 kHz frequency of vibration. The ultrasonic processor was applied on 80 ml volume of adhesive mixture for 30 min at an interval of 5s ON and 5s OFF. The

Table 1: Various weight percentages of constituents of MWCNT-epoxy nanocomposites prepared using UDM technique

Epoxy resins	MWCNTs	Aliphatic hardener
45 gm	0.0 wt. %	15 gm
45 gm	0.25 wt. %	15 gm
45 gm	0.50 wt. %	15 gm
45 gm	1.0 wt. %	15 gm

**Figure 1:** Schematic of UDM technique to prepare the MWCNT-epoxy nanocomposite mixture

produced high intensity ultrasonic waves created alternative high and low pressure inside the mixture that formed small vacuum bubbles. Simultaneously, magnetic stirrer was used with speed of 250 rpm that provided agitation to the MWCNTs and mixed them with adhesive mixture. Due to the combined action of both ultrasonic waves and magnetic stirrer, the molecular interaction between the MWCNTs disrupts and breaks their agglomerates that leads to the uniform mixing of MWCNTs inside the adhesive mixture. This process also generates heat in the adhesive mixture and its temperature rises. To prevent the adhesive mixture from degrading due to the rise in temperature, some ice cubes were placed around the vessel as shown in Figure 1. The prepared adhesive mixture was placed in hot air oven for 24 h at 70°C to remove the solvent. Then, the aliphatic hardener of 1/3rd weight of epoxy resins thoroughly mixed inside the MWCNT-epoxy mixture and subsequently, the vacuum degassing process was used to remove the air bubbles.

Finally, the bubble-free MWCNT-epoxy adhesive was poured into the silicon rubber molds and curing was performed at 50°C for 12 h in a convection oven to prepare the specimens. Other compositions of MWCNT-epoxy nanocomposite samples using aforesaid process are listed Table 1.

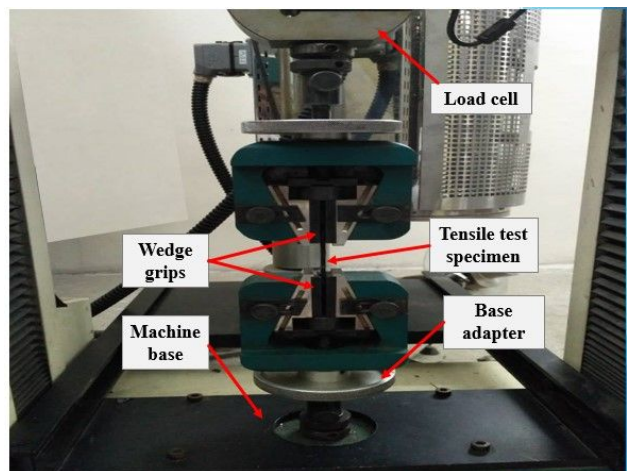
2.3 Preparation of faying surfaces of aluminium substrates

To attain the maximum lap shear strength, the surface of aluminium substrates were prepared by mechanical treatment. In this process, the surface of aluminium substrates were abraded with emery papers of grade 220 to remove the contaminants such as grease, dirt, etc. The abrasion process of aluminium substrate was done for half an hour to reduce the void formation by removing the dirt and other contaminants from its surface and then cleaned by acetone before utilizing it.

2.4 Characterisation of nanocomposite samples

Tensile test

The dumbbelled-shaped tensile test specimens of neat epoxy and MWCNT-epoxy nanocomposites were tested by following ASTM D638 (V) standard using a Hounsfield (H25KS) Universal Testing Machine (UTM) with a cross-head speed of 1 mm/min as shown in Figure 2. The obtained tensile stress-strain curves of nanocomposites spec-

**Figure 2:** Hounsfield universal testing machine (H25KS) used for tensile testing

imen provide the data of tensile strength and toughness. TEM and FESEM analysis was used to understand the structural morphology of MWCNTs and their dispersion quality in the epoxy matrix, respectively. The fracture surface of tensile test specimens was studied to understand the fracture mechanism using an Ultraplus55, Zeiss microscope at an acceleration voltage of 15 kV on a gold-coated surface.

Dynamic mechanical analysis

Dynamic mechanical analysis (DMA) test was performed using DMA 8000 Perkin-Elmer instrument to study the viscoelastic behaviour and thermal transitions of neat epoxy and MWCNT-epoxy nanocomposite samples. A sinusoidal deformation was applied to the specimen of dimensions 9.0 mm long, 6 mm wide and 2 mm thick by varying the temperature range from 30 to 180°C in a single frequency strain mode. The heating rate of 2°C/min was used with a frequency of 1 Hz as per ASTM D4065 standard.

Adhesive joints preparation and lap shear strength test

The lap joints specimens were prepared by applying MWCNT-epoxy adhesive on different faying surfaces of aluminium substrates. The adhesive mixture was gently placed on the faying surface at an appropriate place to form a lap joint by assuring the dimensions, as shown in Figure 3. During curing stage, a uniform pressure of 5 MPa was applied on the mating surfaces of aluminium substrates by the roller at a speed of 5 mm/min to obtain the uniform bond line thickness with an overlapped area of $12.7 \times 25.4 \text{ mm}^2$. The obtained adhesive joint specimens were placed inside hot air oven for 8 h at 50°C temperature for curing process. The lap shear test was conducted by placing each end of a specimen in UTM grips and pulled apart at a speed of 1 mm/min. The force versus extension curves of adhesive joints were recorded and finally, con-

verted into stress-strain curves to examine the lap shear strength test.

3 Results and discussions

3.1 Tensile test

The mechanical properties of neat epoxy and MWCNT-epoxy nanocomposite samples were evaluated using obtained stress-strain curves from the UTM tests. The tensile test specimens of nanocomposites with different weight percentages of MWCNTs (0.25 wt.%, 0.50 wt.% and 1.0 wt.%) were prepared by using UDM technique with an amplitude of 70%. The loading effect of MWCNTs on the tensile properties of nanocomposites was studied from the stress-strain curves which provide the tensile strength (Figure 4). The area under stress-strain curve represents the toughness of specimens (Figure 5). Figures 4 and 5 indicate that the tensile strength and toughness of nanocomposite specimens with 0.50 wt.% of MWCNTs increased by 21% and 46%, respectively, compared to neat epoxy. The increment in strength and toughness are primarily due to the effective uniform dispersion of MWCNTs and their good interfacial bonding with the surrounding epoxy matrix that enhanced the load bearing capacity of resulting nanocomposite. The tensile strengths of nanocomposites with 0.25 wt.% and 0.50 wt.% MWCNTs increased by 12% and 21%, respectively, and the toughness values of corresponding of nanocomposite samples increased by 20% and 46%, as compared to neat epoxy.

It was noticed that the tensile strength and toughness of epoxy nanocomposites increased with an increase in the amount of MWCNTs up to 0.50 wt.% and then the increasing trend declines. This was mainly due to the increase in van der Waals force of attraction between MWCNTs and thus, their clusters form, which eventually create defects and helps in crack nucleation inside the bulk nanocomposite. Several existing studies revealed that the varying

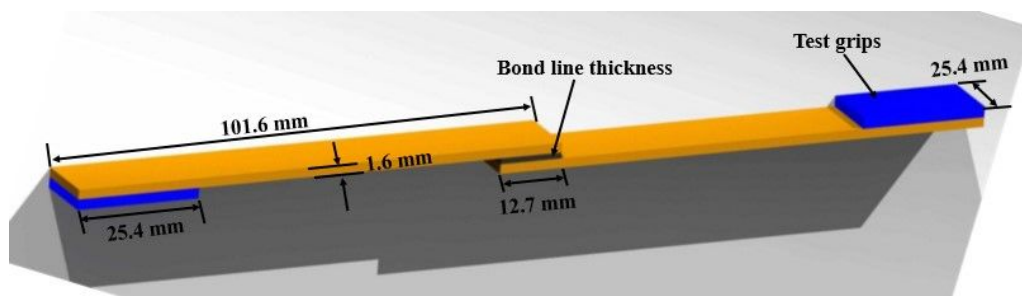


Figure 3: Specimen configuration of single-lap shear adhesive joints of aluminium substrates

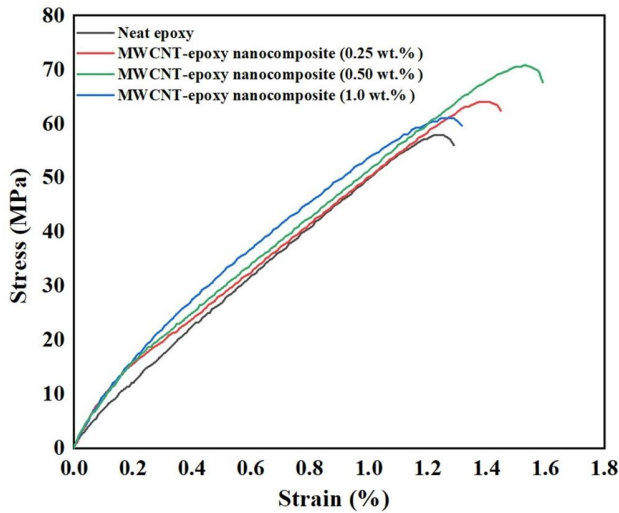


Figure 4: Stress-strain curves of neat epoxy and epoxy nanocomposites with different weight percentages of MWCNTs

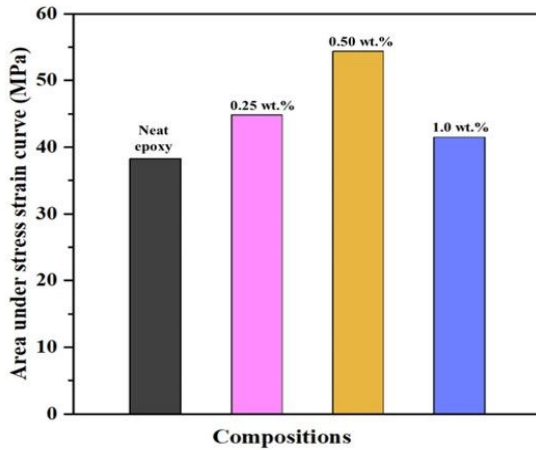


Figure 5: Toughness of neat epoxy as well as epoxy nanocomposites with different weight percentages of MWCNTs

percentage of MWCNTs influences the mechanical performances of their nanocomposites [33–35]. However, there are other factors responsible for enhancing the mechanical performance of CNT-based polymer nanocomposites such as the structure of CNTs and their aspect ratio, CNT alignment as well as dispersion and interaction of CNTs with the surrounding polymer matrix [36]. It can be observed from Figures 4 and 5 that the introduction of 1.0 wt.% of MWCNTs in the epoxy matrix significantly reduced the tensile strength and toughness of nanocomposites, respectively. This is attributed to the formation of clusters of MWCNTs in the epoxy matrix that result in the creation of voids inside the bulk nanocomposite which eventually reduces its strength and toughness.

3.2 Tensile fracture morphology

Figure 6 (a) presents the TEM image of a MWCNT having high aspect ratio. The diameter of MWCNT was clearly examined through TEM image which is nearly 30 to 40 nm whereas its length was difficult to examine due to the non-observability of end points.

The FESEM analysis of samples of neat epoxy and epoxy nanocomposites carried out that helped us to study the extent of dispersion of MWCNTs as well as fracture behaviour of their nanocomposites. Figure 6 (b) is the FESEM image of tensile fracture surface of neat epoxy that shows the clear and smooth fracture pattern, which indicates its brittle fracture due to the poor impact resistance. In neat epoxy, the crack propagated freely with the lower resistance that results in its poor toughness. However, the addition of 0.25 wt.% of MWCNTs in the epoxy matrix, the surface becomes rough and shows protruding like structure. It can be observed from Figures 6d and 6e that the fracture surface of epoxy nanocomposites with 0.50 wt.% loading of MWCNTs demonstrates rough and plateau structures, having some MWCNT fragments on its surface, which shows the crack bridging and pull-out of MWCNTs from the epoxy matrix. These are the important micromechanical toughening mechanisms observed in FESEM analysis that are responsible for increasing the mechanical as well as toughness properties of polymer nanocomposites. The FESEM images also confirm that the uniform dispersion of MWCNTs in the epoxy matrix can be obtained by UDM technique. The simultaneous action of ultrasonication process with magnetic stirrer generates enough energy that breaks the micron-size clusters of MWCNTs and assist in their uniform dispersion in the epoxy matrix. Further, increase in the loading of MWCNTs (>0.50 wt.%) in the epoxy matrix leads to the formation of their clusters as shown in Figure 6(f). The formation of clusters of MWCNTs occurs primarily due to the CNT-CNT interactions due to the van der Waals force of attractions between them which restrict their dispersibility in the epoxy matrix.

3.3 Lap shear strength analysis

The lap shear strength of neat epoxy and nanocomposite adhesives (with different weight percentage of MWCNTs) joints can be studied by stress-strain curves, as shown in Figure 7. The single lap shear adhesive joints were formed by applying neat epoxy as well as nanocomposite adhesives on the mechanically treated surfaces of aluminium substrates with a constant rolling load of 5.0 N to produce an optimum bond line thickness of neat epoxy/adhesive.

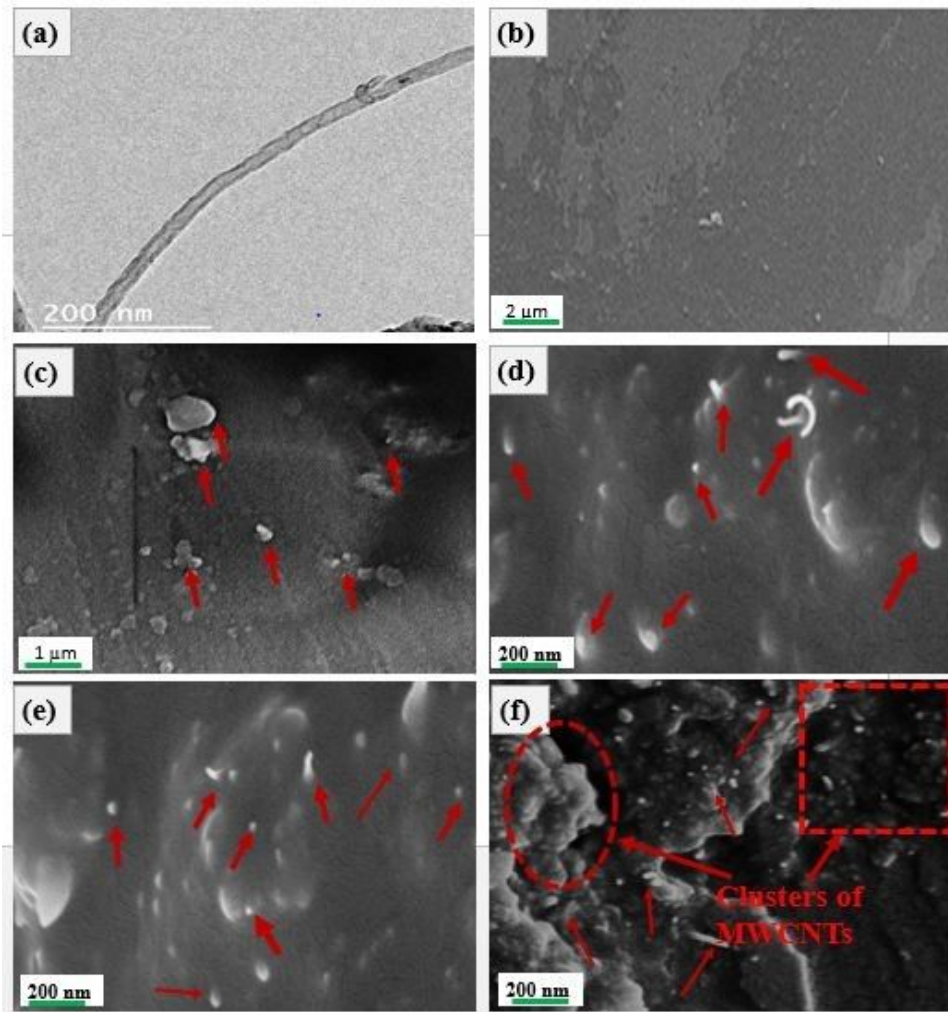


Figure 6: (a) TEM image of a MWCNT; and FESEM images of tensile fracture surface of (b) neat epoxy, (c) epoxy nanocomposite with 0.25 wt.% of MWCNTs, (d & e) epoxy nanocomposite with 0.5 wt.% of MWCNTs (f) epoxy nanocomposite with 1.0 wt.% of MWCNTs

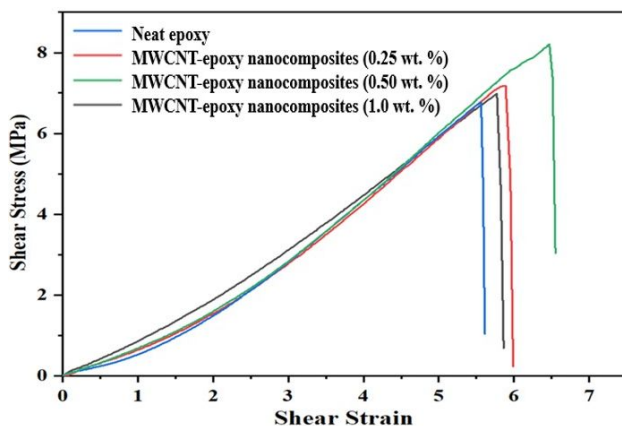


Figure 7: Lap shear strength of joints using neat epoxy and nanocomposites with different weight percentages of MWCNTs

The faying surface of aluminium substrate was mechanically prepared to enlarge the bonding area and enhance the mechanical interlocking to obtain the strong bond. The average results of four specimens having equal weight content of adhesives were considered for analysing and obtaining the reliable results of lap shear tests. Figure 7 shows that the addition of wt.% of MWCNTs up to 0.5 in the epoxy matrix significantly improves the lap shear strength and then further addition of MWCNTs leads to the reduction in shear strength. The lap shear joint of nanocomposite adhesive with 0.5 wt.% of MWCNTs shows the maximum improvement in shear strength up to 22% with respect to the neat epoxy adhesive joint. The high strengthening effect of nanocomposite joint is attributed to the cluster-free dispersion of MWCNTs in the epoxy matrix that results in the effective load transfer from the epoxy to MWCNTs. The high surface areas of MWCNTs improves

the degree of crosslinking with the surrounding epoxy matrix and produce strong interfacial bonding interaction between them.

It can be observed from Figure 7 that the addition of 0.25 wt.% and 0.50 wt.% of MWCNTs in the epoxy matrix increased the lap shear strengths by 7% and 22%, respectively, compared to neat epoxy adhesive joint. Figure 7 also demonstrate that the ductility (strain %) of adhesive joints increased with increase in MWCNTs up to 0.50 wt.% and then it decreased. Therefore, it can be concluded that the shear strength of adhesive joints primarily depends on the dispersion of nanofillers, faying surface preparation and bond line thickness of adhesive joints [34, 38].

3.4 Dynamic mechanical analysis

The viscoelastic property measurements of neat epoxy and epoxy nanocomposites in the temperature range of 30 to 180°C with heating rate of 2°C/min was investigated using DMA. It is a simple method to obtain the storage modulus (G'), loss modulus (G'') and tan delta ($\tan \delta$) of neat polymer and its nanocomposites by measuring their stiffness and damping. Figure 8 shows the storage moduli versus temperature curves of neat epoxy and nanocomposites with different weight percentages of MWCNTs. In the glassy region, the value of storage modulus of neat epoxy is low. It can be noticed from Figure 8 that the increment in weight percentages of MWCNTs in the epoxy matrix enhances the storage moduli of resulting nanocomposites. Some existing studies also suggest that the increment in the storage modulus is primarily due to the high covalent bonding between MWCNTs and epoxy matrix [39]. The addition of 0.50 wt.% of MWCNTs in the epoxy matrix shows highest storage modulus of 2371 MPa, which is primarily attributed to the cluster-free and uniform dispersion of MWCNTs that leads to the higher degree of cross-linking with 3D network of epoxy matrix. The better adhesive interaction and high degree of crosslinking result in decreasing the molecular chain mobility and enhancing the storage modulus of epoxy nanocomposites. The epoxy nanocomposite samples with 0.25 wt.% and 0.50 wt.% of MWCNTs showed the enhancement in the storage moduli as 13% and 26%, respectively, as compared to neat epoxy. The storage modulus of nanocomposite with 1.0 wt.% of MWCNTs is slightly lower than that of nanocomposite with 0.50 wt.% MWCNTs. This is again attributed to the formation of agglomerations of MWCNTs due to the van der Waals force of attraction between them in the epoxy. The cluster formation of MWCNTs in the epoxy matrix enhances the molecular chain mobility that results in the reduction in storage modulus. It can

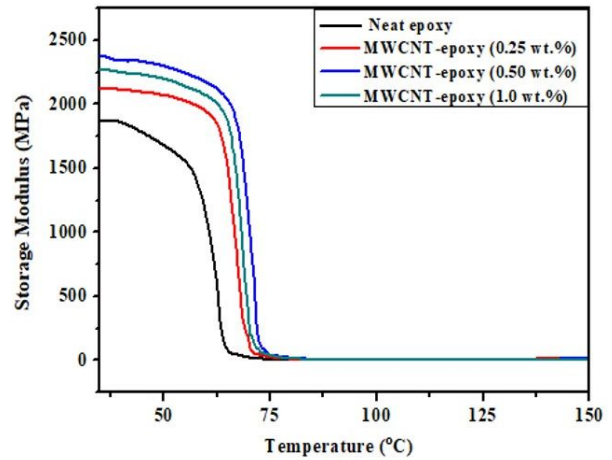


Figure 8: Storage modulus (MPa) versus temperature (°C) of neat epoxy and epoxy nanocomposites with different weight percentages of MWCNTs

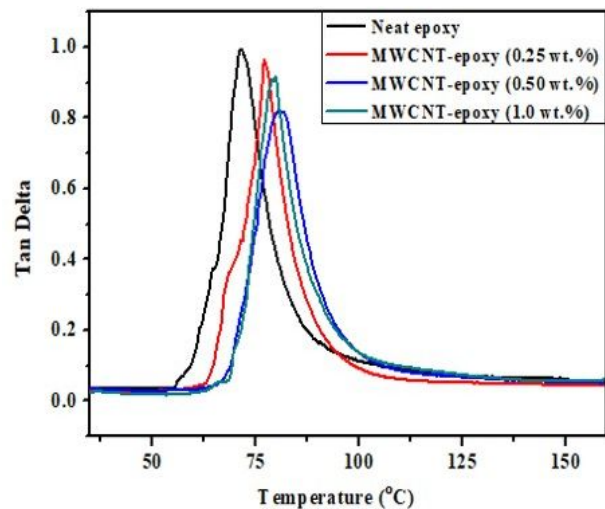


Figure 9: $\tan \delta$ versus temperature (°C) of neat epoxy and epoxy nanocomposites with different weight percentages of MWCNTs

be noticed that the storage moduli of neat epoxy as well as nanocomposite with different weight percentage of MWCNTs decreased with increase in temperature when the temperature reaches to glass transition temperature (T_g). This steady decrease in storage modulus near glass transition temperature attributed to the molecular chain mobility.

The loss factor (known as $\tan \delta$) represents the damping behaviour of materials and can be calculated by dividing loss modulus to storage modulus, G''/G' . The larger area under $\tan \delta$ peak indicates the higher energy dissipation. The uniform dispersion of MWCNTs in the epoxy matrix leads to the decrease in area under $\tan \delta$ peak compared to the neat epoxy, as shown in Figure 9. This is attributed to the interfacial bonding of MWCNTs with the

Table 2: Glass transition temperature (T_g), storage modulus (MPa) and loss factor ($\tan \delta$) of neat epoxy and epoxy nanocomposite with different weight percentage of MWCNTs

MWCNTs content (wt. %)	Glass transition temperature (T_g)	Storage modulus at glassy region (at 35°C)	Loss factor ($\tan \delta$)
0.0	71.42°C	1878 MPa	0.9955
0.25	77.01°C	2130 MPa	0.9588
0.50	80.54°C	2371 MPa	0.8165
1.0	79.15°C	2268 MPa	0.9129

surrounding epoxy matrix and hence, the molecular chain mobility at MWCNT-matrix interfaces decreases, which results in the decrease in damping peak [40]. The values of glass transition temperature (T_g) and $\tan \delta$ of neat epoxy as well as nanocomposites with different wt.% of MWCNTs are summarized in Table 2. The increase of 0.25 wt. % MWCNTs to 0.50 wt. % in the epoxy matrix slightly increases the glass transition temperature (T_g) from 77.01°C to 80.54°C and decrease the corresponding value of $\tan \delta$ from 0.9588 to 0.8165.

The damping in MWCNT-epoxy nanocomposites is mainly occurred due to the interfacial slippage between the MWCNTs and the surrounding epoxy matrix. When there is strong interfacial bonding between MWCNTs and epoxy matrix, then the interfacial slippage between them is very less. The increase in MWCNT content in the epoxy leads to the increase in interfacial slippage due to agglomeration of MWCNTs. Thus, the factors affecting the damping characteristics of polymer nanocomposites are filler percentage, critical shear stress, structural characteristics and crosslinking of nanofillers with the polymer matrix. The high aspect ratio and surface area of CNTs also improve the damping properties of polymer nanocomposites [41]. The uniform dispersion and good interaction of CNTs with the epoxy matrix is the key aspect of high storage modulus and low loss factor.

4 Conclusions

In summary, an innovative ultrasonic dual mixing (UDM) technique was used for the synthesis of MWCNT-epoxy nanocomposites to achieve uniform dispersion and good interfacial interaction of MWCNTs with the epoxy matrix. The simultaneous action of ultrasonic processor with magnetic stirrer produces enough energy which efficiently breaks the agglomerates of MWCNTs and significantly improved their dispersion quality in the epoxy matrix. The results reveal that the mechanical properties such as tensile strength, lap shear strength and toughness as well as

viscoelastic properties such as storage modulus of MWCNT-epoxy nanocomposites significantly enhanced with mere 0.50 wt.% loading of MWCNTs due to their effective uniform dispersion and good interaction with the epoxy matrix. However, further increase in loading contents of MWCNTs reduces the mechanical and viscoelastic properties due to the formation of agglomerates of CNTs that increase the stress concentration and create defects inside the bulk epoxy nanocomposites. The TEM image of MWCNT shows high aspect ratio, whereas the FESEM examination of tensile fracture surface of MWCNT-epoxy nanocomposites indicates the successful cluster-free and uniform dispersion of MWCNTs in the epoxy matrix.

References

- [1] Godara A, Mezzo L, Luizi F, Warriar A, Lomov S V., van Vuure AW, *et al.* Influence of carbon nanotube reinforcement on the processing and the mechanical behaviour of carbon fiber/epoxy composites. *Carbon*. 2009; 47(12):2914–23.
- [2] Yang SY, Lin WN, Huang YL, Tien HW, Wang JY, Ma CCM, *et al.* Synergetic effects of graphene platelets and carbon nanotubes on the mechanical and thermal properties of epoxy composites. *Carbon*. 2011;49(3):793–803.
- [3] Choyal V, Kundalwal SI. Interfacial characteristics of hybrid nanocomposite under thermomechanical loading. *J Mech Behav Mater*. 2017;26(3–4):95–103.
- [4] Sumdani MG, Islam MR, Yahaya ANA. The effects of anionic surfactant on the mechanical, thermal, structure and morphological properties of epoxy–MWCNT composites. *Polym Bull*. 2019;76(11):5919–38.
- [5] Torabi AR, Rahimi AS, Ayatollahi MR. Tensile fracture analysis of a ductile polymeric material weakened by U-notches. *Polym Test*. 2017;64:117–26.
- [6] Alnefaie KA. Strength and modulus of carbon nanotubes under a tensile load. *J Mech Behav Mater*. 2014;23(1–2):15–9.
- [7] Mansourian-Tabaei M, Jafari SH, Khonakdar HA. Lap shear strength and thermal stability of diglycidyl ether of bisphenol a/epoxy novolac adhesives with nanoreinforcing fillers. *J Appl Polym Sci*. 2014;131(6).
- [8] Jojibabu P, Zhang YX, Prusty BG. A review of research advances in epoxy-based nanocomposites as adhesive materials. *Int J Adhes Adhes*. 2020;96:102454.

- [9] Kim JA, Seong DG, Kang TJ, Youn JR. Effects of surface modification on rheological and mechanical properties of CNT / epoxy composites. 2006;44:1898–905.
- [10] Kundalwal SI, Ray MC. Micromechanical analysis of fuzzy fiber reinforced composites. *Int J Mech Mater Des.* 2011;7(2):149–66.
- [11] Kundalwal SI, Ray MC. Effect of carbon nanotube waviness on the effective thermoelastic properties of a novel continuous fuzzy fiber reinforced composite. *Compos Part B Eng.* 2014;57:199–209.
- [12] Reddy JN, Unnikrishnan VU, Unnikrishnan GU. Recent advances in the analysis of nanotube-reinforced polymeric biomaterials. *J Mech Behav Mater.* 2014;22(5–6):137–48.
- [13] Shingare SIKKB, Rathi A. Effect of flexoelectricity on the electromechanical response of graphene nanocomposite beam. *Int J Mech Mater Des.* Springer Netherlands; 2019;15(3):447–70.
- [14] Alian AR, Kundalwal SI, Meguid SA. Multiscale modeling of carbon nanotube epoxy composites. *Polymer (Guildf).* Elsevier Ltd; 2015;70:149–60.
- [15] Kong KTS, Mariatti M, Rashid AA, Busfield JJC. Effect of processing methods and functional groups on the properties of multi-walled carbon nanotube filled poly(dimethyl siloxane) composites. *Polym Bull.* 2012;69(8):937–53.
- [16] Alian AR, Kundalwal SI, Meguid SA. Interfacial and mechanical properties of epoxy nanocomposites using different multiscale modeling schemes. *Compos Struct.* 2015;131:545–55.
- [17] Kumar A, Ghosh PK, Yadav KL, Kumar K. Thermo-mechanical and anti-corrosive properties of MWCNT/epoxy nanocomposite fabricated by innovative dispersion technique. *Compos Part B Eng.* Elsevier Ltd; 2017;113:291–9.
- [18] Mallakpour S, Khadem E. Revised Carbon nanotube – metal oxide nanocomposites: Fabrication, properties and applications. *Chemical Engineering Journal.* 2016;302:344–67.
- [19] Peigney A, Laurent C, Flahaut E, Bacsa RR, Rousset A. Specific surface area of carbon nanotubes and bundles of carbon nanotubes. 2001;39:507–14.
- [20] Song YS, Youn JR. Influence of dispersion states of carbon nanotubes on physical properties of epoxy nanocomposites. *Carbon N Y.* 2005;45(7):1378–85.
- [21] Allaoui A, Bai S, Cheng HM, Bai JB. Mechanical and electrical properties of a MWNT / epoxy composite. *Compos Sci Technol.* 2002;62:1993–8.
- [22] Kumar A, Kumar K, Ghosh PK, Rathi A, Yadav KL, Raman. MWCNTs toward superior strength of epoxy adhesive joint on mild steel adherent. *Compos Part B Eng.* 2018;143:207–16.
- [23] Montazeri A, Chitsazzadeh M. Effect of sonication parameters on the mechanical properties of multi-walled carbon nanotube/epoxy composites. *Mater Des.* 2014;56:500–8.
- [24] Gojny FH, Wichmann MHG, Fiedler B, Schulte K. Influence of different carbon nanotubes on the mechanical properties of epoxy matrix composites - A comparative study. *Compos Sci Technol.* 2005;65(15–16 SPEC. ISS.):2300–13.
- [25] Andrews R, Jacques D, Minot M, Rantell T. Fabrication of Carbon Multiwall Nanotube / Polymer Composites by Shear Mixing. 2002;395–403.
- [26] Guadagno L, Naddeo C, Vittoria V, Sorrentino A, Vertuccio L, Raimondo M, *et al.* Cure behavior and physical properties of epoxy resin-filled with multiwalled Carbon nanotubes. In: *Journal of Nanoscience and Nanotechnology.* 2010;10(4):2686–93.
- [27] Liu P, Su Z. Preparation and characterization of PMMA/ZnO nanocomposites via inSitu polymerization method. *J Macromol Sci - Phys.* 2006;45(1):131–8.
- [28] Zhu J, Kim J, Peng H, Margrave JL, Khabashesku VN, Barrera E V. Improving the Dispersion and Integration of Single-Walled Carbon Nanotubes in Epoxy Composites through Functionalization. 2003;3(8):1107–13.
- [29] Gkikas G, Barkoula NM, Paipetis AS. Effect of dispersion conditions on the thermo-mechanical and toughness properties of multi walled carbon nanotubes-reinforced epoxy. *Compos Part B Eng.* 2012;43(6):2697–705.
- [30] Kumar A, Kumar K, Ghosh PK, Yadav KL. Ultrasonics - Sonochemistry MWCNT / TiO₂ hybrid nano filler toward high-performance epoxy composite. 2018;41:37–46.
- [31] Rathi A, Kundalwal SI. Mechanical and fracture behavior of MWCNT/ZrO₂/epoxy nanocomposite systems: Experimental and numerical study. *Polym Compos.* 2020;(January):1–17.
- [32] Zhang ZQ, Liu B, Chen YL, Jiang H, Hwang KC, Huang Y. Mechanical properties of functionalized carbon nanotubes. *Nanotech.* 2008;19(39):395702.
- [33] Theodore M, Hosur M, Thomas J, Jeelani S. Influence of functionalization on properties of MWCNT-epoxy nanocomposites. *Mater Sci Eng A.* 2011;528(3):1192–200.
- [34] Jojibabu P, Zhang YX, Rider AN, Wang J, Gangadhara Prusty B. Synergetic effects of carbon nanotubes and triblock copolymer on the lap shear strength of epoxy adhesive joints. *Compos Part B Eng.* 2019;178:107457.
- [35] Gojny FH, Wichmann MHG, Köpke U, Fiedler B, Schulte K. Carbon nanotube-reinforced epoxy-composites: Enhanced stiffness and fracture toughness at low nanotube content. *Compos Sci Technol.* 2004;64(15 SPEC. ISS.):2363–71.
- [36] Al-Maharma AY, Sendur P, Al-Huniti N. Critical review of the factors dominating the fracture toughness of CNT reinforced polymer composites. *Materials Research Express.* 2019;6(1):012003.
- [37] Razavi SMJ, Ayatollahi MR, Giv AN, Khoramishad H. International Journal of Adhesion and Adhesives Single lap joints bonded with structural adhesives reinforced with a mixture of silica nanoparticles and multi walled carbon nanotubes. *Int J Adhes Adhes.* 2018;80:76–86.
- [38] Ghosh PK, Patel A, Kumar K. Adhesive joining of copper using nano-filler composite adhesive. *Polymer.* 2016;87:159–69.
- [39] Yoonessi M, Lebroin-Coloïn M, Scheiman D, Meador MA. Carbon nanotube epoxy nanocomposites: The effects of interfacial modifications on the dynamic mechanical properties of the nanocomposites. *ACS Appl Mater Interfaces.* 2014;6(19):16621–30.
- [40] Saba N, Jawaid M, Allothman OY, Paridah MT. A review on dynamic mechanical properties of natural fibre reinforced polymer composites. *Constr Build Mater.* 2016;106:149–59.
- [41] May M, Wang HM, Akid R. Influence of adding multiwalled carbon nanotubes on the adhesive strength of composite epoxy / sol – gel materials. *J Coatings Technol Res.* Springer US; 2016;13(2):325–32.