

## PERFORMANCE CHARACTERISATION OF SILANE-TREATED MWCNT/COPPER WIRE-MESH REINFORCED EPOXY TWO-PHASE THERMAL INTERFACE COMPOSITE MATERIAL

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A two phase epoxy thermal interface composite material was prepared and characterized for their thermal conductivity, electrical insulation and mechanical properties. The principal aim of this work is to produce high performance thermal interface material having two phase structure with silane-treated copper micro wire-mesh and MWCNTs. The phase-I contains copper wire-mesh of 70 vol.% and MWCNTs of 0.5 & 1.0 vol.%, whereas phase-II contain only MWCNTs of 0.5 & 1.0 vol.%. The dielectric constant of two phase thermal interface material measures 3.2, which is almost equal to epoxy resin. But the maximum thermal conductivity measures 0.56 W/mK for 1.0 vol.% of surface-treated MWCNTs dispersed (phase 2) epoxy composite. The silane-treated reinforcements in epoxy matrix keep the composite mechanically stronger and tougher than as-received one. The TEM images show uniform dispersion of silane-treated MWCNTs and network formation in epoxy matrix.

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### 1. Introduction

Particle-dispersed polymer matrix thermal interface materials are capable of conducting heat from source and transmit to sink in electronic gadgets such as microprocessors, integrated circuits and electronic circuits. Heat removal from electronic gadgets plays major role in life span of gadgets and also electric charge dissipation could be a crucial issue [1]. Developing a polymer thermal interface material with thermally conductive and electrically insulative could be useful to extract more heat from electronic gadgets in the absence of current conduction. Epoxy could be used as matrix since it possesses low density, high strength and electrically insulative. MWCNTs could be used as filler material in epoxy since isolated MWCNTs are thermally good conductor (3000 W/mK) and electrically semi-conductor [2]. When high voltage of current is supplied to these material they starts to conduct due to electrical treeing, until that they acts as semi-conducting material. A two phase composite structure, which consist of rapid heat extraction phase and heat dissipation phase could be effective where quick and effective heat transfer is required [3]. The heat extraction phase (source) of TIM may consist of more volume percentage of continuous micro-wire mesh such as copper, aluminium & titanium and filled with high thermal conductivity fillers for speedy heat absorption from heat dissipation zone in electronic gadgets. Whereas the heat sink (phase 2) of TIM may contains high volume percentage of high thermal conductivity with low electrical conductive fillers for effective heat removal in the absence of electric charge dissipation. Surface-treatment of MWCNTs could fetch improved treeing structure

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(network) in polymer matrix, which ensures improved thermal conductivity results compared to as-received MWCNTs in epoxy matrix [4]. Similarly surface-treatment of metal wire-meshes could ensure low brittleness of matrix by reducing the cross-linking density along the wire-mesh-polymer interface [5]. The surface-modification could be done by acid hydrolysis process since it needs simple process parameters like low operating temperature, no controlled atmosphere and no high power requirements [6]. The two phase composite thermal interface material could be prepared by using hand lay-up method, where minimum process parameters like simple start-up arrangements, no high temperature involvement and low application of load [7]. These mechanically tougher high performance thermal interface materials could be used as heat extractor in computer hardware, programmable logic controllers and microcontroller systems, which may improve their life span and performance.

## 2. Experimental procedure

### 2.1. Materials

The two-phase thermal interface material was made up of following materials. The epoxy resin used here was liquid DGEBA resin (Huntsman India, Araldite LY556), having epoxide group of 195 g/mol and density of 1.2 g/cm<sup>3</sup>. Triethylenetetramine a low viscosity amine having kinematic viscosity of 20 cps and density of 0.98 g/cm<sup>3</sup> (Huntsman India Ltd. Mumbai., HY951) was used as curing agent. 3-Aminopropyltrimethoxysilane and MWCNTs of L/D ratio 110/40 nm were purchased from Sigma Aldrich, USA. The copper micro wire-mesh of diameter 350 μm and cross wire gap of 0.5 mm was purchased from Madras metal trading company, Pvt, Ltd, Chennai, India. Additional components like ethanol, acidic acid, pH indicator, which was used for silane-treatment process were purchased from Merck, India Ltd. The reinforcements MWCNTs and copper wire-mesh alone post treated whereas other chemicals and compounds were used in as-received condition.

### 2.2. Silane-treatment process

#### 2.2.1. Silane treatment of MWCNTs

The MWCNTs were silane-treated by acid hydrolysis process; in this the MWCNTs were allowed to etch by H<sub>2</sub>SO<sub>4</sub> of 2 N up to 1 h at 80 °C. The surface etched MWCNTs were then mixed in ethanol-water solution. The solution was prepared with diluted ethanol of 95% and 5% of water. Water is added to adjust the pH of solution with acetic acid in the range 4.5 to 5.5. The silane substance of 3 wt % was then added with ethanol-water solution and stirred continuously for 10 min to acquire homogeneous mixture. The silane-treated MWCNTs were separated using whatman filter paper and dried at 110 °C to form Si-O-Si structure. The functional group attachment on MWCNTs was confirmed by 1H NMR proton spectra. Fig. 1 shows the NMR peaks of silane-treated MWCNT with condensed silane reaction on MWCNTs surface. Table 1 shows the peak values and corresponding molecular groups.

Table 1. functional groups.

Peak value (ppm)	molecular group	presence
4.1	C-O	Silane-treated MWCNT
4.1	C-H	Silane (propyle group)
5.7	C=C-H	Silane-treated MWCNT

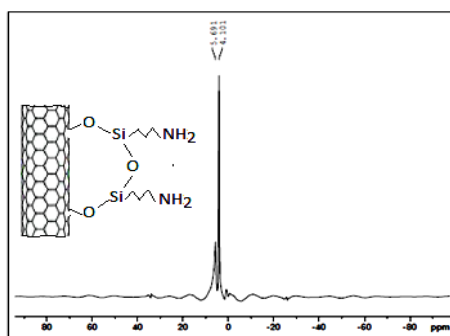


Fig. 1. NMR ( $^1\text{H}$  proton) spectra of silane-treated MWCNT.

### 2.2.2. Silane treatment of copper wire-mesh

The silane-treatment of copper wire-mesh was done by twofold process. First the copper wire-mesh was surface etched by  $\text{H}_2\text{SO}_4$  of 2N concentration. In this process the surface atoms are washed away, which produced rough surfaces and micro pits. Second the surface-etched wire-mesh was then immersed in ethanol-silane (4 wt.%) solution for 10 min to make the copper wire-mesh surface as  $\text{NH}_2$  functional group enriched surface. The acid leached surface facilitates enhanced silane attachment on wire-mesh's surface [8]. The silane solution's pH was maintained between 4.5 to 5.5 with the help of acetic acid and water for better acid hydrolysis. The silane-treated wire-mesh was then dried in a hot oven at  $120^\circ\text{C}$  to eliminate moisture and form Si-O-Si condensed structure. Fig. 2 shows the SEM image of surface leached copper wire-mesh.

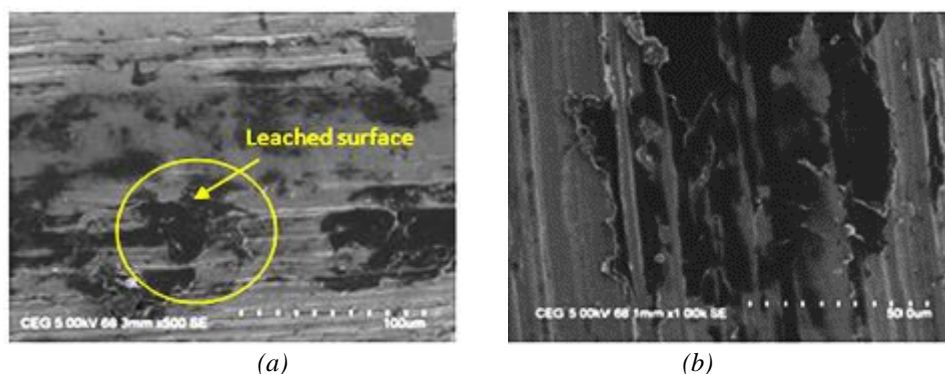


Fig. 2. (a & b) Acid etched copper wire-mesh.

## 2.3. Composite fabrication

The thermal interface material analyzed here was having two phase structure.

### Phase 1 [epoxy+copper wire-mesh and MWCNTs]

A fixed quantity of resin was mixed with 0.5 vol.% and 1.00 vol.% of MWCNTs. Curing agent was then added with resin-MWCNTs solution. The resulted colloidal suspension was then poured into a silicon rubber mould of 1.5 mm thick. 70 vol.% of copper wire-mesh was then reinforced into resin-MWCNTs solution and compressed to remove excess resin solution. The copper wire-mesh reinforced MWCNTs added epoxy composites were cured at room temperature for 24 h and post cured at 48 h. The copper wire-mesh and MWCNTs were added together in this phase for effective heat extraction from source.

### Phase 2 [epoxy+ MWCNTs]

In this phase MWCNTs alone were dispersed in epoxy matrix in the absence of copper wire-mesh to maintain insulation property of epoxy resin. The addition of copper wire-mesh in

phase 2 may conduct current since phase 1 is equipped with copper mesh. The composites may be good in thermal conductive but poor in electrical insulation, hence the phase 1 is equipped with MWCNTs alone. The fabricated phase 1 composite was used as base for building phase 2. The fabricated phase 1 composite material was act as base and the silica rubber mould was fixed over that. The MWCNTs of 0.5 vol.% and 1.0 vol.% added resin solution was poured into rubber mould and cured at room temperature for 24 h, and post cured for 48 h. Fig. 3 shows the graphical image of various form of two phase thermal interface composite material prepared.

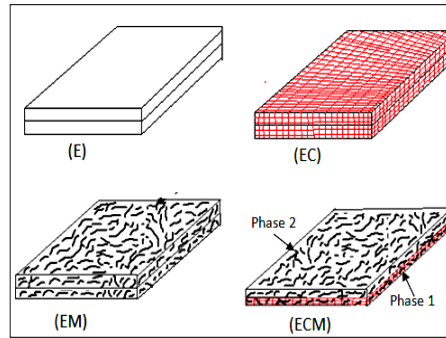


Fig. 3. Graphical image of various form of thermal interface composite material.

#### 2.4. Test specimen preparation

The fabricated two phase thermal interface composite material was checked for visual defects and flaws. The test samples based on ASTM standards were cut by an abrasive water jet machine (Maxium water jet, KENT, USA) having abrasive flow rate of 0.3 g/s, stand-off distance of 2.2 mm and operating pressure of 320 psi. Table 2 shows the composite designations and compositions prepared.

Table 2. Composition and designation of composites.

Composite designation	Epoxy vol.%		Copper wire-mesh vol.%		MWCNT vol.%	
	Phase 1	Phase 2	Phase 1	Phase 2	Phase 1	Phase 2
E	65	35	0	0	0	0
EC	19.5	10.5	40	30	0	0
EM <sub>1</sub>	64.5	34.5	0	0	0.5	0.5
EM <sub>2</sub>	64	34	0	0	1.0	1.0
ECM <sub>1</sub>	19	10	70	-	0.5	0.5
ECM <sub>2</sub>	18.5	9.5	70	-	1.0	1.0

E- Epoxy; C- Copper wire-mesh; M-Multi-walled carbon nanotube

#### 2.5. General characterization

##### 2.5.1. Thermal conductivity

The thermal conductivity of two-phase composite thermal interface material was tested using Lee's disc method. Circular specimens of diameter 11.2 cm were tested to compute average value of thermal conductivity [9].

##### 2.5.2. Dielectric analysis

The dielectric constant and loss of two phase composite material was tested using a LCR HI-Tester (HIOKI-3532-50, JAPAN) followed by ASTM D-150. Test samples of 13 mm diameter were prepared from various composites. The test samples were kept between two parallel conductors as parallel plate capacitor and current was allowed to pass through this in various frequencies. The frequency range used in LCR meter for this present study was from 50 Hz to 500

Hz since; the operating current frequency of electronic gadgets varies between 50 Hz to maximum 150 Hz. The test was repeated in five identical samples of each composite designation.

### 2.5.3. Mechanical characterization

The tensile and flexural strength of fabricated two-phase thermal interface material was tested based on ASTM-D 638 and ASTM-D 790 respectively. A universal testing machine having 5 ton of loading capacity was used for analysing the samples. The machine was equipped with digital encoder and moving at a cross head speed of 2.5 mm/min. Five identical test specimens were tested to find the average.

## 3. Results and discussion

### 3.1. Thermal conductivity

Fig. 4 shows the thermal conductivity graph of various composite designations with as-received and surface-modified reinforcements. The thermal conductivity of pure epoxy resin gives 0.22 W/mK. The additions of copper wire-mesh and MWCNTs into epoxy resin increased its thermal conductivity. The improved thermal conductivity of 37% is observed for copper wire-mesh reinforced epoxy two-phase composite material. This improvement is because of addition of continuous copper wire-mesh, which could transfer the heat from heat source and pass it to heat sink. Similarly, addition of as-received and surface-modified 0.5 vol.% MWCNTs into epoxy resin in both the phases gives improved thermal conductivity of 29% and 42% respectively. Whereas addition of as-received and surface-modified 1.0 vol.% MWCNTs into epoxy resin gives improved thermal conductivity as 35% and 46% respectively. This improvement is because of addition of MWCNTs transmit the heat from source to sink by continues atomic structure.

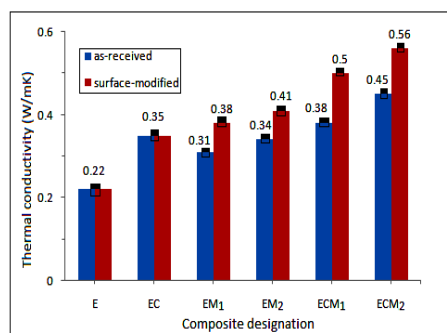
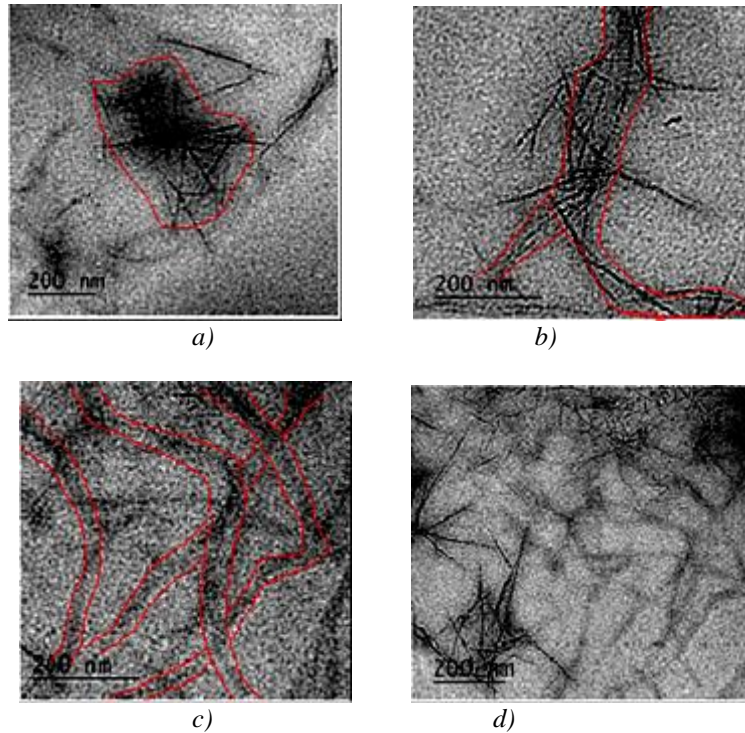


Fig. 4. Thermal conductivity of various composites.

It is observed that the surface-modified MWCNTs in epoxy resin gives improved thermal conductivity on comparing as-received composite designation (EM<sub>1</sub> & EM<sub>2</sub>). This improvement is the cause of effective MWCNTs network in epoxy matrix [10]. Figure 4 shows the TEM images of as-received and surface-modified MWCNTs in epoxy resin. Figure 5(a) shows clustered image of MWCNTs. Figure 5 (b, c & d) shows surface-modified MWCNTs in matrix. The surface-modified MWCNTs dispersed on epoxy matrix more uniformly compared to as-received MWCNTs in matrix. The effective dispersion and network formation led improvement in thermal conductivity. It is observed that the thermal conductivity of composite designations ECM<sub>1</sub> and ECM<sub>2</sub> found to be higher among all composite designations. The improvement of 42% and 51% were observed for as-received composite designation ECM<sub>1</sub> and ECM<sub>2</sub> respectively. But the surface-modified composite designations give improved thermal conductivity of 56% and 61% for ECM<sub>1</sub> and ECM<sub>2</sub> designations. This improvement is because of addition of copper micro-wire and MWCNTs in phase1. The 70 vol.% of copper improved the thermal conductivity by its long continues structure. Compare than particle addition these micro wire mesh structures able to conduct the temperature more effectively [11]. The extracted heat from source transferred effectively through carbon

nanotubes since they acts as heat carriers. This is because the MWCNTs also posses short continues structure, which enables more heat transfer. Figure 5 (b & c) shows network formation of short continues MWCNTs. The formation of more branches in MWCNTs acts a bridge to transfer heat from source to sink.



*Fig. 5. shows TEM images of (a) as-received MWCNTs in epoxy matrix shows cluster, (b, c & d) surface-modified MWCNTs in epoxy matrix shows continuous network.*

### 3.2. Dielectric analysis

Fig. 6 shows the dielectric properties of surface-modified MWCNTs added copper wire mesh-reinforced composite thermal interface materials in room temperature and 100 °C. The previously reported article confirmed that, the surface-treated MWCNTs into epoxy resin dispersed in more uniform manner compared to as-received MWCNTs [12 & 13]. Hence the surface-modified MWCNTs alone used to make two phase thermal interface composite material.



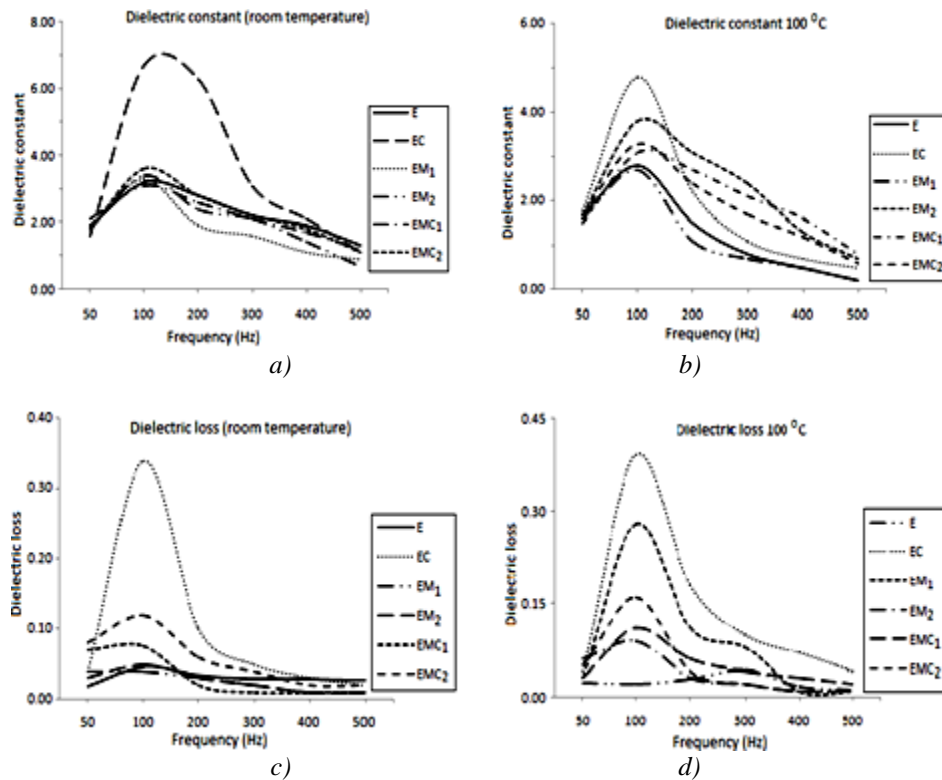


Fig. 6 shows (a) dielectric constant of composites in RT, (b) dielectric constant in elevated temperature (100 °C), (c) dielectric loss in RT and (d) dielectric loss in 100 °C.

Fig. 6 (a & b) shows the dielectric constant of various composites in room temperature and 100 °C. The maximum dielectric constant of epoxy measures 3.2 and 2.8 in room and elevated temperature. The decrease in dielectric constant of 12.5% was observed in elevated temperature. This decrement is because of increased kinetic energy of epoxy molecules in elevated temperature. Since the glass transition temperature of epoxy starts around 65 °C the ability of molecules to move is high thereby polarization affected. The addition of copper in both phase 1 and 2 increased the dielectric constant of composite as 6.7 at 100 Hz in room temperature. This improvement is because of addition of continuous micro wire-mesh in epoxy matrix. At 100 Hz usually polymers get polarized and start transfers the charges. The presence of continuous copper wire-mesh receives the triggered electrons and start conduct. But increment in frequency further hampered the charge flow because of poor relaxation time to dielectric to be polarized. It is observed that the dielectric constant of composite designation EC at 100 °C measures 4.8. This decrement in constant is the cause of poor polarization of epoxy matrix due to increased kinetic energy of epoxy molecules, which affects the electron triggering to copper wire-mesh.

The dielectric constant of composite designations EM<sub>1</sub> and EM<sub>2</sub> measures 3.3 and 3.4. The addition of MWCNTs acts as semiconductor and not fetched any improvement in current conductivity. Compared to epoxy resin the EM<sub>1</sub> and EM<sub>2</sub> gives almost same result, which explicit the unaltered insulation property of composite after dispersing MWCNTs. This is because of poor electrical conductivity of MWCNTs in lateral dimension, which ensures lower current conductivity. The Figure 5(b) shows the dielectric constant of composite designation EM<sub>1</sub> and EM<sub>2</sub> in 100 °C. From graph it is observed that the increase of temperature affected the charge transfer in composite material. When temperature increases the secondary molecules in epoxy resin get enough kinetic energy to rotate thereby the molecules are misaligned with external frequency. This phenomenon affected the polarisation in encapsulated epoxy thermal interface material [14 & 15]. The maximum dielectric constant of 2.7 and 3.8 was observed for composite designations EM<sub>1</sub> and EM<sub>2</sub> respectively. The dielectric constant further decreased with increase of frequency. This decrement is because of poor dielectric polarization due to lower relaxation time

given to polymer composite material. Anyway compare to E and EC composite designation improved dielectric constant was observed. It is further noted that the composite designation  $EMC_1$  and  $EMC_2$  gives maximum dielectric constant of 3.1 and 3.6 in room temperature. This value is almost similar to the dielectric constant of epoxy resin, which measures 3.2. The two-phase composite structure ensures the non flow of current in high frequency. Though copper and MWCNTs are present in epoxy still the dielectric constant of composite designations  $EMC_1$  and  $EMC_2$  are almost similar to epoxy resin. This is because in phase-1 the presence of copper alone conducts the charges and MWCNTs reminds semiconductor whereas in phase-2 the presence of MWCNTs not responds to external frequency thereby affects the continue charge transfer from phase-1 and affects the total permittivity of composite. Similar dielectric constant values were observed in composite designations  $EMC_1$  and  $EMC_2$  (3.1 and 3.3) at 100 °C.

Fig. 6 (c & d) show the dielectric loss of composites in variable frequencies at room temperature and 100 °C. It is observed that the dielectric loss of pure epoxy measures 0.05 at 100 Hz. In other frequencies the composites show lower loss value. This is because of at 100 Hz polarization takes place and charges about to move. But due to high insulation of epoxy resin the transferred charges converted as heat. It is observed that when frequency increases the loss also decreases. This is because when frequency increases polarization depleted. The poor polarization never allows the charges to transfer thereby no heat generation was observed at higher frequencies [16 & 17]. It is observed that the improved dielectric loss of 85%, 38% and 58% were observed for composite designations EC,  $ECM_1$  and  $ECM_2$  respectively. This large heat dissipation is the cause of increased frequency, which is applied over the copper contain composite. The composite designations  $EM_1$  and  $EM_2$  give 0.04 and 0.05 respectively, which is almost equal to epoxy resin. Similar results were observed in composites at 100 °C. The pure epoxy measures a dielectric loss of 0.02 whereas, the improved dielectric loss of 94.8%, 92%, 78%, 81% and 87.5% were observed at 100 °C for composite designations EC,  $EM_1$ ,  $EM_2$ ,  $EMC_1$  and  $EMC_2$  respectively. This increase of heat dissipation is the cause of misalignment of molecules due to higher energy. At higher temperature the epoxy molecules are gained more energy and start rotates freely away from primary C-C molecular chain. This phenomenon affects the polarization and increase the heat dissipation effect [18 & 19]. Thus the two phase composite structure, which consist of both copper and MWCNTs in phase-1 and MWCNTs alone in phase-2 ( $ECM_1$  and  $ECM_2$ ) resulted almost same dielectric constant compared to pure epoxy resin.

### 3.3. Mechanical properties

Fig. 7 shows the tensile and flexural strength of as-received and surface-modified MWCNT and copper wire mesh reinforced epoxy two phase thermal interface material. It is observed that the tensile strength of pure epoxy measures 65 MPa. The addition of as-received reinforcement further changes the tensile strength. The decrement of 8%, 3% and 6% were observed for composite designations EC,  $EMC_1$  and  $EMC_2$  respectively. This decrement is the cause of increase of cross-linking density in epoxy matrix by the addition of as-received copper microwave. The positive copper atoms attract the free OH molecules in epoxy matrix and form higher cross linking density along the polymer-wire mesh interface, which lead brittle fracture when tensile load is applied [20 & 21]. The addition of MWCNTs alone to epoxy resin in both phase 1 and 2 marginally increased the tensile strength. Improvement of 13.3% and 7.1% were observed for  $EM_1$  and  $EM_2$  composite designations. This improvement is because of load sharing ability of continuous MWCNTs in epoxy matrix, which intern reduce the stress concentration on matrix [22 & 23]. It is further noted that the additions of surface-modified MWCNTs and copper wire mesh in to epoxy resin in phase 1 and phase 2 improved the mechanical properties. The improvement of 9.7%, 26%, 31%, 17% and 13% were observed for composite designations EC,  $EM_1$ ,  $EM_2$ ,  $EMC_1$  and  $EMC_2$  respectively. This improvement is because of addition of surface-modified MWCNTs and copper wire mesh. Highest tensile strength of 94% was observed for composite designation  $EM_2$ . This improvement is the cause of uniform dispersion of surface-modified MWCNTs, which transfer the applied load effectively in matrix and reduce the stress concentration. The composite designation  $EMC_2$  gives 17% of improvement in tensile strength, which shows that surface-modified copper wire mesh restrict the high cross-link formation on wire mesh-polymer interface. This is because surface-modified copper wire mesh covered by silane



substance, which intern reduce the attraction of OH molecules from epoxy resin to form high cross-linking nature [24 & 25]. Similarly the surface-modified copper wire mesh chemically bond with epoxy matrix, which ensures the effective load transfer between matrix and metal wire mesh.

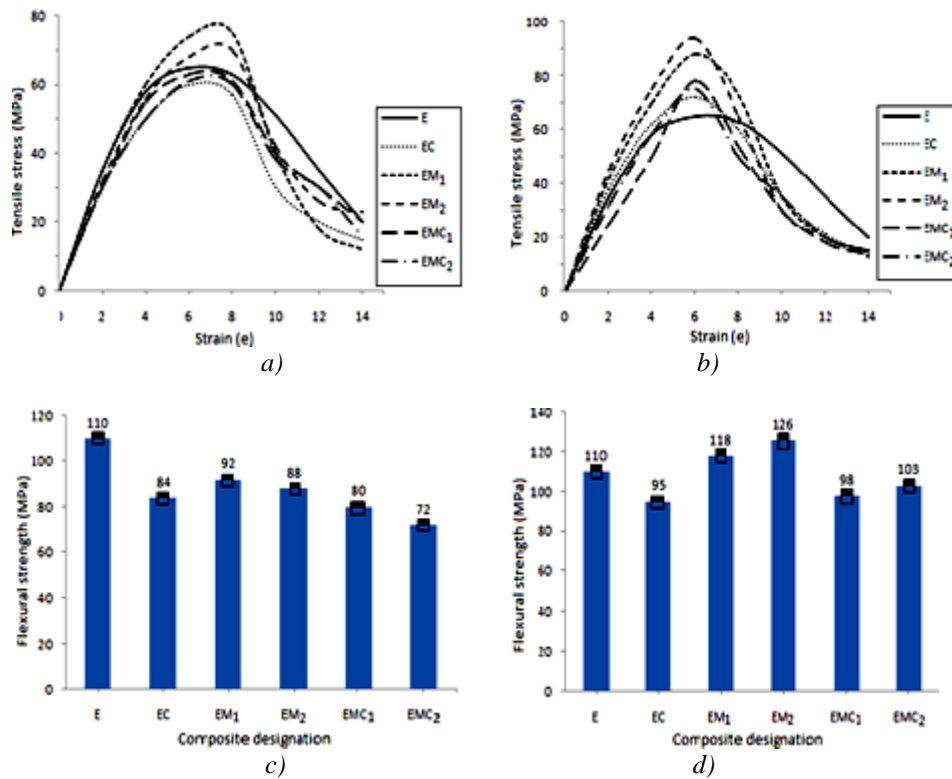


Fig. 7. Shows (a and c) tensile and flexural strength of as-received MWCNT/copper reinforced composite, (b and d) shows tensile and flexural strength of surface-modified MWCNT/copper reinforced composite.

The flexural strength of as-received MWCNTs and copper wire mesh in epoxy resin two phase composite material shows downward trend. The decrement in flexural strength of 24%, 16%, 20%, 27% and 34% were observed for composite designations EC, EM<sub>1</sub>, EM<sub>2</sub>, EMC<sub>1</sub> and EMC<sub>2</sub> respectively. This decrement is because of as-received MWCNTs formed cluster in matrix and hampered the effective load transfer. Similarly the as-received copper micro wire mesh attracts the unsaturated OH molecules and created highly cross linked wire mesh-polymer interface. These phenomena significantly affected the flexural properties of composite TIM [26]. But the composite contain surface-modified MWCNTs and copper wire-mesh in epoxy resin gives improved flexural strength compared to as-received reinforcements contains epoxy composite. The improvement of 7% and 13% were observed for EM<sub>1</sub> and EM<sub>2</sub> composite designations on comparing pure epoxy. The uniformly dispersed surface-modified MWCNTs are the cause of this improvement. The other composite designations also shows improved flexural strength on comparing as-received reinforcements based composite. The uniform dispersion of MWCNTs, hindrance of forming large cross-linking on wire mesh-polymer interface and effective load transfer between wire mesh and epoxy through chemical bonding are the causes for this improvement.

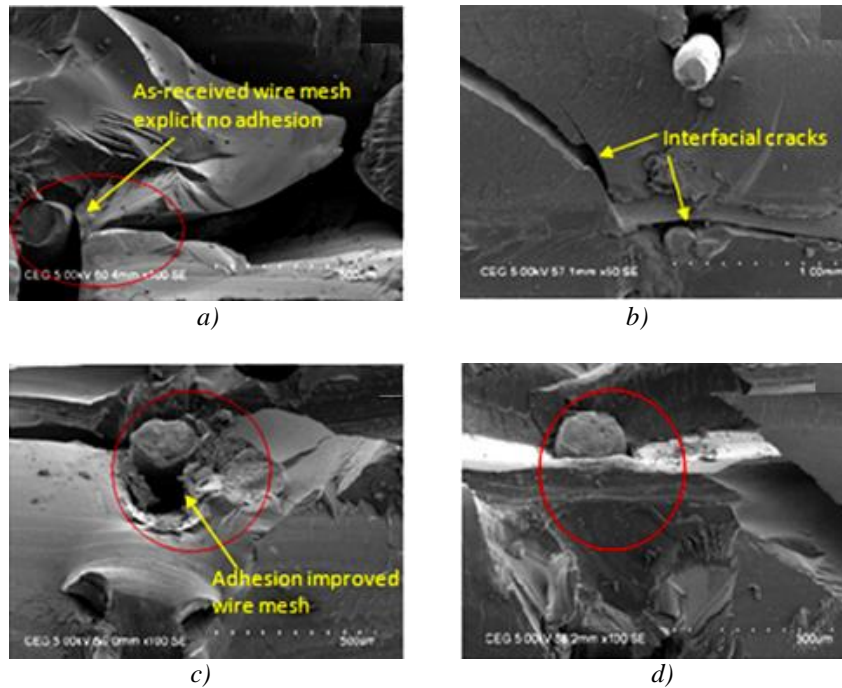


Fig. 8. Shows fractograph of (a & b ) as-received copper wire mesh in epoxy matrix and (c & d) surface-modified copper wire mesh in epoxy matrix.

Fig. 8 shows the fractograph of composites under mechanical loading. The SEM micro analysis was performed using a scanning electron microscope 1515, HITACHI, JAPAN. The samples were surface coated with gold before testing. Fig. 8 (a & b) shows as-received copper wire mesh in epoxy matrix. The interfacial cracks in matrix were observed along the wire mesh-polymer interfaces, which explicit poor adhesion of wire mesh with epoxy matrix. Figure 8(c & d) shows the adhesion improved copper wire mesh with epoxy resin. The interfacial cracks were suppressed and improved adhesion on wire mesh surface resulted improved tensile and flexural properties [27].

#### 4. Conclusion

A two phase composite thermal interface material was fabricated using MWCNTs and copper micro wire mesh and analysed for thermal conductivity, dielectric properties and mechanical behaviour. The two phase composite structure, which consist of MWCNTs and copper in phase 1 and MWCNTs alone in phase 2 giving better performance in thermal conductivity and dielectric performance compare to other composite structures. The two phase composite thermal interface material of designation (EMC) has the ability of transferring heat from source to sink in the absence of current dissipation. Very high thermal conductivity and lower dielectric constant is the functional key of this composite designation. The surface-modified MWCNTs and copper wire mesh shows improved performance compare to as-received MWCNTs and copper wire mesh in epoxy matrix. The surface-modified reinforcements in epoxy matrix show improved tensile and flexural properties. Hence the composite thermal interface material is mechanically stronger and tougher with superior thermal conductivity and electrical insulation. These high performance and mechanical stronger materials could be used in electronic gadgets to remove excess heat and improves their life span.

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