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Enhancing the thermal conductivity of polymer composites

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ABSTRACT

Carbon fiber-reinforced Polymericomposites (CFRP) are becoming the most important structural materials for aircraft structures, satellites, missiles, and other aerospace applications. However, extending these materials for thermal and electronic applications is restricted by the poor thermal conductivity. In order to improve the thermal conductivity of CFRP composites, two methods were followed. In the first method, the polymer matrix was made conductive by incorporating various fillers (copper powder, carbon nanotubes and graphene). Using these conductive matrices, CFRP composites were developed by vacuum bag molding technique. In the second approach, in addition to fillers, a continuous copper mesh was embedded for fabricating the CFRP composite. The in-plane and out- of -plane thermal conductivity of the CFRP composites was measured by hot disc. From the results obtained, it was found that the in-plane thermal conductivity of the CFRP decreased with the filler inclusion whereas the out- of- plane conductivity of the composite was enhanced. In case of copper mesh embedded CFRP composites, both in the plane and out of plane thermal conductivity was enhanced further, the effect of filler addition and copper mesh inclusion on the mechanical strength was studied by measuring the interlaminar shear strength (ILSS) and was substantiated by NDT characterization.

Keywords— *Carbon fiber reinforced composites (CFRP), Carbon nanotubes (CNT), Copper mesh, vacuum bag technique, thermal conductivity, Hot disc equipment*

1. INTRODUCTION

Carbon fiber reinforced polymer composites (CFRP) are gaining importance as structural materials in aircraft industries due to their light weight combined with superior strength and stiffness properties. As the miniaturization and weight saving demands are increasing even for microelectronic applications related to aircraft, CFRPs are gaining importance in these applications too. However, the heat dissipation is becoming the most critical parameter to be addressed in this context. Because of heat generation, the performance and life of the devices will be severely affected. [1–4].Though carbon fibers are having in-plane thermal conductivity in the range of 8-70 W/mK.[15] the CFRP exhibit the thermal conductivity values of 2-3 W/(mK) in fiber direction and 0.5-0.6 W/(m·K) in the through-the-thickness direction [5]. Because the epoxy resins widely used in this CFRPS exhibit very low the conductivity in the range of (0.15–0.25 W/mK [6-7]. The general strategy to increase the thermal conductivity of polymers is doping materials with high thermal conductivities, such as ceramics [8-11], metals powders [12], carbon nanotubes [13] and grapheme [14] carbon black, carbon fibers [15-19]. Among these doping materials, carbon-based fillers are found to be the best promising fillers, with high thermal conductivity and lightweight [20-21]. It is worth noticing that significant scatter of data are typically reported for thermal conductivity of fillers. This is caused by several factors, including filler purity, crystallinity, and particle size and measurement method.

The thermal conductivity of multiwalled carbon nanotubes and graphene powder was reported to be ~3,000 W/mK [22-24] and 100-400 W/mk [15] respectively. Single graphene sheets constituting graphite show intrinsically high thermal conductivity of about 800W/m K[25] or higher (theoretically estimated to be as high as 5300W/m K[26-27] , this determining the high thermal conductivity of graphite, usually reported in the range from 100 to 400W/m K. Expanded graphite (EG), an exfoliated form of graphite with layers of 20–100 nm thickness, has also been used in polymer composites[28] for which the thermal conductivity depends on the exfoliation degree[29]. Carbon black is reported to contribute to electrical conductivity rather than thermal conductivity [33-30].

Many studies have been reported the improvement either in in-plane or in out-of-plane thermal conductivity. However, for all the practical purposes, it is necessary to improve both in-plane and out of plane thermal conductivity. Further, it was reported that CFRPs while exhibiting the enhanced thermal conductivity with the inclusion of fillers, showed a declining trend in mechanical properties.

In the present study, an effort has been made to enhance out of plane thermal conductivity of Epoxy/CFRP with the Carbon nanotube (CNT) and graphene inclusions and both in the plane and out of plane thermal conductivity with copper mesh inclusion without compromising on the interlaminar shear strength of the CFRP.

The enhancement of thermal conductivity is substantiated with the volume fraction calculations and NDT.

2. EXPERIMENTAL

2.1. Materials

The polymer matrix system used is a commercially available aerospace grade epoxy resin LY5052 with density 1.17 [g/cm3] Viscosity @ 25 °C 1000 – 1500 [cps.] with amine curing agent Aradur 5052 supplied by M/s. Huntsman Advanced Materials (India) Private Limited, Mumbai, India.

Carbon nanotubes with a diameter ranging from 15-25mm, length ranging from 5-15mm with a purity of 99.1% were procured from Nanoshell, LLC, USA.

Graphene powder with 99.5% purity, with layer thickness< 1.6 nanometers, platelet planar 0.32 to 5 micrometers, 0.0215 gm.cc supplied by Nanoshell, LLC, USA.

Laboratory grade Copper powder with particle size less than 63 microns was procured from Nice Chemicals.

The dimension of the copper mesh is 50*50mm, 0.002inch diameter and it was procured from Hawkeye International Limited. The carbon fabric used is a 210GSM, bidirectional, and has a dimension of 300*3500mm. The carbon fabric was procured from Hindoostan Mills LTD, Mumbai

2.2 Preparation of conductive matrix.

Various conductive matrices were prepared by dispersing the fillers into the epoxy polymer using THINKY Mixer Model: ARE-310. Initially, the mixture was subjected to 1000 rpm for 5 min, 1500 rpm for 5 min and 1800 rpm for 5 min followed by deaeration at 2000 rpm for 2 min to remove the air trap. The resin to curing agent ratio was maintained 100:40. The filler weight % was maintained in such a way that the viscosity of the matrix is convenient enough to wet the carbon fabric reinforcement during the preparation of CFRP. Table 2.1 presents details of conductive matrices prepared. A controlled formulation without any filler was prepared for comparative study

Table 2: Details of conductive matrices

S.no	Resin wt %	Curing agent wt %	Filler type	Percentage of filler with respect to the resin
1.	100	40	Carbon nanotubes	2.5
2.	100	40	Graphene	40
3	100	40	Copper powder	40
4	100	40	--	--

2.3 Preparation of CFRP composites

CFRP composites of dimensions 300*300*0.2mm were prepared using Vacuum bag moulding technique. The fiber to resin ratio was maintained as 60:40. Hand layup technique was used to wet the Caron Fabric. Vacuum bag set up as shown in figure 1 was used for fabricating various CFRP composites. Table 2 presents the details of various CFRP composites.

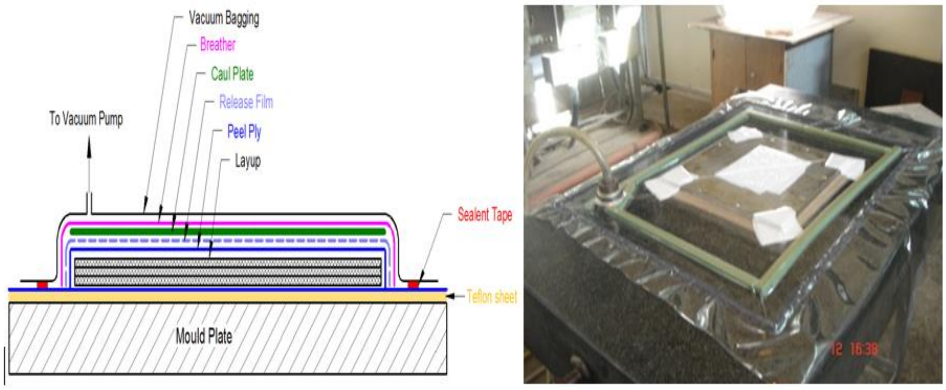


Fig. 1: Vacuum bag setup

Composite laminates (CFRP) were allowed to cure @ room temperature for 24 hrs later post cured @100°C for 4 hrs using a hot air oven.

Table 2: Details of the CFRP composites

S. No	Details of the composition (Resin/Curing Agent/Filler/Mesh/Reinforcement)	Designation
1	LY5052/Ardur5052 with Cabon fiber reinforcement	CFRP control
2.	LY5052/Ardur5052/CNT with Cabon fiber reinforcement	CNT filled CFRP
3.	LY5052/Ardur5052/Graphene with Cabon fiber reinforcement	Graphene filled CFRP
4.	LY5052/Ardur5052/Copper powder with Carbon fiber reinforcement	Copper powder filled CFRP
5.	LY5052/Ardur5052/Two layers of copper mesh embedded inside the CFRP	CFRP with Copper mesh embedded inside
6.	LY5052/Ardur5052/ Two layers of copper mesh embedded outside the CFRP	CFRP with Copper mesh embedded outside

2.4. Nondestructive test

NDT analysis of composites was carried out for assisting quality of the laminates. The composite quality was examined by Ultrasonic wave penetration and the data was represented by a C-scan which provided the features of a defect like delamination and voids. The specimen was mounted in the Ultrasonic scanning equipment. Water was used as a medium to transfer signal from transmitter to specimen and specimen to receiver. Inspection process was carried out and data was represented in the computer screen as a color code Color corresponding to the signal intensity for each position of the specimen was indicated. Figures2-5 depict the C-scans of CFRP control, CNT filled CFRP, CFRP with Copper mesh embedded inside, CFRP with Copper mesh embedded outside respectively

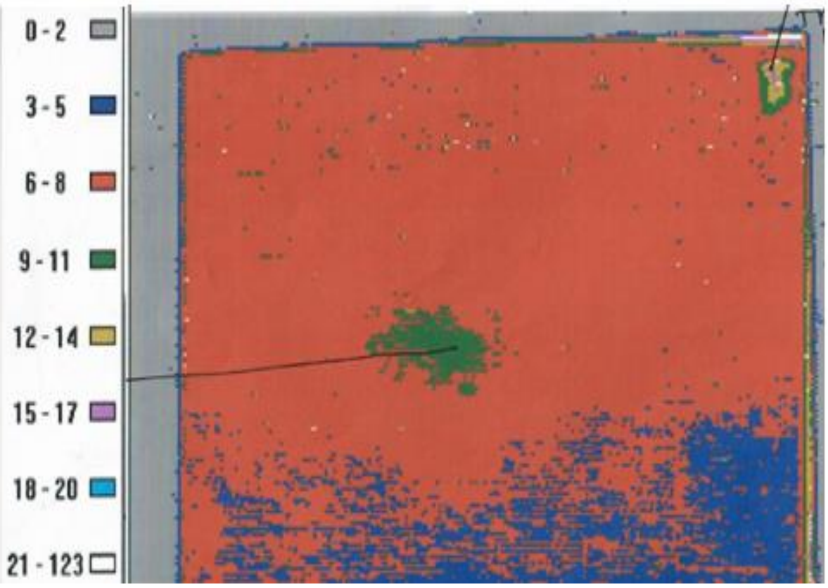
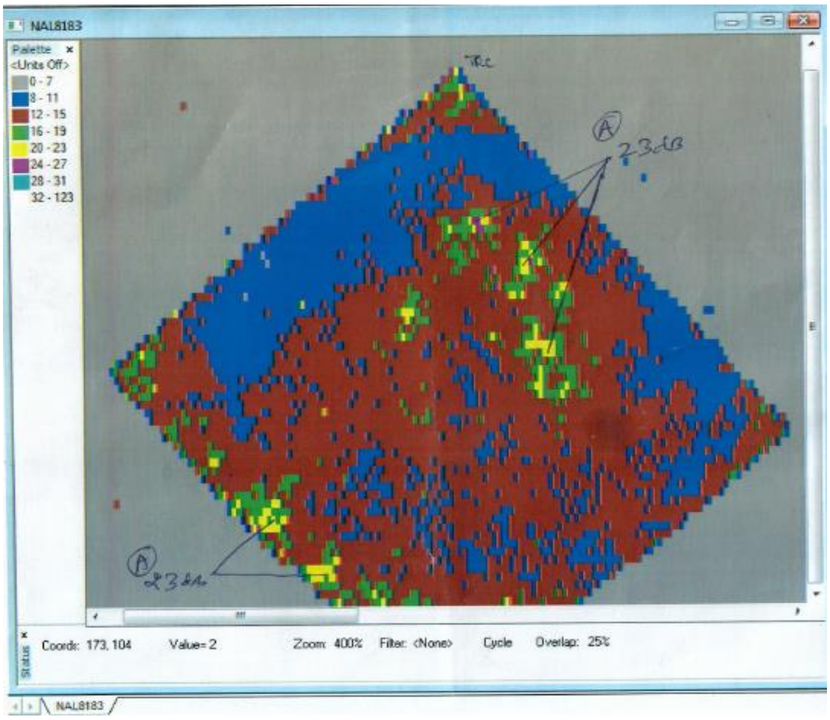


Fig. 2: CFRP control



Coords: 173, 104 Value= 2 Zoom: 400% Filter: <None> Cycle Overlap: 25%

Fig. 3: CNT filled CFRP

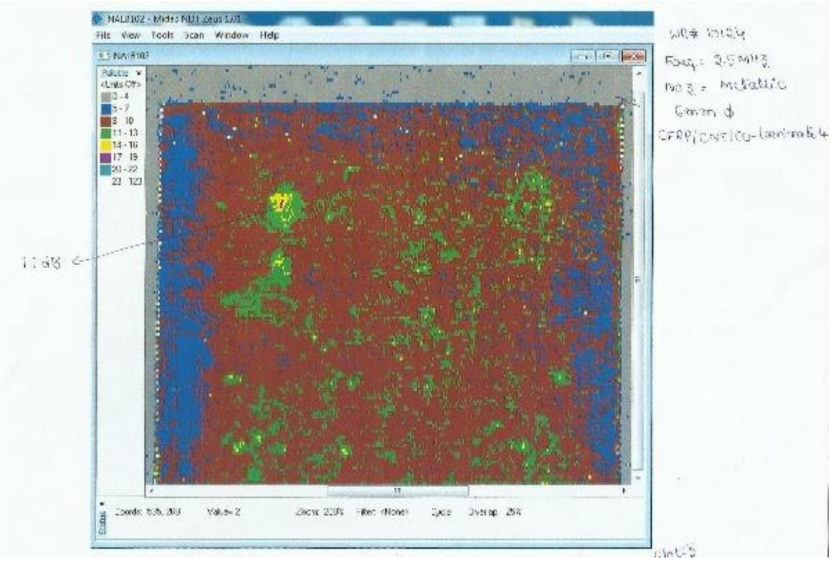


Fig. 4: CFRP with Copper mesh embedded inside

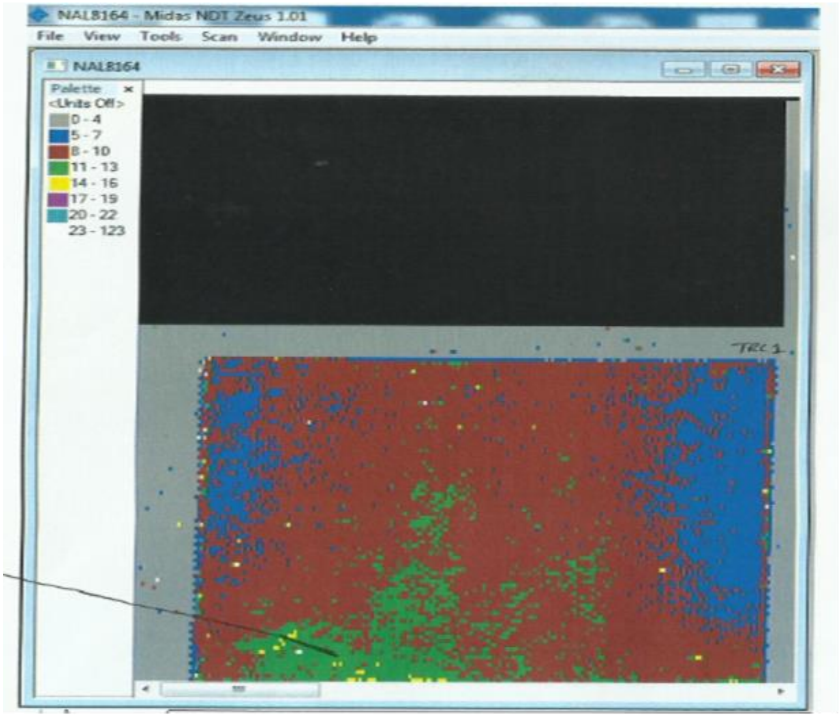


Fig. 5: CFRP with Copper mesh outside on both sides

2.5. Volume fraction Calculations

The Volume fraction of the reinforcement and the polymer matrix were calculated using the formula ----

Volume fraction the resin can be calculated using the formula $V_r = 1\text{-}V_{f.}$
Where, V_f is the volume fraction of fiber.

$V_f = 1/((1+ \rho_r/ \rho_f (W_f/))$ Where ρ_r is the density of the resin, ρ_f is the density of the fiber, W_f is the weight fraction of the fiber and W_r is the weight fraction of the resin. Table3 presents the calculated volume fractions of the fiber and the polymer matrices for various composites

Table 3: Calculated volume fractions of the fiber and the polymer matrices for various composites

S .no	Different laminates	Volume fraction	
		Fiber	Resin
1.	CFRP control	0.39	0.61
2.	CNT filled CFRP	0.35	0.65
3.	Graphene filled CFRP	0.35	0.65
4.	Copper powder filled CFRP	0.35	0.65
5.	CFRP with Copper mesh embedded inside	0.35	0.65
6.	CFRP with Copper mesh embedded outside	0.35	0.65

2.6 Thermal conductivity measurements by Hot disc method

The hot-disk method utilizes a Nickle sensor which acts as both a heat source as well as a thermometer. The source and the thermometer will measure the changes in the temperature of the specimen and the increase in the time-dependent temperature. Two square CFRP composite samples with the dimensions 20*20*2mm and 15*15*2mm were used for measuring the specific heat capacity and thermal conductivity measurements respectively. Figure 6 depicts the test samples.

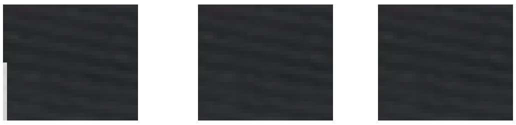


Fig. 6: Test specimens

The sensor was sandwiched between two test specimens of the same configuration as shown in the figure. The current was passed through the nickel sensor. Once the sensor gets heated, heat dissipates throughout the specimen placed on either side. By comparing the temperature versus the time response in the sensor, the thermal conductivity was calculated and displayed directly. Fig-7: present the schematic view of the test arrangement

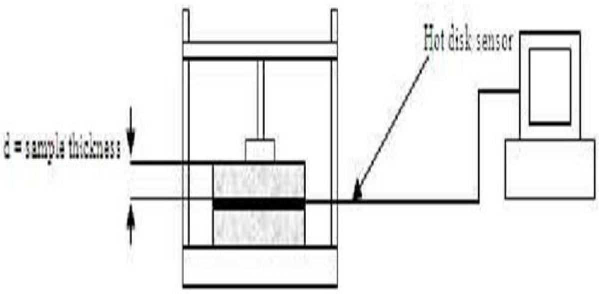


Fig. 7: Schematic view of a test arrangement

The thermal conductivity values measured for various CFRP composites are presented in the Table 4.

Table 4: Thermal conductivity results

S. no	Different CFRP laminates	In-plane thermal conductivity (W/mK)	Out of plane thermal conductivity (W/mK)	The volume fraction of the resin
1.	CFRP control	2.01	0.21	0.61
2.	CNT filled CFRP	1.55	0.55	0.65
3.	Graphene filled CFRP	1.17	0.86	0.65
4.	Copper powder filled CFRP	1.02	0.65	0.65
5.	CFRP with Copper mesh embedded inside	2.70	0.38	0.65
6.	CFRP with Copper mesh embedded out side	3.50	0.40	0.65

2.7 Mechanical testing

The shear parallels to the plane of the lamina of a composite, defined as the Interlaminar shear strength (ILSS), was determined in accordance with ASTM D2344 using universal testing machine INSTRON5982with a load cell capacity of 100KN. Specimens of 20 *10* mm were used with a crosshead speed of 2mm/min Fig 8 shows specimen under ILSS test.

Fig. 8: Specimen under ILSS evaluation

Table 5: The Interlaminar shear strength of CFRP composites

S.no	Different CFRP laminates	ILSS RESULTS (MPa)
1.	CFRP control	40±1
2.	CNT filled CFRP	32±1
3.	Graphene filled CFRP	46±1
4.	Copper powder filled CFRP	48±1
5.	CFRP with Copper mesh embedded inside	43±1
6.	CFRP with Copper mesh embedded outside	41±1

3. RESULTS AND DISCUSSIONS

From the NDT analysis of the various CFRP composites and C-scans presented in figs2-5**,** it is observed that the attenuation of the wave was well within 10 dB for plain CFRP copper mesh embedded CFRP indicating the good quality of the laminates. In the case of CNT filled laminates more scattering is observed with higher dB values (above 10) indicating improper compaction. These results were further confirmed by measuring the Interlaminar shear strength of the composites. From table 5 it is seen that the ILSS values have been improved with the addition of fillers as well as with the copper mesh. Except in the case of CNT filled CFRP composite. Highest ILSS values were realized for Copper filled CFRP. This was due to the improper compaction that leads to decrease the strength.

The thermal conductivity measurements revealed the fact that the in-plane thermal conductivity of CFRP decreased with the incorporation of fillers. This is due to the fact that the volume fraction of resin (v_r) in the composite increases with the addition of fillers. The composite volume fraction is the sum of v_f and $v_r = 1$. Hence when v_r increases the v_f decreases. This was clearly seen from the values presented in table 3. Since the fiber conductivity is many times higher than resin conductivity, the magnitude of the decrease in conductivity will be more influenced by the decrease in fiber volume fraction. Hence the in-plane thermal conductivity of composite Kc = υf * Kf + υm * Km (where Kf is the thermal conductivity of the fiber and Kmis the resin matrix conductivity) decreases due to the addition of filler. From the thermal conductivity measurements, it is seen that all the CFRP composites filled with carbon-based fillers and mesh due to the same reason the in-plane thermal conductivity was reduced. The out of plane thermal conductivity of the CFRPs was enhanced due to the addition of fillers this was due to the improved layer to layer contact. throughout the thickness of the composite. Introduction of copper mesh into the CFRP resulted in the drastic enhancement of both in-plane thermal conductivity and out of plane thermal conductivity. Further, it is seen that the CFRP composite with copper mesh outside exhibited higher in-plane thermal conductivity as compared to the CFRP composite having the mesh in the side. This is due to the fact that the in-plane thermal conductivity was influenced by the highly conductive copper mesh present at the surface of the composite.

4. CONCLUSION

From our research studies it was found that with the inclusion of conductive fillers into the resin matrix, the out of plane thermal conductivity of CFRP composites was enhanced whereas the in-plane thermal conductivity showed a declining trend. The copper mesh embedding further into the filled Composite resulted in enhancement both in-plane and out of plane thermal conductivity. The percentage of filler inclusion was optimized for both processing and ensuring the better Interlaminar shear strength of the composite. Which is very sensitive to the processing conditions. Further improvement of thermal conductivity can be achieved by layer to layer contact in the composite.

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