

Thermal Synergistic Effect on CFRP Laminates with Modified Fiber/Matrix Systems for Heat Transfer Applications

J. Aravind, M. Manu, K. E. Reby Roy,* M. Mubarak Ali, Shukur Bin Abu Hassan, S. Rajalekshmi, and Desin S. Daniel

Carbon fiber reinforced polymer (CFRP) laminates are modified to enhance their suitability for various thermal applications. A synergistic approach utilizing the effect of various conductive and insulative modifiers with diglycidyl ethers of bisphenol A (DGEBA) epoxy resin and/carbon fiber (CF) is explored. In CFRP laminates developed after modifications made in epoxy resin using a thermoplastic material, such as polycarbonate (PC) and/or acrylonitrile butadiene styrene (ABS), exhibit high thermal resistance (TR) of 77.1% compared to unmodified CFRP. In contrast, modifications made using conductive mediums like phosphonium (P), imidazolium (I), or silanized-graphene oxide (SGO) have lower TR of 25.7%, 30.5%, and 32.4%, respectively. A temperature gradient (TG) enhancement of 75% is reported for the 1.5 wt% PC/ABS modified CFRP laminates. On the contrary, modifications using 0.5 parts per hundred (phr)P, 0.5 phr I, and 1 g L⁻¹ SGO in epoxy reduce the TG by 25%, 30%, and 32%, respectively. Thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) analyses are done to explore the thermal characteristics of each case of modification. Finally, scanning electron microscopy images confirm the distribution profile of the modifiers used. Based on the types of modifications performed, the current study can offer insightful information on the thermal performances of modified CFRP laminates.

1. Introduction

Aerospace and aeronautical applications use lightweight, high-strength continuous carbon fiber (CF) materials.^[1–4] Thermosetting (TS) reinforced CF (CFRP) composites are easy to make and have great mechanical properties.^[5] CFRP composites have excellent thermal, electrical, and mechanical properties.^[6] For energy-conscious societies, CFRP composites reduce weight without compromising strength. The TS resin in CFRP is brittle under extreme temperature cycles or dynamic loading conditions, limiting its use.^[5–7] CFRP systems' low elongation after cure and brittle failure characteristics limit their use in aerospace, automotive, and wind turbine blade applications.^[8–9] Based on mechanical design, CFRP materials were in high demand earlier. However, specific applications have recently been focused on enhancing heat removal and electric discharge paths in areas like lightning strike protectors, anti/defrosting measures,^[10–12] battery thermal management units,^[13–16] and inner walls for Dewar units.

In general, depending on the modifiers used and the modification methodology chosen, CFRP composites can be made conductive or insulative. To modify CFRP composites, three main techniques are typically used: resin modification, fiber modification, and resin and/or fiber modifications.^[17–19] Direct resin modifications using a thermoplastic (TP),^[20–25] graphene,^[26,27] graphene oxide (GO),^[9,28] blend, fiber, and particle reinforcements as modifiers^[8,29] are among the most adaptable and simple to modify techniques. The diglycidyl ether of Bisphenol A (DGEBA) serves as the basis for the resin system frequently used in CFRP systems for aerospace studies. The electrical conductivity of CFRP laminates is typically improved by DGEBA modifications using ionic fluid systems like imidazolium^[30–32] and phosphonium.^[33–34] The electrical conductance of CFRP laminates has been reported to be improved by the use of particulate modifiers like GO.^[9,35] On the contrary, CFRP laminates may exhibit a more insulative nature to heat and electric current when using thermal and electrical insulators like TP, TS, and TP/TS modifiers in DGEBA.

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Table 1. Technical data sheet of materials collected.

S. No	Material	Specification
1	Carbon fiber fabric cloth	3K genuine, 220 g m ⁻² , 2 × 2 Twill Weave, Manufacturer Part No. CBC24030, Carbon black Composites, India.
2	Epoxy resin	Diglycidyl ether of bisphenol A (DGEBA) (YD 128), epoxide equivalent weight (EEW) of 185 to 194 g eq. ⁻¹
3	Hardener	Modified cyclo-aliphatic amine (TH 7301), Aditya Birla Chemicals Limited, Thailand
4	Thermoplastic-based modifiers	
	(a) Polycarbonate (PC)	Virgin quality, Makrolon 2856, Covestro AG, Germany
	(a) Acrylonitrile butadiene styrene (ABS)	Virgin quality, Cicolac MG 47F, Sabic, Saudi Arabia
5	Ionic fluid based modifier	
	(a) Imidazolium (I)	1- <i>n</i> -butyl-3-methylimidazolium chloride (C4MImCl)—Sigma Aldrich
	(a) Phosphonium (P)	Trihexyl(tetradecyl)phosphonium bis(2,4,4-trimethylpentyl) phosphinate—Sigma Aldrich.
6	Silane	3-Aminopropyltriethoxysilane (3-APTES)—Thermo Fischer Scientific
7	Graphene Oxide (GO)	Platonic Nanotech Private Limited, India

Research has been extensively carried out in determining the extent to which electric conductance in CFRP^[9,33,35] can be enhanced by modifying CFRP systems with graphene, GO and ionic fluids. However, in all these works, the effect of these modifications on the thermal conductance behavior of CFRP laminates is hardly defined. The influence of low modifier concentrations of TP, GO and Ionic fluids in the thermal conductive nature of CFRP laminates is vaguely studied and quantified. Also, the presence of higher concentrations of modifiers in epoxy increases resin viscosity^[33,36] drastically. The high viscosity after modification will restrict the DGEBA resin flow leading to unexpected errors in specimens prepared using vacuum infusion.^[8] The low modifier concentration is emphasized throughout this paper to maintain cost considerations and control the increase in viscosity of modified DGEBA. Consequently, the focus of this study is to investigate and quantify the thermal properties of CFRP laminates modified with a lower concentration of modifiers. The present work compares the effectiveness of CFRPs modified using silanized graphene oxide via electrophoretic deposition (EPD) with conventional ionic fluid and TP blend modified CFRPs.

2. Experimental Section

2.1. Materials

The materials selected for the present research work are shown in Table 1. Except for carbon fiber (CF) fabrics, all other materials were used as obtained from the seller. The CF fabric was cut into dimensions of 30 cm × 30 cm. The cut CF fabrics were then wiped with acetone solution and then treated in a hot air oven preheated to a temperature of 100 °C for 60 min.^[37]

2.2. Processing Methods

2.2.1. Modification of Epoxy Resin Using PC/ABS as Modifying System

The modification of epoxy using the PC/ABS blend was defined in our previous work.^[8] For this present work, PC/ABS blend

of 90 parts PC and 10 parts ABS were opted for modifying DGEBA epoxy resin by melt-mixing technique at 180 °C. A light-yellow coloration was observed in the DGEBA resin after melt-mixing. Case 1 of Figure 1 depicts the processing methodology used. From here on the PC/ABS (90/10) modified CFRP laminates will be termed 90/10 m-CFRP. CF fabrics initially treated with acetone with no further modification were used as reinforcement for preparing CFRPs in this case.

2.2.2. Modification of Epoxy Resin Using Ionic Fluid as Modifying System

In this system, 100 g of DGEBA resin was combined with various concentrations of I (0.5, 1.0, and 1.5 phr) and P (0.5, 1.0, and 1.5 phr). An ultrasonicator-bath (Power SONIC410) was used to sonicate the mixture for 45 min at a temperature of 40 °C and a frequency of 100 kHz after the ionic fluid had been dispersed in DGEBA resin using a magnetic stirrer at 70 °C for 45 min.^[38] Following that, the mixture was cooled to room temperature. CF fabrics initially treated with acetone with no further modification were used as reinforcements along with modified DGEBA at room temperature for fabricating CFRPs in this case. Case 2 of Figure 1 illustrates the processing method that was employed. From this point forward, CFRP laminates modified with Imidazolium (I) and Phosphonium (P) will be referred to as (I 0.5 m-CFRP, I 1.0 m-CFRP, and I 1.5 m-CFRP) and (P 0.5 m-CFRP, P 1.0 m-CFRP, P 1.5 m-CFRP) respectively.

2.2.3. Modification of CF via Electrophoretic Deposition (EPD) with Silanized-Graphene Oxide (SGO) as Modifying System

A magnetic stirrer mixed GO and deionized water at 70 °C for 1 h. After overnight filtration and drying at 90 °C, silane-functionalized graphene was obtained.^[29,39] In an EPD bath with deionized water, dried SGO was taken at concentrations of 0.5, 1, and 1.5 g L⁻¹. SGO was coated on the surface of CF fabrics by applying a current of 20 A for 30 min.^[40] The entire EPD bath was immersed in the Power SONIC410 bath at 40 °C to ensure perfect deposition, bubble-free interface, and even SGO coating

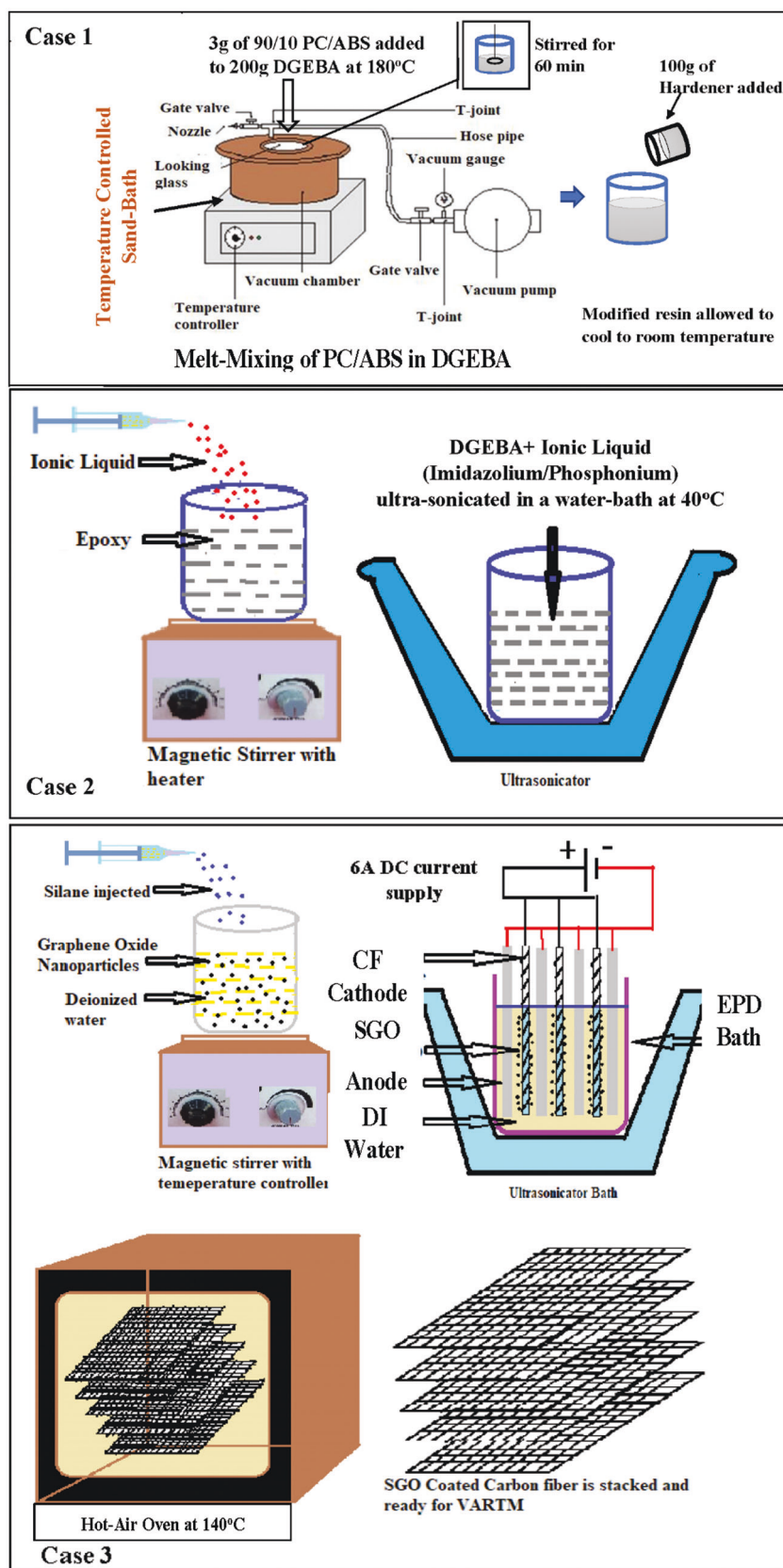


Figure 1. Case 1, Case 2, and Case 3 define the pictorial representation of processing methods defined in Sections 2.2.1, 2.2.2, and 2.2.3 respectively.

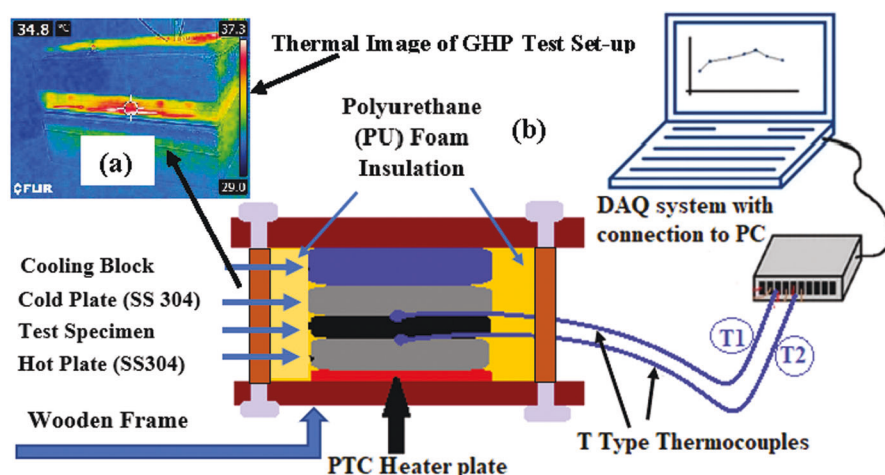


Figure 2. a) Thermal image from FLIR E4 thermal imaging camera and b) GHP Test set-up with DAQ and system unit to record interface temperature.

on CF fabrics. The SGO-coated CF fabrics were then heat-treated in a hot air oven at 140 °C for 2 h to initiate the bonding process. Case 3 of Figure 1 illustrates the CFRP processing. Then, 100 g of m-DGEBA resin from each case was mixed with 50 g of TH7301 (2:1 as per manufacturer criteria). To make CFRP laminates, the m-DGEBA/TH7301 mixture was infused into the SGO coated CF fabrics via vacuum-assisted resin transfer molding (VARTM). In further discussions, SGO modified CFRP laminates will be referred to as G 0.5 m-CFRP, G 1.0 m-CFRP, and G 1.5 m-CFRP.

2.2.4. Fabrication of CFRP Laminates

For carbon fabric, there is a relation existing between the thickness and weight of the fabric as mentioned in Equation (1)

$$\text{Consolidated thickness (in mm)} = \text{Weight in } [(g/m^2)/1000] \text{ mm} \quad (1)$$

where weight of the carbon fabric = 220 g m⁻² (from data sheet); consolidated thickness for one layer of fiber = 220/1000 = 0.22 mm; number of sheets of carbon fiber used = 9 numbers; thickness of 9 layers of carbon fiber sheets = 9 × 0.22 = 1.98 mm; approximate/total thickness of the composite casted using VARTM = 2.20 mm; number of epoxy layers = 10 layers; thickness of 10 layers of epoxy layers is determined as per the following method:

Approximate/total thickness of composite (CFRP laminate) – Thickness of 9 layers of carbon fiber sheets = 2.2 – 1.98 = 0.22 mm; and

$$\begin{aligned} &\text{Thickness of one layer of epoxy} \\ &= \frac{\text{Thickness of 10 layers of epoxy layers}}{\text{Number of epoxy layers}} \\ &= \frac{0.22}{10} = 0.022 \text{ mm} \end{aligned} \quad (2)$$

The prepared CFRP laminates using VARTM^[4,8,37] were polished to reach an average thickness of 2 mm, and circular speci-

mens of 50 mm diameter were cut for thermal performance analysis using the test set-up as shown in Figure 2b.

2.3. Evaluation of Thermal Synergistic Effect Using GHP Experiment

The through-thickness thermal conductivities of CFRP laminates were determined using the guarded hot plate (GHP) method by assuming a steady-state technique. The thermal conductivity could be determined by just knowing the test sample thickness and interface resistance between the CFRP laminate (sample) and the stainless steel (SS) 304 (substrate).^[9,41] A steady-state condition with only 0.5 °C difference in temperature^[9] was observed for the GHP experiment. The neat and modified CFRP laminates were tested in a thermally insulated chamber with a positive temperature coefficient (PTC) heating element as its base. The laminate specimens were prepared in disc shape with a diameter of 50 mm and thickness of 2 mm. The chamber was designed to achieve minimum heat loss from the test chamber, confirmed by the thermal images obtained from FLIR E4 thermal imaging camera, as shown in Figure 2a) The customized test set-up comprises T-type thermocouples, OMEGA HFS-4 heat flux sensor (sensitivity = 1.6 μV (W⁻¹ m⁻²)) and a data acquisition unit (DAQ) connected to the system unit as shown in Figure 2b. The T-type thermocouples were calibrated using a constant temperature water bath.

The test set-up was calibrated using SS 304 and cured DGEBA disc-shaped specimens whose thermal conductivity values were well defined.^[30,36,42] It was observed that the PTC heating unit can provide a constant heat source for which the calculated thermal conductivities of SS304 and polytetrafluoroethylene (PTFE) at 100 °C were 16.2 ± 0.04 W m⁻¹ K⁻¹ and 0.25 ± 0.005 W m⁻¹ K⁻¹ respectively. The observed thermal conductivity values were in good agreement with the values stated by the suppliers and with those cited in previous works^[43–45] as well. The effect of modifications on the morphology was determined from field emission scanning electron microscopy (FE-SEM) images. The effect of modifications on other thermal parameters of CFRP specimens was determined using differential scanning calorimetry (DSC)

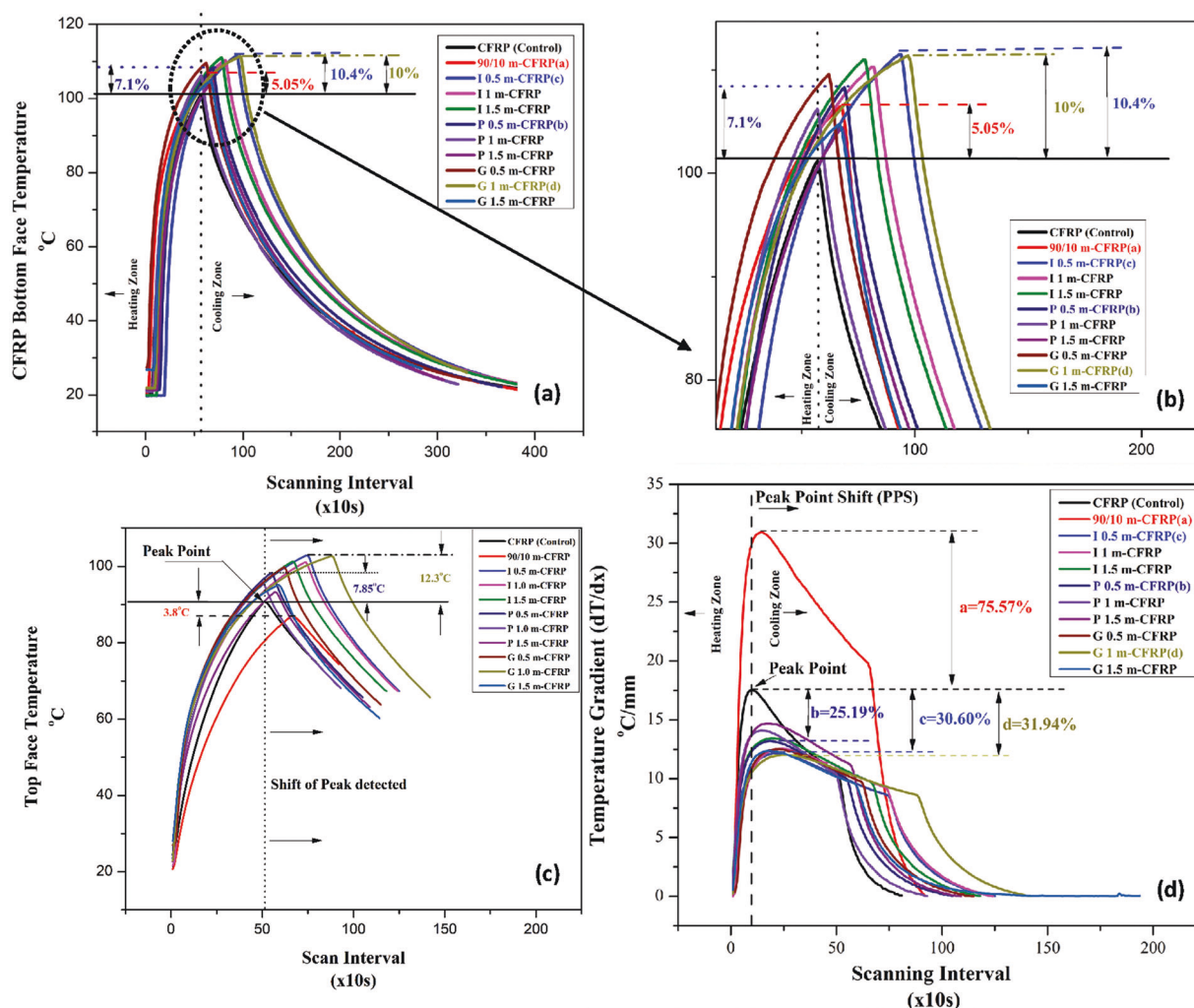


Figure 3. GHP experiment data showing a) bottom face temperature, b) zoomed portion of peak temperatures from bottom face, c) top face temperature, and d) dT/dx – time curve for various CFRP laminates.

(DSC SETLINE- SETARAM) between -30 to 200 °C at a heating rate of 5 °C min^{-1} and thermogravimetric analysis (TGA) (TGA Labsys Evo instrument-SETARAM) between 30 °C to 600 °C at a heating rate of 10 °C min^{-1} . The thermal conductivities of the modified matrix were determined using the KD2Pro Thermal properties meter (SL368).

3. Results and Discussion

3.1. Experimental Determination and Analytical Validation of Through-Thickness Thermal Conductivity of CFRP Laminates Using Lewis and Nielsen's Equation

The equation for predicting the through-thickness thermal conductivity of composites (K_p)^[46] is shown in Equation (3)

$$K_p = K_m + \frac{2(1 - V_m)(K_{FR} - K_m)K_m}{V_m(K_{FR} - K_m) + K_m} \quad (3)$$

where K_m is the thermal conductivity of the matrix, V_m is the volume fraction of matrix, and K_{FR} is the thermal conductivity of the fiber. The K_p value of neat CFRP was obtained as 0.7403 $\text{W m}^{-1} \text{K}^{-1}$ by substituting the values $V_m = 0.42$ (experimentally determined), $K_m = 0.24$ $\text{W m}^{-1} \text{K}^{-1}$ (measured using KD 2Pro) and confirmed from previous works,^[42] $K_{FR} = 2$ $\text{W m}^{-1} \text{K}^{-1}$ at 100 °C.^[41] A PTC heat source was employed to carry out the GHP experiment. The heat flux was measured using a heat flux sensor, which was then used to determine the thermal conductivities of laminates. The interface and peak temperatures recorded during the GHP experiment are shown in Figure 3. According to the Fourier Law for heat transfer under steady state condition we have

$$q'' = K_{\text{exp}} \frac{dT}{dx} \quad (4)$$

where q'' is the heat flux (W m^{-2}), K_{exp} is the experimental thermal conductivity of the material in $\text{W m}^{-1} \text{K}^{-1}$ and dT/dx is the temperature gradient (°C mm^{-1}). Thus, by substituting the value

Table 2. Interface temperature data and thermal conductivity obtained from the GHP experiment.

Case studies		Experimental data			% Error	
Sl. No.	Specimen	Bottom face peak temperature [°C]	Top face peak temperature [°C]	Peak dT/dx [°C/mm]	Thermal conductivity, K_p [W m ⁻¹ K ⁻¹]	% Change ($1 - \frac{K_{exp}}{K_p}$) 100
1	Unmodified	101.4	90.78	17.84	0.74	2.7
2	90/10 m-CFRP	106.7	87.0	31.16	0.42	2.3
3	I 0.5 m-CFRP	112.0	103.1	12.35	1.07	1.8
4	I 1.0 m-CFRP	110.8	101.3	12.55	1.05	1.9
5	I 1.5 m-CFRP	111.4	101.5	13.46	0.98	2.0
6	P 0.5 m-CFRP	108.4	81.8	13.23	1.0	2
7	P 1.0 m-CFRP	106.50	95.8	14.12	0.93	2.8
8	P 1.5 m-CFRP	104.4	93.4	14.70	0.89	2.2
9	G 0.5 m-CFRP	109.5	99.9	12.56	1.05	1.9
10	G 1.0 m-CFRP	111.4	102.8	11.94	1.11	1.8
11	G 1.5 m-CFRP	104.7	95.2	12.40	1.06	1.8

Table 3. Thermal properties of unmodified and modified CFRP laminates.

Case studies		Derived thermal properties				
S. No.	Specimens	Thermal resistance [K W ⁻¹] at 100 °C	Specific heat capacity ($C_{p,c}$) at 100 °C [J g ⁻¹ K ⁻¹]	Thermal conductivity, K_{exp} at 100 °C [W m ⁻¹ K ⁻¹]	Thermal conductivity, K_m at 100 °C [W m ⁻¹ K ⁻¹]	Thermal diffusivity (α_{dc}) at 100 °C [cm ² s ⁻¹]
1	Unmodified	1.05	1.9	0.76 ± 0.02	0.24	3.33
2	90/10 m-CFRP	1.86	2.14	0.43 ± 0.01	0.14	1.68
3	I 0.5 m-CFRP	0.73	2.17	1.09 ± 0.007	0.39	4.18
4	I 1.0 m-CFRP	0.75	–	1.07 ± 0.013	–	–
5	I 1.5 m-CFRP	0.8	–	1.00 ± 0.021	–	–
6	P 0.5 m-CFRP	0.78	2.13	1.02 ± 0.03	0.35	3.97
7	P 1.0 m-CFRP	0.83	–	0.96 ± 0.015	–	–
8	P 1.5 m-CFRP	0.87	–	0.92 ± 0.024	–	–
9	G 0.5 m-CFRP	0.74	–	1.07 ± 0.017	–	–
10	G 1.0 m-CFRP	0.71	1.63	1.13 ± 0.009	0.41	5.77
11	G 1.5 m-CFRP	0.74	–	1.08 ± 0.011	–	–

of q'' and dT/dx in Equation (4), the K_{exp} value can be obtained. Table 2 displays the calculated K_p values for all modified CFRP laminates. All CFRP laminate specimens have the same constant specimen thickness, which is 2 mm on average. Table 2 shows a variation in K_{exp} with respect to K_p of 1.8% to 2.8%, which was well within the reported variation of 2% to 4%, as previously stated by Zhang et al.^[46]

Table 3 displays K_{exp} along with additional thermal characteristics from the GHP experiment, such as thermal diffusivity and thermal resistance. The Lewis and Nielsen's formula, which Simon et al. mention in their work,^[41,47–49] was used to validate it. According to previous works,^[47] the deviation of K_{exp} from Lewis and Nielsen's formula with analytically determined thermal conductivity is shown in Figure 4. Similar to what Kim et al.^[50] stated, the CFRP laminates used in the current research work consist of 58% fiber (fillers) and 42% matrix (epoxy/modified epoxy resin). Lewis and Nielsen's formula is used to calculate the thermal con-

ductivity of CFRP composite laminates using the matrix thermal conductivities (K_m) obtained from KD2Pro equipment.^[51]

As demonstrated in our previous work,^[4,8] uniform dispersion occurs when PC and ABS were melt mixed in DGEBA resin. The excess resistance to heat flow along the through-thickness of CFRP laminates caused by the presence of uniformly dispersed phase of PC/ABS in DGEBA was confirmed by the drop in thermal diffusivity (49.54%) and increase in thermal resistance (77.1%) values as shown in Table 3. Furthermore, the formation of a heterogeneous morphology of PC/ABS phase over DGEBA, as shown in Figure 5a, resulted in enhanced thermal resistance of 90/10 modified CFRP laminates. As shown in Table 2, a trend of increased thermal conductivity was seen when DGEBA was modified using ionic fluids and SGO, with a maximum improvement of 48.7% for the SGO 1.0 g L⁻¹ modified CFRP system. The thermal conductivity exhibits a declining trend as the ionic fluid concentration increases.^[33,36] Thermal conductivity initially

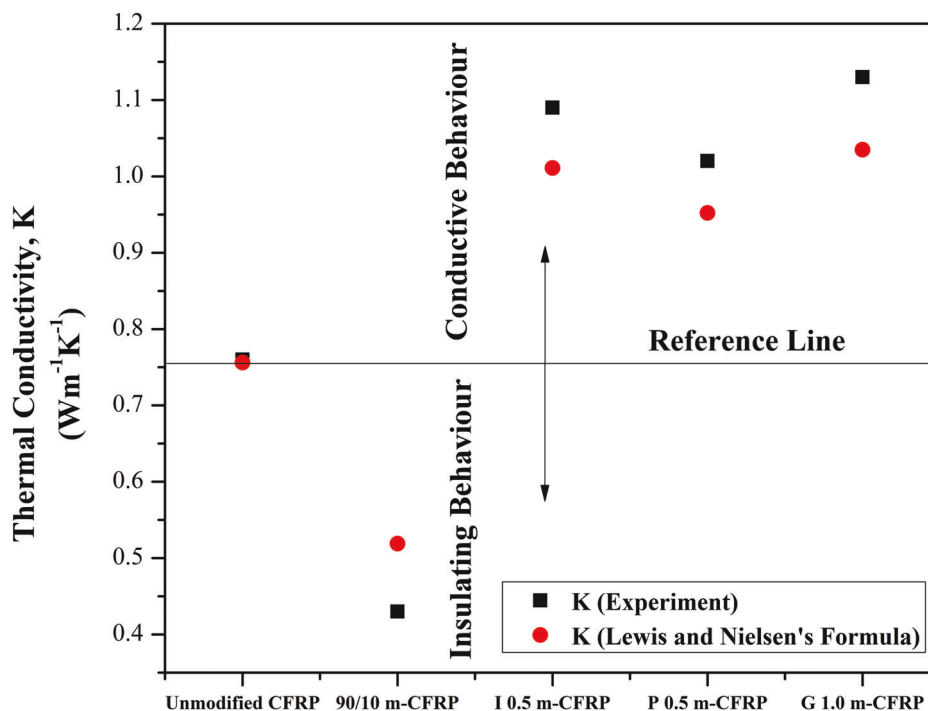


Figure 4. Experimental thermal conductivity data validation using Lewis and Nielsen's formula.

increases as a result of the SGO modification, up to 1.0 g L^{-1} of SGO concentration. At 1.5 g L^{-1} , the SGO has an agglomeration nature, which could explain the decline in thermal conductivity with an increase in concentration.^[9,18,28,35] SEM micrographs confirm the tendency of SGO to agglomerate at 1.5 g L^{-1} concentration, as shown in Figure 5b. The experimental results made it clear that 1.0 g L^{-1} SGO gives the CFRP samples a superior conductive quality over other CFRPs modified using ionic liquids. The case of SGO 1.0 g L^{-1} modified CFRP showed the highest value of thermal conductivity, and after that, it began to decline but was still more conductive than unmodified CFRPs.

The enhanced conductive nature of modified CFRP can be substantiated by the fact that graphene oxide nanoparticles inherently possess good thermal characteristics. Also, SGO has successfully formed a strong bond between the fiber and epoxy, providing a smooth passage for thermal energy transfer.^[29] Moreover, SGO forms a flexible —O—Si—O— bond in the CFRP system, which is a means of vibrational transfer of thermal momentum, consequently resulting in high thermal conductivity.^[52] However, in contrast to this trend, there was a drop in conductivity for the case of G 1.5 m-CFRP, which can be corroborated by the thermal hindrances formed at fiber epoxy interphases due to SGO agglomeration, which was evident in the SEM analysis as shown in Figure 5b.^[53]

3.2. Effect of Modifiers in the Variation of Specific Heat Capacity of Modified CFRP Laminates

When compared to unmodified CFRP, the thermal properties of 90/10 m-CFRP, I 0.5 m-CFRP, P 0.5 m-CFRP, and G 1.0 m-CFRP were all excellent (Table 2). Therefore, unmodified CFRP,

90/10 m-CFRP, I 0.5 m-CFRP, P 0.5 m-CFRP, and G 1.0 m-CFRP were selected for a more in-depth analysis. To further establish the improved thermal properties, it was necessary to determine the effect of modifications on the specific heat capacity (C_p) of CFRP laminates. According to Figure 6, C_p for 90/10 m-CFRP, I 0.5 m-CFRP, and P 0.5 m-CFRP, respectively, increased by 12.63%, 14.21%, and 12.1% at 100°C compared to unmodified CFRP. In G1.0 m-CFRP, the C_p value at 100°C exhibits a 14.21% reduction compared to unmodified CFRP. Graphene oxide, by its nature, has a lower C_p value than other ionic and plasticizer modifiers. This lower specific capacity is transferred to the CFRP system and lowers the overall C_p value. Also, this might result from the enhanced flexibility of bonds in DGEBA due to silane modification. As a result, lower heat energy resulted in the sudden temperature rise, confirming lower C_p values than unmodified CFRP laminates.^[53] Additionally, in the case of I 0.5 and P 0.5 modified CFRP laminates, the cross-linking density of DGEBA shows a reduction in the stiffness of bonds.^[30–33,38,54]

The bonds absorb more vibrational energy before converting it to thermal energy as a result of the reduction in bond stiffness. The increased C_p values in comparison to unmodified CFRP may be due to the decrease in bond stiffness. As seen in Figure 7, the TGA further confirmed the change in thermal stability caused by the rise in C_p . A heterogeneous phase of PC/ABS was formed over the DGEBA matrix in the case of 90/10 m-CFRP, as demonstrated in SEM micrographs (Figure 5(a)). The additional thermoplastic phase acts as an insulating layer, resulting in excess thermal energy accumulation, leading to higher C_p values than unmodified CFRP. The synergism between PC/ABS/DGEBA causes the rise in C_p value, as confirmed in our earlier publications.^[8]

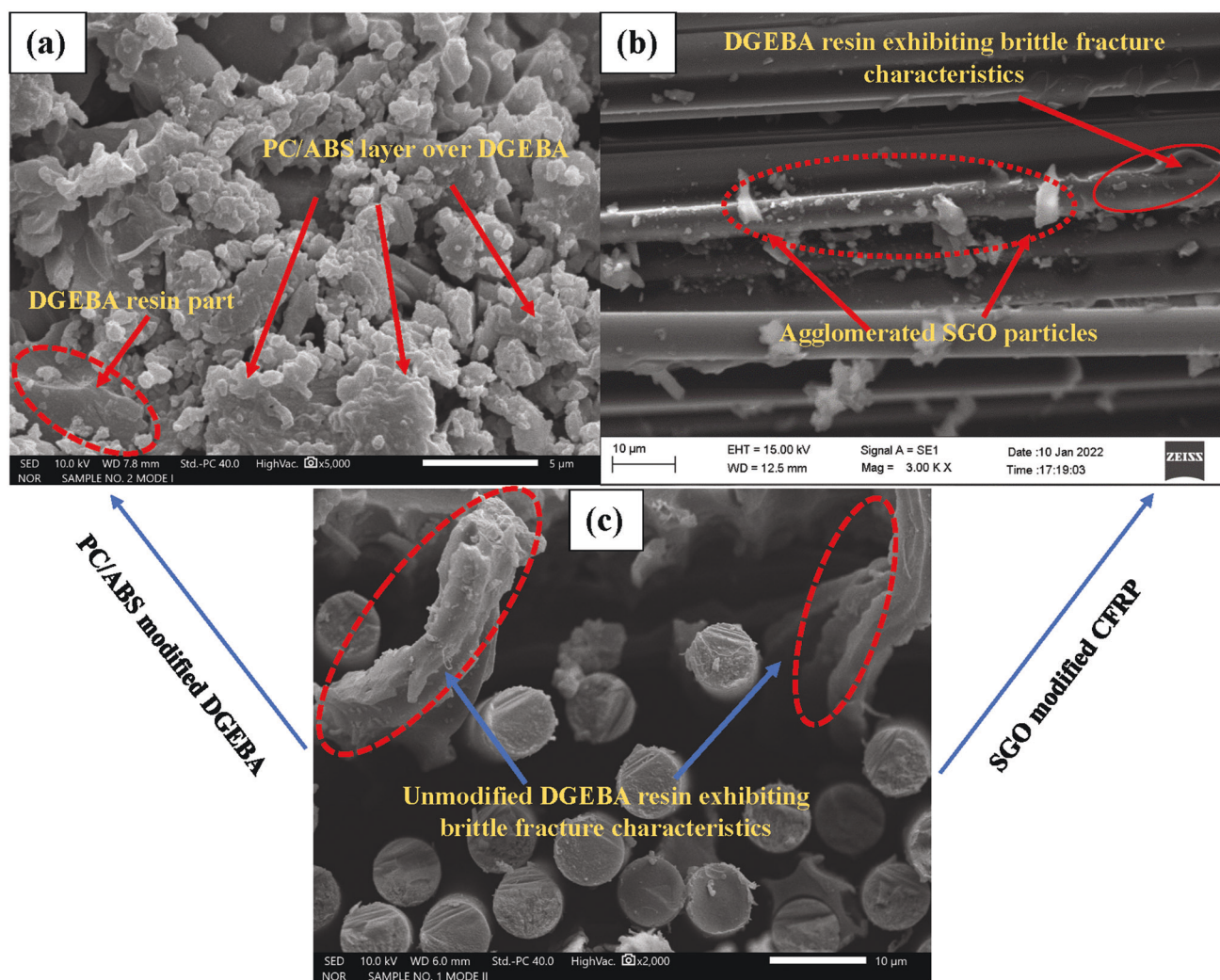


Figure 5. Comparison of morphologies a) 90/10 m-CFRP, b) G 1.5 m-CFRP, and c) unmodified CFRP laminates.

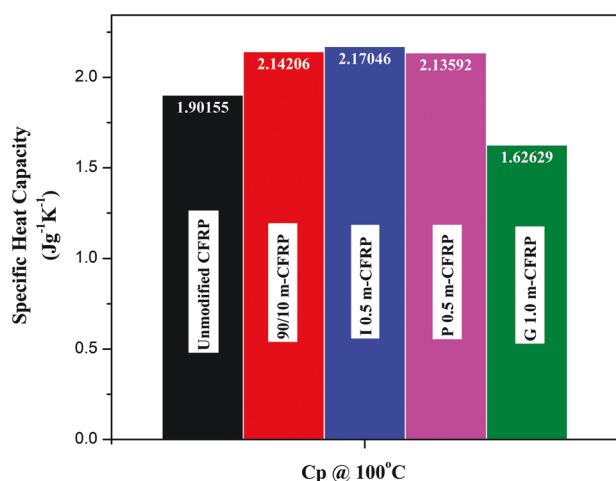


Figure 6. Specific heat capacity of CFRP unmodified compared with various modified CFRP laminates obtained from DSC.

3.3. Quantitative Determination of the Extent of Insulative/Conductive Behavior of Modified CFRP Laminates

As was covered in earlier sections, modifications to CFRP tend to change the thermal conductivity and specific heat capacity of laminates. The use of PC/ABS (90/10) tends to decrease the thermal conductivity of CFRP laminates by 43.4% and increase their specific heat capacity by 12.6%. However, the CFRP laminates showed an increase in thermal conductivity and C_p values when modifications involving ionic fluids were made.

The I 0.5 modification exhibited an increase in C_p and thermal conductivity of 43.4% and 14.21%, respectively. Thermal conductivity and C_p were improved by 34.2% and 12.1%, respectively, as a result of the P 0.5 modification. According to the aforementioned data, the I 0.5 modification outperforms the P 0.5 modification in terms of performance and can guarantee improved overall thermal performance of modified CFRP laminates over unmodified CFRP laminates. The thermal diffusivity of CFRP laminates tends to alter after modifications, as shown in Figure 8. Only in the 90/10 modified CFRP laminates did the thermal

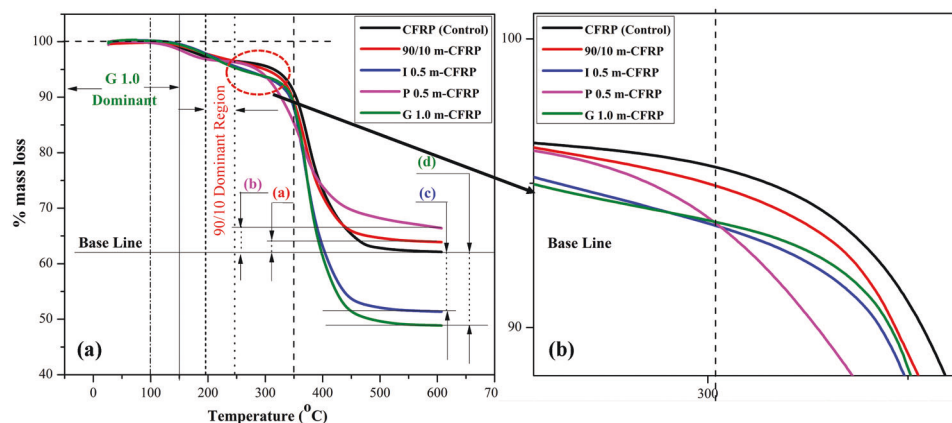


Figure 7. a) TGA overlay of unmodified, 90/10 m-CFRP, I 0.5 m-CFRP, P 0.5 m-CFRP, and G 1.0 m-CFRP laminates and b) zoomed portion of TGA curve at 300 °C.

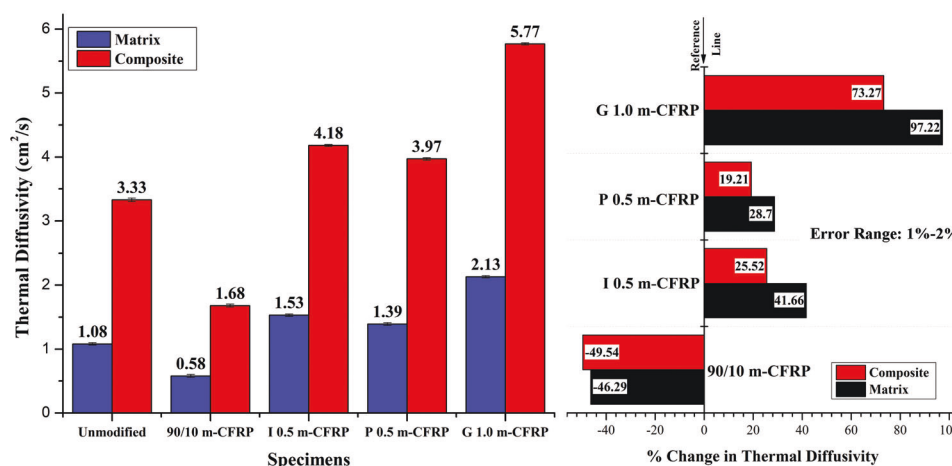


Figure 8. Experimental data a) change in thermal diffusivity values calculated from GHP experiment and b) % change in thermal diffusivity compared with unmodified CFRP laminates.

diffusivity value decrease, whereas all other modifications tended to make CFRP laminates more thermally diffusive. When compared to the I 0.5 and P 0.5 modifications, the G 1.0 modifications tend to show the greatest increase in thermal diffusivity. Thermal conductivity has increased in CFRP as a result of the anomalous behavior of the G 1.0 modifications, in contrast to a decrease in the C_p value. The result is that G 1.0 m-CFRP has a $5.77 \text{ cm}^2 \text{ s}^{-1}$ thermal diffusivity value, which is 73.2% higher than unmodified. Additionally, I 0.5 and P 0.5 m-CFRP laminates are outperformed by G 1.0 m-CFRP by 38% and 45.3%, respectively. This study therefore, confirms the effectiveness of SGO modifiers over other conductive and insulative type modifiers in CFRPS for heat dissipation applications.

3.4. Synergism Effect of Modifiers/DGEBA in Thermal Stability Characteristics of Modified CFRP Laminates

Figure 7a compares the thermal degradation of 90/10, I 0.5, P 0.5, and G 1.0 modified CFRP laminates to unmodified CFRP.

Based on the data, it was found that modified CFRP systems with 90/10 ($a = 3.5\%$) and P 0.5 ($b = 7.5\%$) have better mass retention properties than unmodified CFRP systems. However, as shown in Figure 7a, I 0.5 ($c = 16.7\%$) and G 1.0 ($d = 21.6\%$) modified CFRP laminates tend to show higher mass reduction at 600 °C than unmodified CFRP systems.^[51] The silane content in SGO particles is responsible for the greater weight loss observed in the case of the G 1.0 m-CFRP system.^[39] For unmodified, 90/10 modified, and P 0.5 modified CFRP laminates, a mass loss of 3.5% at 250 °C was noted. I 0.5 and G 1.0 modified CFRP laminates, in contrast, experienced a mass loss of 4.39% and 4.69%, respectively, at 250 °C. At 150 °C, P 0.5 modified CFRP and unmodified CFRP systems exhibit at most 0.75% mass loss whereas, I 0.5, G 1.0, and 90/10 modified CFRP systems exhibit almost no mass loss.^[30] The thermal stability of DGEBA resins tends to increase with the presence of heterogeneous thermoplastic phase with higher T_g values, which increased the thermal stability of CFRP laminates at 150 °C.^[24] It is interesting to note that after 147 °C (T_g of PC), 90/10 modified CFRP systems perform better than unmodified and P 0.5 modified CFRP systems, and

overtake I 0.5 and G 1.0 modified CFRP systems at 200°C. The 90/10 modification is the one that performs the best among all the others after 200°C and maintains the trend until 250°C. According to Figure 7b, unmodified and 90/10 modified CFRP systems both show a mass loss of just 5% at 300°C, while I 0.5, P 0.5, and G 1.0 modified CFRP systems show a mass loss of 6.4%.^[31] I 0.5, P 0.5, and G 1.0 modifications have a tendency to reduce the bond network and make the crosslinking flexible, which causes a slight increase in mass loss. DGEBA crosslinked networks increased flexibility causes a slow transfer of heat energy, which results in localized heating effects resulting in this enhanced mass loss. After 300°C, this characteristic becomes dominant. It is worthwhile to mention that 90/10, I 0.5 and G 1.0 modified CFRP laminates all perform equally well, whereas P 0.5 modified CFRP laminate was the least performing till 196°C.^[55] However, P 0.5 modified CFRP laminates tend to retain maximum mass even after 600°C. The high mass retention tendency of P 0.5 modified CFRP might be due to the lower thermal diffusivity values of CFRP laminates resulting after P 0.5 modification compared to I 0.5 and G 1.0 modifications, as shown in Table 3.

3.5. Analytical Confirmation of the Effectiveness of Modified CFRP Laminates as Potential Materials for Specific Applications

In the present study, it was found that adding 1.5 wt% of PC/ABS (90/10) to DGEBA resin can reduce CFRP laminates' thermal conductivity by 43.4%, enhancing the material's competency for use as the inner wall of cryogenic Dewar vessels. When comparing 90/10 modified CFRP laminates to unmodified CFRP laminates, performance evaluation using the analytical methods described in^[56] demonstrated that a reduction in CF layers could be achieved. Using 90/10 modified CFRP laminates (18 CF layers) results in a reduction of 2 CF layers with the added benefit of improved thermal insulation properties compared to unmodified CFRP laminates (20 CF layers). Thus, compared to unmodified CFRP systems, the reduced CF layers can result in a sizable cost reduction. While earlier studies^[35] suggested that increasing the thermal conductivity of CFRP laminates by adding SGO particles can enhance their use in the design of thermal management systems. Analytical results showed that 1 mm thick G1.0 modified CFRP laminates could remove the same amount of heat as 1.5 mm thick unmodified CFRP laminates. Thus, using G 1.0 m-CFRP can ensure an optimal operating temperature range and reduce the heating of battery compartments. Therefore, compared to other traditionally used materials, effective thermal management can be achieved with the least amount of additional load. In future scope, incorporating expensive high conductive nanomaterials in CFRP laminates using the EPD technique can improve the thermal conductivities to a range of 20 W m⁻¹ K⁻¹, which would extend its application in defense technology. Please refer Supporting Information.

4. Conclusion

The results of the GHP experiment confirm that the I 0.5 modification and the G 1.0 modification are the most effective ways to improve the thermal conductivity of CFRP laminates. To improve the thermal properties of a material for use in conducting

applications, the G 1.0 modification is the way to go. Tests using thermogravimetric analysis (TGA) show that I 0.5 and G 1.0 modified CFRP perform better than P 0.5 modified CFRP in thermal conductive applications over a temperature range of 100 to 150°C. In general, 90/10 modified CFRP performs better than all other modifications and unmodified CFRP over the temperature range of 200 to 250°C. When compared to unmodified CFRP, 90/10 modified CFRP performs admirably over a temperature range of 100 to 200°C, with a maximum weight loss percentage of just 2.08%, making it ideal for applications requiring high thermal insulation properties. This paper is expected to guide the manufacturers in selecting the best suitable limited quantity modifier in CFRPs for thermal insulative/conductive specific applications.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

CFRP laminates, synergism, temperature gradient, thermal conductivity, thermal stability

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