

Study on the heat transfer reinforcement of glass fiber/epoxy resin composites by grafting and dispersing graphene oxide

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A B S T R A C T

The grafted-graphene oxide/glass fiber/epoxy resin (grafted-GO/GF/EP) and dispersed-graphene oxide/glass fiber/epoxy resin (dispersed-GO/GF/EP) composites were successfully prepared by grafting graphene oxide (GO) on glass fiber (GF) with coupling agent and random dispersion of GO. A scanning electron microscope (SEM) was employed to characterize the micro structure. The heat transfer RVE models of the grafted-GO/GF/EP and dispersed-GO/GF/EP composites were established respectively. Based on RVE heat transfer simulation model, the heat transfer characteristics of the grafted-GO/GF/EP and dispersed-GO/GF/EP composites were analyzed. The results show that under the condition of 0.1 vol% GO content and heated for 60 s, the thermal conductivities of dispersed-GO/GF/EP and grafted-GO/GF/EP composites are increased by 3.48% and 82.37% respectively. The dispersion of GO increases the heat transfer paths in EP and forms a connected GO-GO heat transfer network. However, with the increasing of GO content, the agglomeration of GO becomes more obvious. The dispersion and interfacial bonding of the GO/GF/EP composites were improved by coupling agent, which effectively reduced the interfacial thermal resistance and significantly improved the thermal conductivity of the composites.

1. Introduction

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Polymer composites have been widely used in aerospace, military, automobile and medical devices due to their high specific stiffness and strength, large damping, corrosion resistance and low thermal expansion performance [1-4]. Among polymers, epoxy resin (EP) is considered as one of the most important thermoset polymer for the outstanding characteristics such as good mechanical properties, enhanced thermal stability, good chemical resistance and fatigue resistance [5,6]. However, due to the low thermal conductivity of EP (the thermal conductivity is $\sim 0.2 \text{ W/ m} \cdot \text{K}$), the applications in many fields such as heat conduction materials, heat-resistant materials and high heat flux density electronic devices are greatly restricted. Therefore, adding high heat conducting filler into polymer is one of the most effective methods to improve the thermal conductivity of EP. Various nano materials such as nano silica [7], carbon nanotubes [8,9], graphene and graphene oxide [10-13] are introduced to improve the thermal conductivity of polymers. Compared with conventional materials, dispersed polymer composites have better mechanical, thermal and electrical properties, low density and high strength [14]. Graphene with perfect structure has almost the highest thermal conductivity ($\sim 5000 \text{ W/ m} \cdot \text{K}$) [15]. Combined with excellent 2D morphology and high surface area, graphene can significantly improve the physical and mechanical properties of polymer with very low filling amount, which has a wide range of potential applications in polymer matrix and the development of high-performance polymer composites [16,17].

As the main derivative of graphene, graphene oxide (GO) is the most easily obtained precursor of graphene. Due to the polar sheets of sp^2 hybrid carbon atoms and sp^3 hybrid carbon atoms are closely arranged into a two-dimensional single-layer, GO has extraordinary mechanical properties and superior thermal conductivity, which can form strong physical interaction with polymer, and disperse stably in water and polar organic solvents [18,19]. Zhang et al. [20] modified carbon fiber (CF) by grafting polyhedral oligosilicate (POSS) onto the CF surface using GO as the linkage. The results showed that the presence of GO linkage and POSS could significantly enhance both the area and wettability of fiber surface, leading to an increase in the interfacial strength between fibers and resin. Jing et al. [21] compared pure glass fiber, silane coupling agent modified glass fiber (GF-S) and graphene oxide coated glass fiber (GF-GO) into poly lactic acid (PLA) respectively. It was found that the excellent nucleation ability of GF-GO led to the crystallization of GF-GO interface in the isothermal crystallization process of PLA, which enhanced the thermodynamic properties of PLA/GF-GO composites. A liquid crystalline epoxy resin (LCE) was synthesized by chemical grafting of GO sheet and γ -ethoxymethoxysilane (KH560) by Qi et al. [22]. This material has higher thermal stability and glass transition temperature than pure epoxy resin. The above results show that GO is a promising nanomaterial filler to improve the thermal conductivity of polymer composites.

However, the type of filler is not the only factor affecting the thermal enhancement of polymer composites. The lack of interfacial bonding between filler particles and matrix is one of the main problems that hinder the improvement of thermal conductivity of polymer composites [17,23]. Improving the interface interaction can effectively reduce the interfacial thermal resistance and improve the thermal conductivity of polymer composites. In addition, due to the special two-dimensional structure, high surface energy and the inherent Van der Waals interaction, the GO layers are prone to gather in the polymer matrix, thus losing the nano effect [24,25]. In order to find an effective method to enhance the dispersion and interfacial bonding of GO in polymer matrix, various dispersion and preparation processes have been widely investigated. A research result from Aradhana et al. showed that the strengthening effect is closely related to the dispersion state of GO [26]. The agglomeration of GO fillers makes the properties of the composites far less than expected, even negative effects. Moreover, the aggregation of GO lamellae in the polymer matrix will lead to serious defects, thus reducing the overall performance of the composite [27]. Huang et al. [28] used ultrasonic technology to disperse GO and coated boron nitride with GO (h-BN@GO). The compatibility of h-BN with epoxy resin was improved and the interfacial thermal resistance between h-BN and EP was reduced by employing hybrid agent.

Obviously, the effect of heat transfer enhancement of GO dispersion in EP has some limitations. Especially when the doping content is high, the huge surface area of GO makes it easy to aggregate. Thus, the grafting of GO on the surface of the substrate can further enhance the heat transfer. The common grafting methods include electrophoretic deposition (EPD) [29,30], coupling agent [31], and dip coating process [32]. These treatment methods introduce some functional groups, perforations and deep ridges on the fiber surface, which changing the roughness and promoting the adhesion to the polymer. However, the above methods are also subject to many limitations, such as high temperature, expensive catalyst and pollution, and reducing the mechanical properties of glass fiber [33]. By contrast, coupling agent treatment can not only protect the fiber from abrasion, but also provide a good interface for the bonding between filler particles and polymer matrix. Therefore, silane coupling agents are widely used in the surface modification of GO. The GO sheets were covalently grafted onto the surface of glass fibers (GFs) by Chen et al. via amidation [34]. The maximum interlaminar shear strength (ILSS) value of GO grafted on GFs (GO-g-GF1.5) obtained by this method was 36.23 MPa, which is significantly higher than that of pure glass fiber (25.63 MPa) and GO coated on GFs (GO-c-GFs) (29.76 MPa) by electrostatic assembly strategy. In conclusion, considering the abundant functional groups on the surface of GO, grafting is a feasible method to improve the dispersion and interfacial adhesion.

In this work, GO was introduced into EP matrix by random dispersion and coupling agent grafting on the surface of GF, thus the dispersed-GO/GF/EP and grafted-GO/GF/EP composites were prepared respectively. The effects of GO interface interaction and dispersion on the heat transfer characteristics of GO/GF/EP composites were studied. In addition, through the microscopic characterization of dispersed-GO/GF/EP and grafted-GO/GF/EP samples, representative volume element (RVE) heat transfer mathematical models were established. The heat transfer characteristics of the samples prepared by dispersing and grafting methods were analyzed by numerical simulation. The heat transfer path and enhancement effect were compared and analyzed. The results of this study guide the preparation process selection of GO modified GF/EP composites, and reveal the mechanism of GO on the heat transfer characteristics of GO/GF/EP composites in different doping states from the perspective of heat transfer enhancement mechanism.

2. Experimental

2.1. Materials

GO was provided by Suzhou Hengqiu Graphene Technology Co., Ltd., with a lamellar diameter of 0.2–10 μm , layer number of 1–20, the oxygen content >50%, and a purity of ~98%. Glass fiber was G15000 fabric provided by Shandong Guangwei composite material Co., Ltd., with surface density of 150 g/m^2 . Bisphenol-A diglycidyl ether (E 51) was purchased from Shanghai Autun Chemical Technology Co., Ltd. Curing agent 4,4'-Methylenedianiline (DDM) was obtained from Shanghai Zhanyun Chemical Co., Ltd. Coupling agent 3-aminopropyl triethoxy silane (KH-550) was provided by Nanjing Chuangshi Chemical Additives Co., Ltd. All materials were used without further modification.

2.2. Methods

2.2.1. Synthesis of GO-grafted GF

A two-step method was employed for grafting GO on GF (as shown in Fig. 1). GO suspension in ethanol solution was prepared by high-speed stirring and ultrasonic vibration. Then the dispersed GO was attached to the surface of GF by physical adsorption. After drying, GO adsorbed on the surface of GF was chemically grafted in the form of covalent bond through the action of coupling agent (KH-550), which effectively avoided the agglomeration of GO caused by the direct mixing with KH550. The part of the coupling agent group is inorganic group, which can

interact with the reinforcement fiber. Thus the interfacial interaction between inorganic and organic materials is effectively improved, which is conducive to the improvement of comprehensive thermal conductivity of the composites.

Firstly, GF was placed in the resistance furnace at 400 °C for 30min to remove the sizing agent. Then put the cooled GF into acetone for immersion cleaning. Soak the cleaned GF in NaOH solution for 40 min. Alkali treatment can hydroxyl the surface of GF, which is beneficial to grafting. After soaking, the treated GF was washed with deionized water for three times and dried. The GO ethanol solution with mass fraction of 75% was prepared, and stirred in a high-speed mixer for 1500 r/min for 1h, and then ultrasonic treatment (power of 100W, and the oscillation frequency of 40 kHz) for 1h to obtain uniformly-dispersed GO suspension. After that, the GF was immersed in the GO suspension for 15 min, and then took it out for drying. The deionized water and anhydrous ethanol were mixed into 100 ml solution at the ratio of 7:2. KH-550 solution was obtained by adding KH-550 at 1 vol%. Finally, the dried GF and KH550 solution were mixed in acidic solution for 30 min until GO was grafted onto GF via silane coupling reaction. Then the GF was taken out and dried again for standby. The matrix volume fraction used was ~72.35%.

2.2.2. Preparation of grafted-GO/GF/EP

The curing agent DDM was added into EP according to the ratio of EP: DDM 4.3:1, and stirred for about 10 min until there were no bubbles. After GO grafting treatment, the GF was put into the mold and immersed in the EP. Then the mold was put into the blast oven and cured at 80 °C for 2.5h, and heated to 150 °C for 2h. The process of the experiment is shown in Fig. 2.

2.2.3. Preparation of dispersed-GO/GF/EP

The curing agent DDM was added into EP according to the ratio of epoxy resin: curing agent = 4.3:1, and stirred evenly. The GO/EP GO/GF/EP and dispersed-GO/GF/EP composites. Before SEM observation, liquid nitrogen was used for quick freezing, and then brittle fracture was made under mechanical action to obtain ideal cross-section morphology. The morphologies of samples were observed at a working voltage of 1.5–3 KV.

Fig. 4 (a) shows that there are obvious small protrusions on the surface of GF, which indicates that GO has been successfully grafted onto the surface of GF after modification. From Fig. 4 (b), it can be more clearly seen that the surface of GF-GO is wrinkled and rough, and there are many irregular polymers distributed. This is due to the modification of GO surface by silane coupling agent, which greatly improves the surface roughness of GF. Moreover, there are no obvious gullies and clusters on the surface of GO and GF after modification, which indicates that GO and GF have good interfacial compatibility. The adhesion of GO on the surface of GF makes the interface form a three-dimensional structure, which greatly improves the interface performance of the composite and enhances the thermal conductivity. Fig. 4 (c) shows the micro morphology of dispersed-GO/GF/EP composite with 0.05 vol% GO under random dispersion condition. GO sheets are uniformly dispersed in EP matrix, and only small amount of lamellae are stacked. The agglomerated GO is gathered on the surface of GF, resulting in the weakening of the surface adhesion between GF and EP. The aggregation of GO prevents the interface interaction between EP matrix and GF, and even reduces the thermal conductivity of the composites.

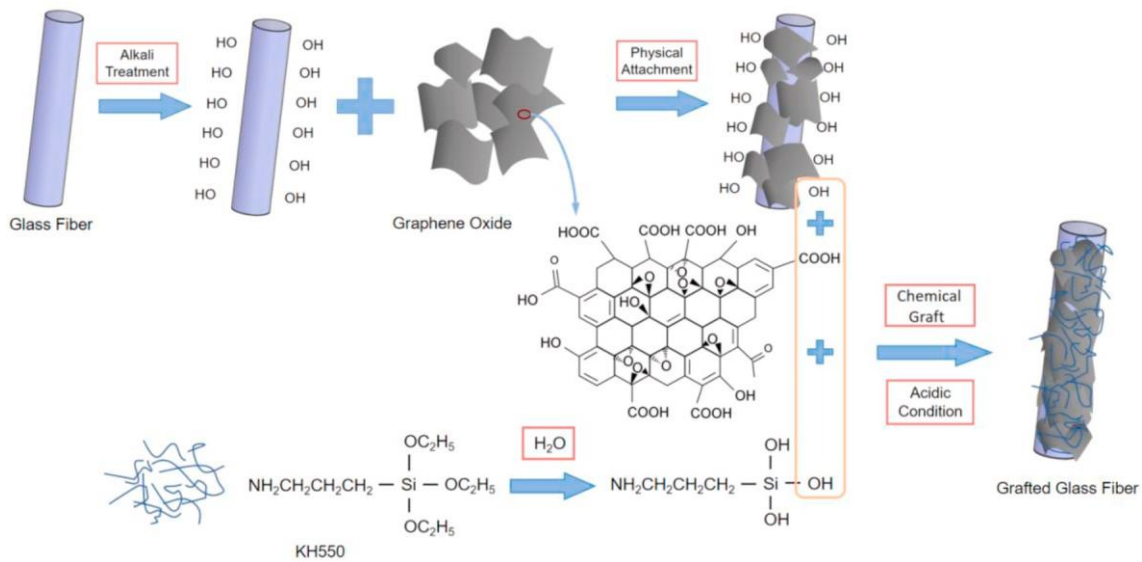


Fig. 1. Schematic diagram of GO grafted on GF.

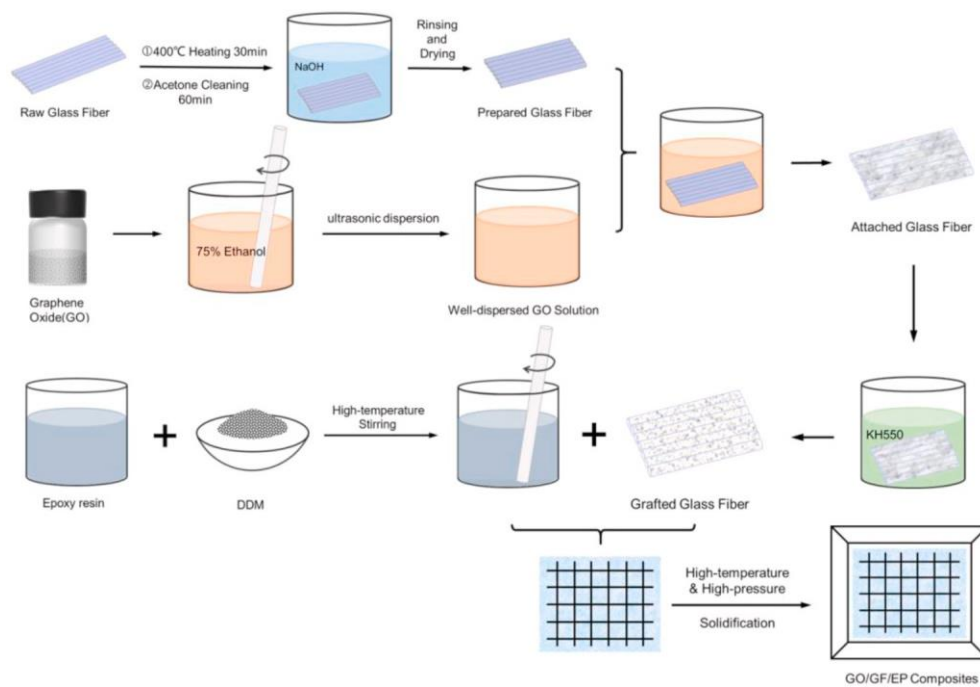


Fig. 2. Preparation of grafted-GO/GF/EP composites.

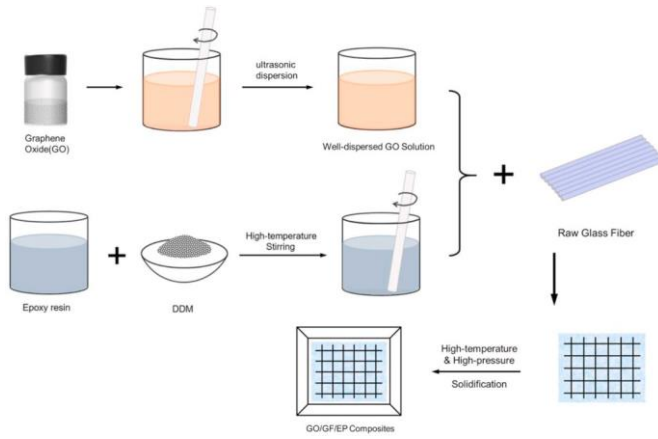


Fig. 3. Preparation of dispersed-GO/GF/EP composites.

2.3.2 Heat transfer characteristics test

The heat transfer performances of the dispersed-GO/GF/EP and grafted-GO/GF/EP were acquired by heat transfer characteristics test. The experiments were conducted at 10 °C. As shown in Fig. 5, the electric heating system was designed as an adjustable heating system to control the heating temperature by changing the DC power supply (Maxon MS605D, rated power of 300W). The temperature distribution was measured by infrared thermal imager (Testo 869). The thermal sensitivity of the infrared thermal imager is 0.06 °C, and the measurement error is ± 1 °C. The temperature distribution images were composites with 0.05 vol% GO was prepared by dispersing GO suspension. The two mixed solutions were respectively treated with ultrasonic vibration for 15 min with ultrasonic cleaning machine. After ultrasonic treatment, the two solutions were mixed and ultrasonic treated again for 10–15 min. Then, the GF was immersed in the prepared solution and put it into the mold. Then the mold was put into the blast oven and cured at 80 °C for 2.5 h, and heated to 150 °C for 2h. The process of the experiment is shown in Fig. 3.

2.3 Apparatus and measurement

2.3.1 Samples characterization and morphology

Scanning electron microscopy (SEM, Hitachi SU-8010) was employed to characterize the cross-section morphology of the grafted-processed by the TESTO IRSOFT-PC software. During the experiments, the dispersed-GO/GF/EP and grafted-GO/GF/EP test pieces were bonded on the heating pad with high thermal conductivity adhesive. The data acquisition cycle was set as 15 s.

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3. Finite element model

3.1. Heat transfer RVE model

GO/GF/EP composites can be approximately regarded as composed of numerous representative volume element (RVE) arranged in accordance with certain rules. According to the SEM image of the composites in this paper, the heat transfer RVE models of grafted-GO/GF/EP and dispersed-GO/GF/EP were established by employing the commercial finite element software. As shown in Fig. 6, the composite material is composed of GF, EP and GO. The geometry of RVE: 78 (length) \times 78 (width) \times 30 (thickness) μm^3 . The diameter of glass fiber is 12 μm , the spacing between glass fiber layers is 1.5 μm , and the average diameter of GO sheets is 0.6 μm .

In the RVE model of dispersed-GO/GF/EP (see Fig. 6(a) and (b)), the GO lamellae are randomly distributed on the columnar surface of GF, and the GF is embedded in the EP without obvious delamination or separation. In the RVE model of dispersed-GO/GF/EP (see Fig. 6(c) and (d)), the GO sheets are randomly and uniformly distributed, which embedded in EP matrix. No obvious delamination or separation phenomenon was occurred when the GF and GO were embedded into the epoxy resin matrix. Therefore, it can be considered that there has no contact thermal resistance between GO, GF and EP. On the basis of the RVE models, the properties of grafted-GO/GF/EP and dispersed-GO/GF/EP composites are investigated.

In the finite element calculation, the discretization of the complex geometry of GO/GF/EP composites leads to high calculation cost. Therefore, the following assumptions and simplification are established in the process of modeling:

GO sheets are simplified as nano scale equal diameter circular structure. According to the principle of minimum surface energy, GO shows the natural state of three-dimensional surface undulation due to the two-dimensional structure.

(1) It is assumed that the properties of GO sheets and EP are temperature independent. In order to simplify the calculation, the dependence of temperature is ignored [35].

(2) The steady-state heat transfer without volume heat generation is considered [36].

(3) It is considered that the interface between GO sheets and EP matrix is well bonded.

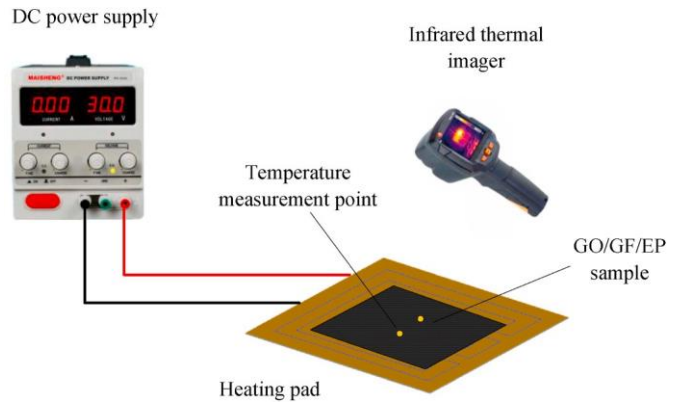


Fig. 5. The diagram of heat transfer test.

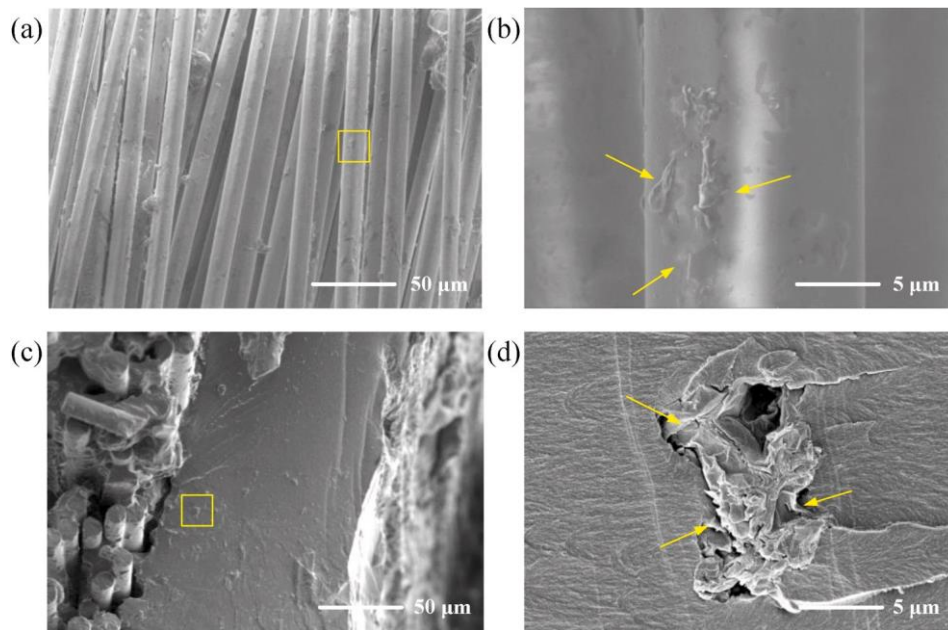


Fig. 4. The SEM images of (a) grafted-GO/GF/EP with (b) enlarged image, and (c) dispersed-GO/GF/EP with (d) enlarged image.

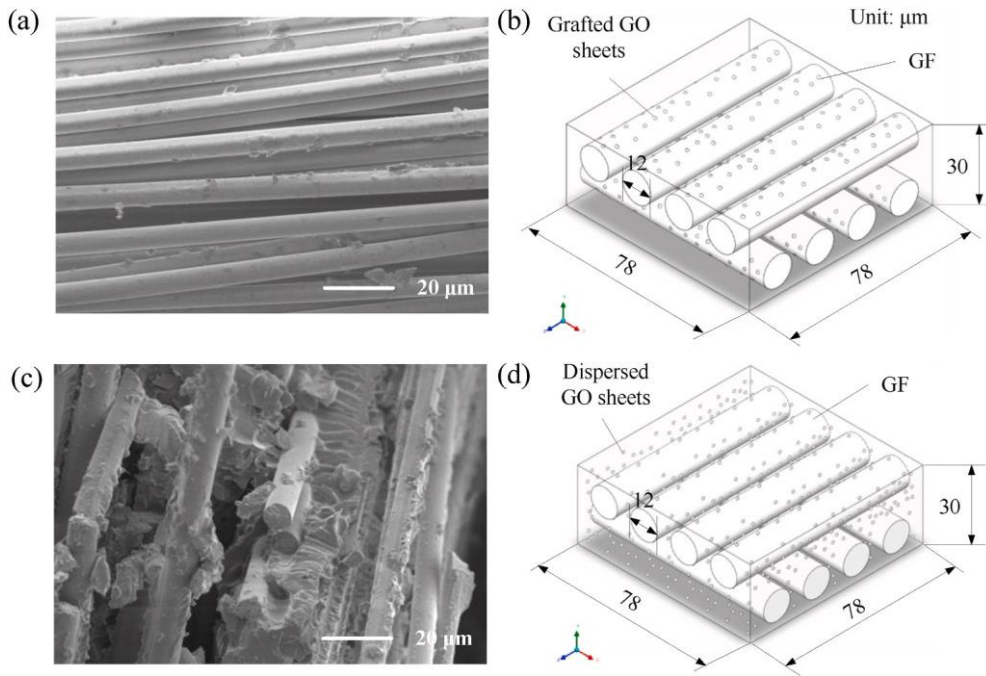


Fig. 6. The heat transfer modeling of grafted-GO/GF/EP and dispersed-GO/GF/EP composites.

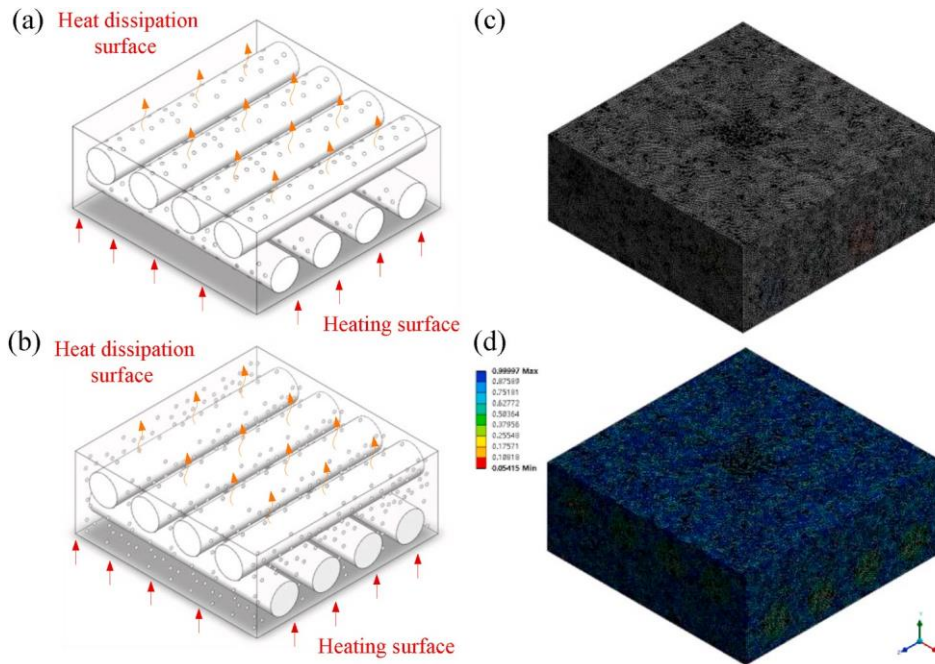


Fig. 7. The (a)–(b) boundary conditions, (c) mesh and (d) quality of the heat transfer models.

3.2. Meshes and boundary conditions

Three dimensional transient heat transfer analysis of grafted-GO/ GF/EP and dispersed-GO/GF/EP composites were performed respectively. The grids numbers of grafted-GO/GF/EP and dispersed-GO/GF/EP models are 692760 and 872590. The element qualities of grafted-GO/GF/EP and dispersed-GO/GF/EP models are 0.75 and 0.74, which indicating that the quality of the grids meets the requirements of the calculation as shown in Fig. 7.

The model adopts periodic boundary condition (PBC) and the boundary setting can ensure that each RVE model has the same loading mode, and there is no separation between adjacent RVEs. The bottom surface of the RVE model is set as the heating surface (δ) and the specific temperature (T). The upper surface (δ') is set as the heat dissipation surface and the specific heat transfer coefficient. The other symmetrical surfaces are set as insulation surfaces and the two opposite sides are set as η and η' , ζ and ζ' respectively. The heat transfer boundary conditions are as follows: the initial temperature of the material is 10 °C, the heat flux of the heating surface is 6000 W/m², and the heat transfer coefficient of the heat dissipation surface is 5 W/ m² · °C. The PBC equations of surfaces and nodes can be summarized as follows:

$$\lambda_{\delta} = \lambda, \quad T_{\delta'} = T \quad (1)$$

$$T_{\eta} - T_{\eta'} = 0, \quad T_{\zeta} - T_{\zeta'} = 0 \quad (2)$$

3.3. Material description

The sample in this study is the grafted-GO/GF/EP and dispersed-GO/ GF/EP composites, which uses EP as matrix material and GO as reinforcement material. The thermo physical parameters of materials of dispersed-GO/GF/EP and grafted-GO/GF/EP composites are as shown in Table 1.

4. Results and discussion

4.1. Validation of simulation

Fig. 8 shows the average temperature of the dispersed-GO/GF/EP and grafted-GO/GF/EP composites under the heating voltage of 2.6 V. Compared the experimental data with simulation data, it can be seen that the temperature rise curves of the dispersed-GO/GF/EP and grafted-GO/GF/EP composites keep the same trend. Meanwhile, the maximum standard deviation of the dispersed-GO/GF/EP between the experimental data and the simulation data is 0.70 °C. For the grafted-GO/GF/EP, the maximum standard deviation between the experimental and the simulation data is 0.68 °C. Based on the analysis of standard deviation, the numerical simulation results are in good agreement with the experimental data, which shows that the numerical simulation model of dispersing and grafting can predict the heat transfer characteristics of dispersed-GO/GF/EP and grafted-GO/GF/EP respectively. In the follow-up study of this paper, the numerical simulation data was used for research and analysis.

Table 1
Thermo physical parameters of materials.

Materials	Graphene oxide	Glass fiber	Epoxy resin
Density (kg/m ³)	990	2400	1500
Young's modulus (MPa)	1.34×10^6	7.30×10^4	3.41×10^3
Poisson's ratio	0.2	0.2	0.4
Thermal conductivity (W/(m·K))	4840	1.3	0.363
Thermal expansion coefficient (/°C)	8.2×10^{-6}	54×10^{-6}	80×10^{-6}
Specific heat capacity (J/(kg·K))	700	794.2	550

4.2 Heat transfer capability analysis

4.2.1 Heat transfer of dispersed-GO/GF/EP

For the samples prepared by dispersion method, GO is completely embedded in the EP matrix. The heat flows pass through both the interface of GO/EP, and the in-plane direction of GO. The high thermal conductivity of GO significantly increases the in-plane heat flux. As shown in Fig. 9, the cross section is taken at X = 21 μm and the GO content of dispersed-GO/GF/EP is 0.05 vol%. The results show that the heat flux inside GO (point A and point B) is 13804.81 and 10178.26 W/m², while the heat flux around GO (point C and D) is only 4865.29 and 5135.72 W/m². Many oxygen-containing functional groups on the surface of GO, such as hydroxyl, carboxyl and epoxy groups, promote the chemical cross-linking between GO and EP, thus form covalent bonds. The existence of covalent bonds enhances the interface strength between EP and GO, reduces the interface thermal resistance, and improves the heat flux. However, the hydrophilic GO in the organic polymer has poor wettability and dispersion, and there are strong hydrogen bonding and dipolar interaction between oxygen-containing groups between the layers, which makes it difficult to disperse and peel GO in the EP matrix. Therefore, GO cannot give full play to its enhanced heat transfer performance, which limits the improvement of thermal conductivity of composites.

4.2.2 Heat transfer of grafted-GO/GF/EP

Fig. 10 shows the heat flux distribution near the branch of GO on the GF. The cross section is taken at X = 69 μm, and the GO content of grafted-GO/GF/EP is 0.1 vol%. At points A and B, the heat flux values are 2.12×10^6 W/m² and 1.34×10^6 W/m² respectively. While at points C and D, the heat flux values are 4681.93 W/m² and 1222.41 W/m². The heat flux in GO sheets is significantly higher than that in EP, which indicates that heat is preferentially transferred through GO. Compared with the heat flux values, the heat flux inside the GO sheets is 452 times higher than that around the GO sheets. This shows that the branching method can better improve the interface interaction between GO and EP, and give full play to the high thermal conductivity of GO. This is due to the interface bonding between GF and EP matrix enhanced by coupling agent.

Moreover, the surface of GO is grafted with oxygen-containing active groups and alkyl molecular chains. These molecular structures reduce the surface energy and increase the lipophilicity of GO, which improve the wetting dispersion and interfacial bonding of GO/EP. The alkyl chains introduced into GO surface also increase the interlaminar spacing and inhibit the entanglement of resin molecular chains, so that the resin molecules can overcome the hydrogen bond between GO interlayers and the dipole interaction between oxygen-containing groups to enter into GO interlayers, thus improving the exfoliation dispersion of GO. Therefore, the coupling agent acts as a molecular bridge between the thermal conductive filler GO and EP, and improves the interfacial bonding strength, which conducting to the improvement of the thermal conductivity of the composite.

4.2.3. Heat transfer comparison of grafted and dispersed GO/GF/EP

Fig. 11 shows the temperature distribution of GF/EP, grafted-GO/GF/EP, and dispersed-GO/GF/EP composites under the heating conditions of 6000 W/m². The GO content of grafted-GO/GF/EP and dispersed-GO/GF/EP is 0.1 vol%. By displaying the temperature distribution on the surface of RVE model, the thermal conductivity of the composites can be visualized. It can be seen from the figure that the temperature distribution of the dispersed-GO/GF/EP and the pure GF/EP is similar. The overall temperature of the upper surface of the grafted-GO/GF/EP is higher than that of the others. The area of the high temperature zone is also significantly larger than that of the other two groups, which indicating that the heat transfer effect of the grafted-GO/GF/EP is better.

Fig. 12 shows the minimum and maximum temperature distribution curves on the heat dissipation surface of the GF/EP, the grafted-GO/GF/EP, and the dispersed-GO/GF/EP samples. It can be seen from Fig. 12(a) that two different doping methods can improve the thermal conductivity of the composites. The surface temperature of pure GF/EP composites increased from 10 °C to 4.31 °C, the temperature of dispersed-GO/GF/EP increased to 4.46 °C, and that of grafted-GO/GF/EP increased to 7.86 °C. Compared with the undispersed GO group, the thermal conductivity of the composites with GO grafted GF is increased by 82.37%. While the thermal conductivity of the dispersed-GO/GF/EP only increased by 3.48%, which indicates that the thermal conductivity of the composites with GO grafted on GF is better than that of the composites with GO dispersed in EP. In addition, according to the temperature curve, the heat transfer rate of the grafted-GO/GF/EP is 0.33 °C/s in 10–35 s, which is slightly higher than that of the dispersed-GO/GF/EP (0.24 °C/s). After heating for 35 s, the heat transfer rate between the grafted and the dispersed composite is almost the same, which can reach 0.46 °C/s (see Fig. 12(b)).

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Fig. 14 compares the effects of the formation of GO-GO heat conduction network on the thermal conductivity of the composites. It can be seen that due to the formation of the heat transfer paths, the heat is quickly transferred to the whole material through the heat transfer channels. Compared with the composites without GO-GO heat transfer network, the temperature of the heat dissipation surface is increased by 0.5 °C, and the heat transfer effect is increased by 5 times.

4.2.2 Heat transfer path of grafted-GO/GF/EP

The temperature distributions of grafted-GO/GF/EP and pure GF/EP composites are shown in Fig. 15. The cross section is taken at X = 48 μm. As shown in Fig. 15(a), the GO sheets random branches on the outer surface of GF cylinder (in the red circle). It can be seen that the temperature isotherms fluctuate around the GF and bends to the GO sheets (mark with yellow arrows). Due to the grafting of GO on the surface of GF, the heat transfer path of the joint between GF and EP was changed. The heat is transferred upward through the heat conduction network formed between GO sheets. However, when the distance between the two GF bundles is large, the thermal conduction network between GO-GO will be broken, which leads to a significant decline in the thermal conductivity of the composites.

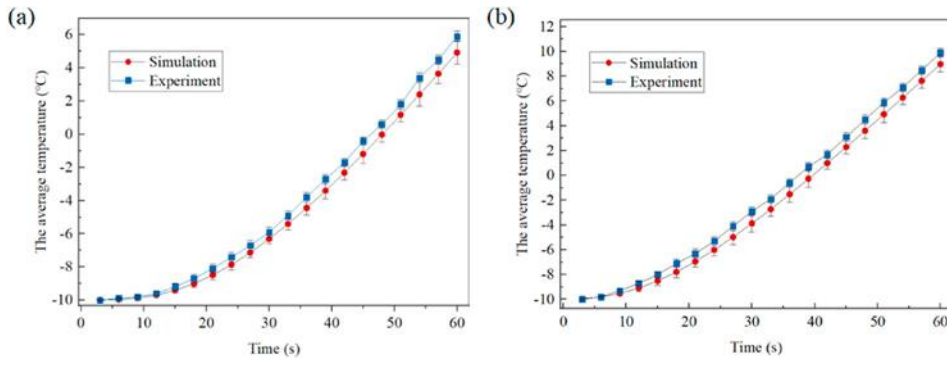


Fig. 8. Comparison of experimental and simulation data of (a) dispersed-GO/GF/EP and (b) grafted-GO/GF/EP composites.

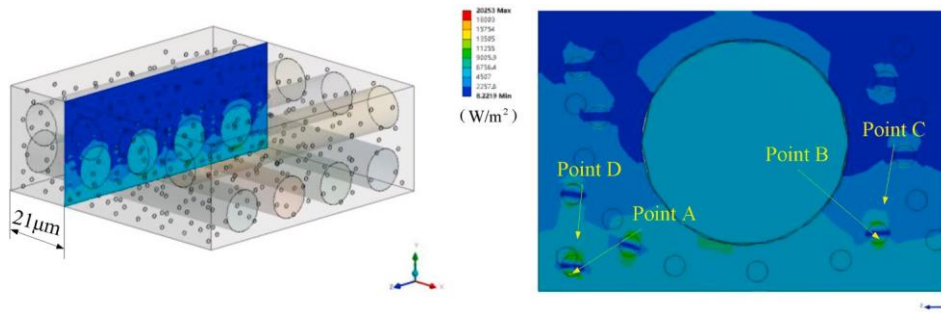


Fig. 9. The heat flux of GO dispersion area.

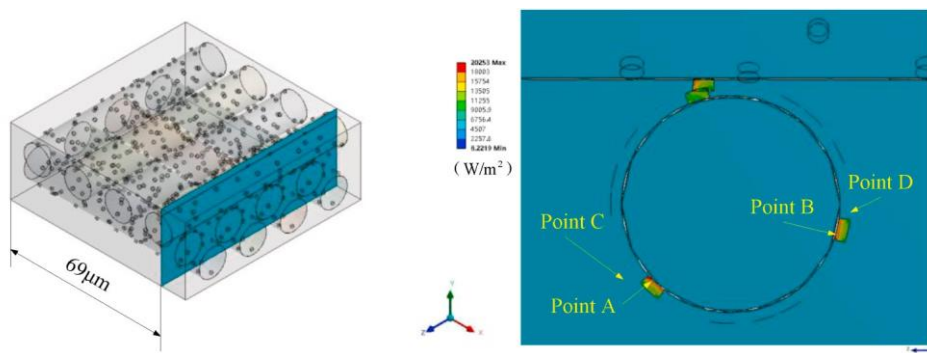


Fig. 10. The heat flux of GO branch area.

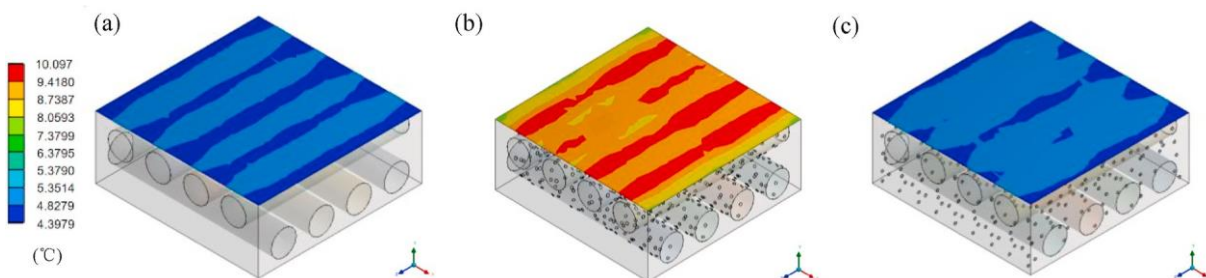


Fig. 11. Temperature distributions of (a) GF/EP, (b) grafted-GO/GF/EP, and (c) dispersed-GO/GF/EP composites.

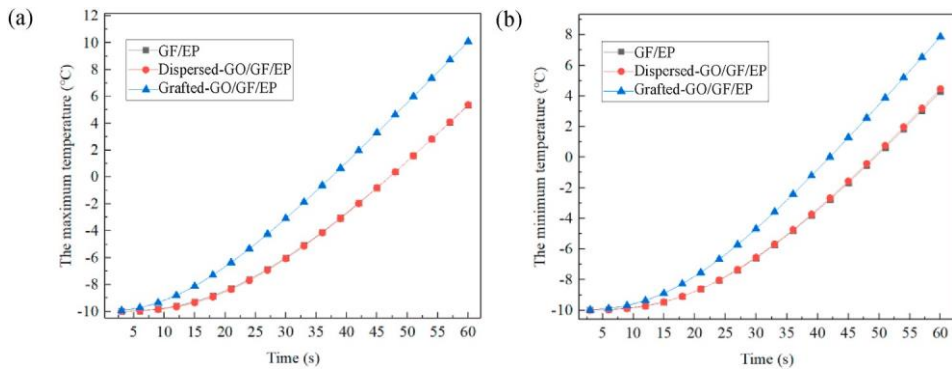


Fig. 12. The (a) maximum and (b) minimum temperature on the heat dissipation surface.

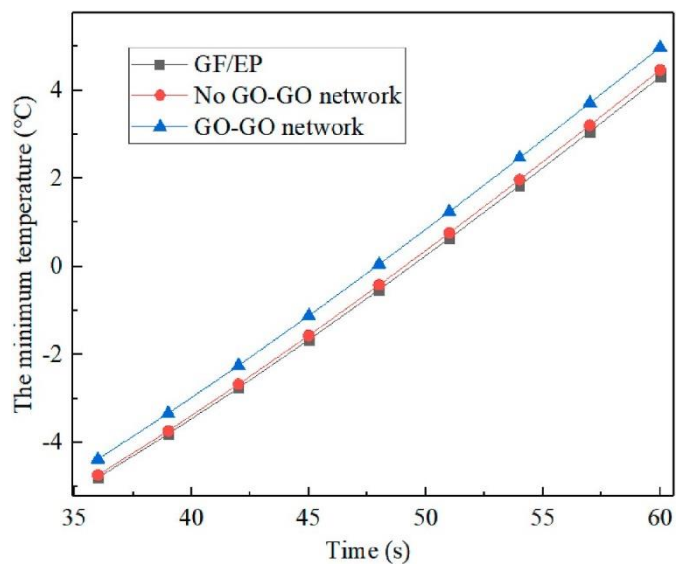


Fig. 14. The heat flux of GO branch area.

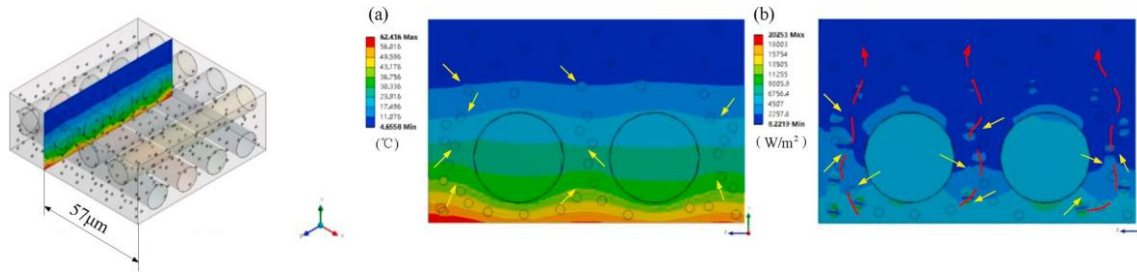


Fig. 13. The (a) temperature and (b) heat flux of GO dispersion area.

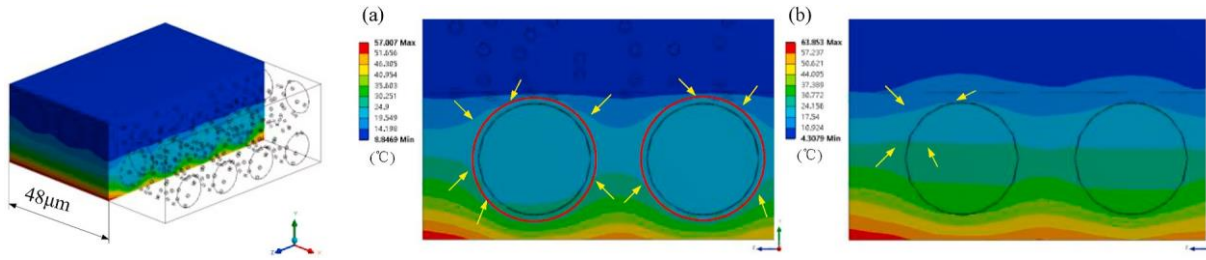


Fig. 15. Heat transfer path of grafted-GO/GF/EP.

4.4 Analysis of influencing factors of heat transfer

Dispersing content of GO dispersion and interfacial bonding of GO in EP are improved, and the interfacial heat transfer performance is enhanced. As shown in Fig. 17 (a), when the GO diameter is 0.6 μm , 1.2 μm , 1.8 μm , and 2.4 μm , the Fig. 16 shows the temperature distribution of the composites with different GO doping contents. It can be seen that with the increasing of GO doping content, the thermal conductivity of grafted-GO/GF/EP composites is improved in varying degrees. However, GO doping content has little effect on the thermal conductivity of dispersed-GO/GF/EP composites, which the thermal conductivity of the composites increases by only 4%. It indicates that the heat transfer enhancement effect of GO doping is weakened by the high interfacial thermal resistance in the composites, thus the effect on the thermal conductivity of dispersed-GO/GF/EP composites is not obvious. On the contrary, when the GO content is 0.05 vol%, the thermal conductivity of the grafted-GO/GF/EP increases by 60.32%, which indicating that a better heat transfer network is formed. Increasing the GO content is conducive to enhancing the heat transfer effect of the composite.

However, the doping content of GO in grafted-GO/GF/EP is not as high as possible. When the GO content is more than 0.05 vol%, the heat transfer enhancement effect is no longer significant. When the GO content is 0.05 vol%, 0.10 vol%, and 0.15 vol%, the surface temperature of the composite increases by 2.61 $^{\circ}\text{C}$ (60.55%), 3.55 $^{\circ}\text{C}$ (82.37%), and 4.23 $^{\circ}\text{C}$ (98.14%). The reason is that the GO agglomeration in EP matrix results in the loss of nano effect, which leads to the increasing of interface thermal resistance. In addition, the GO agglomeration leads to obvious pores and defects in the EP, which is not conducive to the heat transfer in the composites. Diameter of GO Fig. 17 shows the temperature distribution of grafted GO/GF/EP and dispersed-GO/GF/EP with different GO diameters. For grafted-GO/GF/EP composites, the increase of GO diameter contributes to the modification by coupling agent. On the one hand, the large GO diameter increases the possibility of successful grafting. On the other hand, GO with larger surface area can react with more coupling agent molecules, which leads to the introduction of more oxygen-containing active groups and alkyl molecular chain structure on the GO surface. Thus, the wetting temperature of grafted-GO/GF/EP is 5.73 $^{\circ}\text{C}$, 7.86 $^{\circ}\text{C}$, 8.49 $^{\circ}\text{C}$, and 8.95 $^{\circ}\text{C}$ respectively. The thermal conductivity is improved by 32.95%, 82.37%, 96.98%, and 107.66% compared with that of GF/EP.

As shown in Fig. 17 (b), for the dispersed-GO/GF/EP composites, the thermal conductivity of the composites increases with the increase of GO diameter. Different from the grafted-GO/GF/EP, although the thermal conductivity improvement rate of the dispersed-GO/GF/EP is less than 25%, the increase of GO diameter always has a positive effect on the thermal conductivity of the composites. With the increasing of GO diameter, the thermal conductivity of the dispersed-GO/GF/EP increased by 3.02%, 7.78%, 15.54% and 21.81%, respectively. In addition, by comparing the effects of GO doping contents and diameter on the thermal conductivity, it can be found that the effect of GO diameter is more obvious, which indicating that GO diameter is a more significant factor.

4.4.3 Number of GO layers

Fig. 18(a) shows the temperature distribution of composites with different layers of GO sheets. It can be seen that the thermal conductivity of the composite is greatly improved by the grafting of single-layer GO, and the minimum temperature of its surface reaches $6.88\text{ }^{\circ}\text{C}$. Compared with the pure GF/EP, the thermal conductivity of the grafted-GO/GF/EP is improved by 59.63%. With the increase of GO layers, the thermal conductivity of the composites improves steadily. The thermal conductivity of the composite grafted with five layers GO increased by 97.02%. However, the thermal conductivity of the composite grafted with 10 layers of GO is only 4.22% higher than that of grafted with five layers of GO. The results indicate that the increase of GO layers can increase the contact thermal resistance between GO layers and reduce the enhancement effect of grafting on the thermal conductivity of the composites.

As shown in Fig. 18(b), the increase of GO layers has no significant effect on the thermal conductivity of the composites. The composite dispersed with two layers GO has the best thermal conductivity, which surface temperature is $4.53\text{ }^{\circ}\text{C}$. However, the maximum increase of GO layers on the overall thermal conductivity of the composites is less than 5.10%, which indicates that the increase of GO layers has no significant effect on the heat transfer enhancement of the dispersed-GO/GF/EP. This is due to the influence of doping multilayer GO on the dispersion in EP, which increases the agglomeration phenomenon.

4.4.2 Distance of GF layer

Fig. 19 shows the thermal conductivity of composite materials with different glass fiber layer distances. With the increase of glass fiber layer distance, the thermal conductivity of the composites decreased significantly. When the distance between glass fiber layers is 1 mm, the temperature of the grafted-GO/GF/EP is $4.28\text{ }^{\circ}\text{C}$ after heating for 60 s.

Compared with the composite with $1.5\text{ }\mu\text{m}$ GF layer distance, the thermal conductivity is reduced by 45.62%. For the dispersed-GO/GF/EP, the effect of GF layer spacing on the heat transfer performance of composites is more obvious. When the GF layer spacing is $6.0\text{ }\mu\text{m}$, the temperature of the dispersed-GO/GF/EP decreases to $1.62\text{ }^{\circ}\text{C}$. Thus, the interlayer spacing of glass fiber is an important factor affecting the thermal conductivity of composites.

With the increasing of the glass fiber layer spacing ($>6.0\text{ }\mu\text{m}$), the temperatures of the grafted-GO/GF/EP and dispersed-GO/GF/EP even decrease to below $0\text{ }^{\circ}\text{C}$. When the glass fiber layer spacing is $30.0\text{ }\mu\text{m}$, the temperatures of the branch group and dispersion group is $4.66\text{ }^{\circ}\text{C}$ and $7.72\text{ }^{\circ}\text{C}$, respectively. It indicates that when the GF layer distance is large, the GO-GO heat conduction network in the composite is disconnected, which the heat cannot be transferred from the heat conduction network formed by GO smoothly. Moreover, the thermal conductivity of the composites decreased significantly due to the large space between the GF layers and the large interfacial thermal resistance between the layers filled with EP.

5. Conclusions

In this paper, GO with high thermal conductivity was used as nano reinforcement and introduced into epoxy resin matrix composites in dispersed and grafted ways. Based on the micro heat transfer RVE

model, the heat transfer characteristics of dispersed-GO/GF/EP and grafted-GO/GF/EP prepared by two different doping methods were analyzed. The results show that GO is grafted onto the surface of GF and the alkyl chain is introduced into GO surface in the grafted-GO/GF/EP composites, which improving the exfoliation dispersion of GO in EP, and further significantly effecting the thermal conductivity of the composites.

With the increasing of GF layer spacing, the heat conduction network between GO and GO is broken. The heat transfer effect of the dispersed- GO/GF/EP and grafted-GO/GF/EP is greatly affected by the GO diameter. Increasing the GO diameter is beneficial to chemical modification, improves the GO/EP interface interaction, and reduces the interface thermal resistance. The dispersed-GO/GF/EP and grafted-GO/GF/EP composites with two-layer GO dispersion have the best thermal conductivity under the condition of 2.4 μm GO diameter and 0.1 vol% content. The thermal conductivities of dispersed-GO/GF/EP and grafted-GO/GF/EP are increased by 21.81% and 107.66% respectively. With the increasing of GO layer, there is no significant effect on the heat transfer enhancement of dispersed-GO/GF/EP. This is due to the negative effect of multilayer GO on the dispersion in EP and the increasing of agglomeration.

CRedit author statement

Junjie Ren: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation. Long Chen: Investigation, Data curation, Formal analysis, Visualization, Zhanqiang Liu: Writing-review & editing. Qinghua Song: Visualization, Writing-original draft. Chao-zong Liu: Investigation, Formal analysis, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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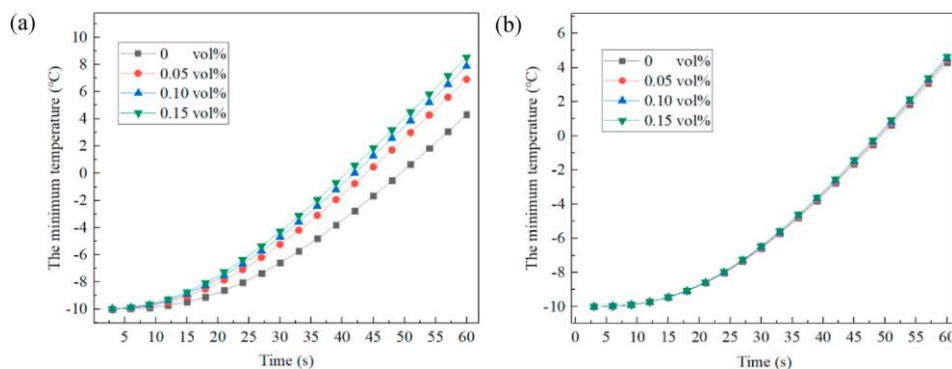


Fig. 16. The temperature of (a) grafted-GO/GF/EP and (b) dispersed-GO/GF/EP with different GO contents.

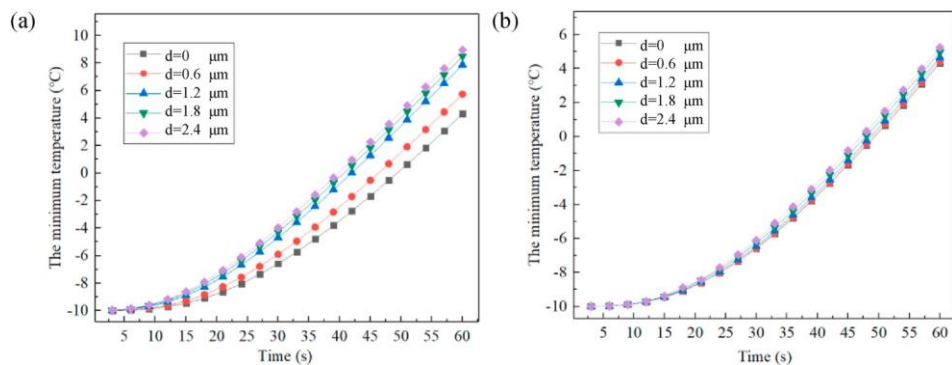


Fig. 17. The temperature of (a) grafted-GO/GF/EP and (b) dispersed-GO/GF/EP with different GO diameters.

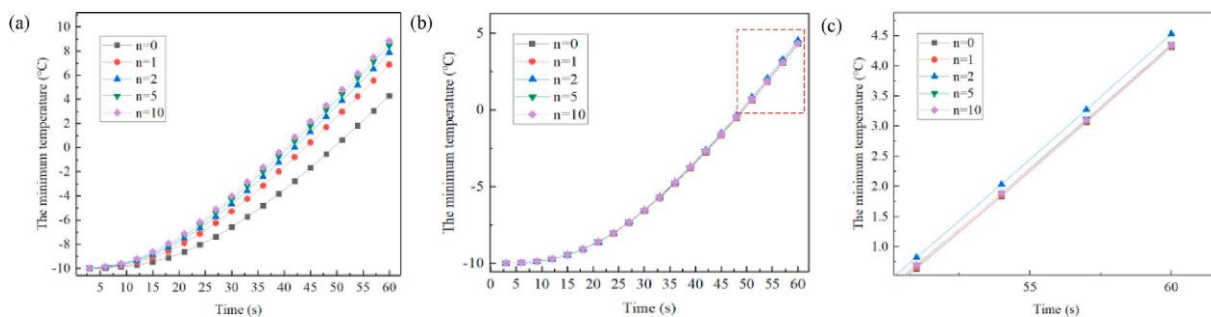


Fig. 18. The temperature of (a) grafted-GO/GF/EP, (b) dispersed-GO/GF/EP, and (c) enlarged image with different GO layer numbers.

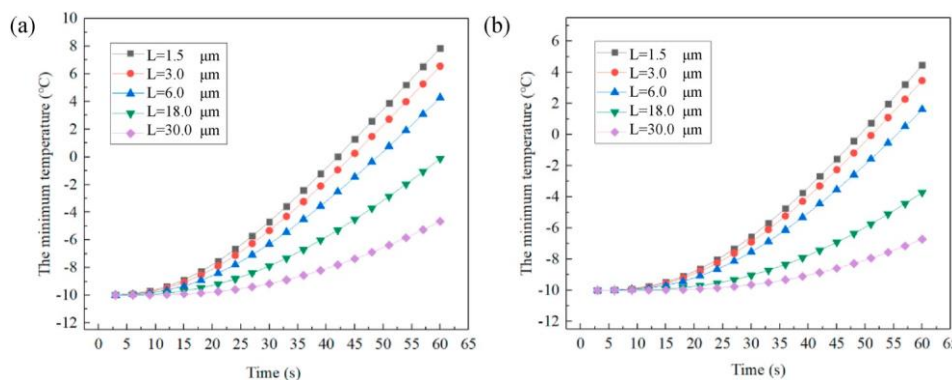


Fig. 19. The temperature of (a) grafted-GO/GF/EP and (b) dispersed-GO/GF/EP with different GF layer distances.

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