Composites: Part A 168 (2023) 107463

Contents lists available at ScienceDirect

Composites Part A

journal homepage: www.elsevier.com/locate/compositesa

Synergistically enhancing the electrical conductivity of carbon fibre reinforced polymers by vertical graphene and silver nanowires

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ARTICLE INFO

Keywords:
Electrical conductivity
Carbon fibre reinforced polymers
Vertical graphene
Silver nanowires

ABSTRACT

Increasing the electrical conductivity of carbon fibre reinforced polymers (CFRPs) holds great promises for a range of applications, such as removing the need for metallic meshes in the protection against electromagnetic interference and lightning strikes. Herein, a hybrid method of improving the electrical conductivity of CFRPs by functionalizing carbon fibres with vertical graphene (VG) and modifying the matrix with silver nanowires (AgNWs) is introduced. The results revealed that the hybrid method increased the through-thickness and the in-plane electrical conductivities by almost 38 times and 39%, respectively, without adversely affecting mechanical properties. Finite element modelling revealed that the unprecedented synergy is due to the significant reduction in the contact resistance between carbon fibres by the combination of VGs on the fibres and the AgNWs in the matrix. Computational modelling showed that the electrical conductivity increase can reduce the joule heat density by around one thousand times under simplified lightning strike conditions.

1. Introduction

One common issue in the application of carbon fibre reinforced polymers (CFRPs) is their inherently low electric conductivity, especially in their through-thickness direction [1,2,3], which poses challenges for protection against lightning strikes, electromagnetic interference, and high energy electromagnetic radiation [4,5,6] in aircraft, spacecraft, wind turbines, and all-electric vehicles [7,8,9,10]. For example, without adequate conductive protection, lightning strikes can result in severe structural damage and potentially catastrophic consequences [11,12,13,14]. To address this issue, copper or aluminium expanded foils are added on the external surface of composites [15,16] to conduct the lightning current and thus avoid potential damages to the aircraft. However, the expanded metal foils not only add weight but also complexities in terms of manufacturing and repairs [17]. Moreover, metal foils and the CFRP structure have distinct mechanical properties, which can cause additional stresses and hence can result in failures [18,19]. Therefore, it is highly desirable and of great practical importance to explore new measures to enhance the electrical conductivity without the use of metal foils and without compromising the lightweight benefits of CFRPs.

Incorporating conductive nanofillers into the matrix of CFRPs has been reported as an appealing solution to enhance the electrical conductivity without adding significant weight [20]. For example, Senis et al. [21] dispersed graphene oxide (GO) (6.3 vol%) to into an epoxy resin, leading to 200 % improvement in the CFRP's through-thickness electrical conductivity. Wu et al.[22] and Raj et al. [23] carried out in-situ aligning of conductive nanofillers such as graphene nanoflakes (GNFs) and carbon nanofibers (CNFs) in the epoxy composites under an alternating current (AC) electrical field, the electrical conductivity of epoxy composites can be improved by 7–8 orders with this procedure. Similar, Singer et al. [24] did an in-situ aligning of carbon nanotubes (CNTs) with an AC electrical field after introducing the CNTs into epoxy to fabricate CFRP laminates. With aligned CNTs in the epoxy, an over 8-

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https://doi.org/10.1016/j.compositesa.2023.107463
Received 30 November 2022; Received in revised form 19 January 2023; Accepted 24 January 2023
Available online 26 January 2023

fold improvement in the electrical conductivity of composites was achieved. However, it has also been reported that as the conductive nanofiller concentration increased, the relative electrical conductivity improvement became less pronounced [25,26]. More importantly, high loadings of conductive nanomaterials within the matrix may lead to nonuniform distribution and a highly viscous un-cured epoxy mixture that is difficult to infuse into carbon fabrics [27].

A complementary strategy of grafting carbon fibres with conductive nanofillers has also demonstrated promising results [28]. Pozegic et al. [29] used a plasma enhanced chemical vapour deposition (PECVD) technique to graft CNTs onto the carbon fibre surface to explore the electrical conductivity of the resultant CFRP, reporting a 510 % increase in the through-thickness and a 330 % increase in the in-plane electrical conductivity of the composites, respectively. Russello et al. [30] reported a direct deposition of CNTs on unidirectional carbon fibre tow tapes and used them to fabricate thin-ply laminates. It was observed that the deposited CNTs boosted the through-thickness electrical conductivity of the CFRP by eight times. However, the electrical conductivity improvement was less pronounced as the laminate thickness increased. Lee et al. [31] adopted an electrophoretic deposition technique to deposit reduced graphene oxides (rGO) on the surface of carbon fibres and reported an increase of 25 % in the in-plane and 250 % in the through-thickness electrical conductivity. Qin et al. [32] used graphene nanoplatelets (GNPs) to dip-coat carbon fibres and observed a 165 % through-thickness conductivity improvement of the composites. Wu et al. [33] combined freeze-drying with the sizing process to deposited silver-plated CNTs onto the carbon fibres to achieve 385.8 % improvement of the through-thickness conductivity. Compared to the matrix modification approach, fibre modification approaches can avoid the problem of increasing resin viscosity [34]. However, functionalizing carbon fibres alone, which may require aggressive reacting conditions and elevated temperatures, has been found to achieve limited increase in electrical conductivity without excessively degrading the mechanical properties of the fibres.

One major issue common to both the methods is that the improvement in electrical conductivity peaks at a low volume fraction of the nanofiller materials, because of the challenges of high resin viscosity and potential damages to carbon fibres. Inspired by recent research into the synergistic effect of multiple conductive nanofillers in improving the electrical conductivity of composites [34,35], herein we present a study into the effectiveness of functionalising both the matrix and carbon fibres using different nanomaterials to achieve greater improvement in electrical properties beyond those attainable by either method alone.

In this work, the carbon fibres and epoxy resin were simultaneously modified with different conductive nanofillers. In particular, the carbon fibres were grafted with vertical graphene (VG) using a PECVD process, and silver nanowires (AgNWs) were dispersed in an epoxy matrix. The deposition of VG on carbon fibre requires a short processing time (10 min) at a low temperature ~ 400 °C, which has been reported not to degrade the mechanical properties of carbon fibres [36]. The CFRPs were then fabricated from the modified fibres and the matrix using wet lay-up and hot press procedures, with their in-plane and through-thickness conductivities both measured, and the results compared with unmodified composites. The electrical conductivity results reveal that using AgNWs and VG together, the CFRP's through-thickness electrical conductivity can be improved by over 38 times, significantly greater than the state-of-the-art values in existing relevant studies. The in-plane electrical conductivity of the CFRP laminate has been improved by 39 %. These enhancements have been achieved without any adverse effect on the tensile properties of the composites. This work demonstrates the merits of using a hybrid modification approach and hybrid conductive nanofillers in improving the electrical conductivity of CFRPs.

2. Experimental section

2.1. Materials

A plain weave T300 carbon fibre fabrics (C06343B Carbon Plain, Toray Industries Inc., Japan) was used in this work, which have an areal density of 19.8 mg/cm^2, average thickness of 0.23 mm, and average fibre diameter of around 7 μm. The fibres were grafted with VGs radially on the surface via a plasma-enhanced chemical vapor deposition (PECVD) process at a low reacting temperature of ~ 400 °C using methane (CH_4) as precursor gas [37].

An epoxy resin and amine curing agent (Raku Tool EL-2203 and Raku Tool EH-2970–1, GMS Composites, Australia), with a ratio of 10:3 by weight, were used as the matrix, which can be cured at room temperature for 24 h followed by a post cure at 120 °C for 14 h.

AgNWs were prepared *via* a polyol method [38,39]. Analytical grade Silver Nitrate ($AgNO_3$), Polyvinylpyrrolidone (PVP, Mw 40000), Sodium chloride (NaCl), and glycerol were obtained from Sigma-Aldrich, Australia.

2.2. Preparation of VGs deposited carbon fibres using PECVD method

VGs were grown on carbon fibres using a PECVD technique. A carbon fibre fabric was fixed to a supporting platform vertically in the centre of the PECVD's vacuum chamber, with a 10 mm distance between the plasma coil and the supporting platform. After the base pressure in the chamber reached 0.02 Pa, Ar gas was introduced with a flow rate of 10 sccm (standard cubic centimetre per minute). When the Ar gas in the chamber increased the pressure to ~ 1.5 Pa, a 13.56 MHz radio-frequency power source was switched on at 1000 W to ignite the plasma. The carbon fibre fabric was pre-treated with Ar plasma for 10 mins to ensure a better adhesion between VGs and carbon fibres. Once the plasma pre-treatment was complete, H_2 and CH_4 gases with a flow rate of 20 sccm were introduced into the chamber for another 10 mins to deposit VGs on carbon fibres [36]. By adjusting the deposition distance, plasma density, gas flow rate or the deposition time, the radial height of VGs deposited on carbon fibre surface can be changed [36]. Basically, a taller VG deposition will enable a higher volume fraction of conductive fillers in the carbon fibre reinforced composite, thus potentially leading to a larger electrical conductivity improvement. As the focus of this work is to investigate the influence of hybridizing VG and AgNWs on the electrical conductivity of carbon fibre composites, all the VG deposited carbon fibre fabrics are prepared under the same conditions for the purpose of simplification and consistency.

2.3. Synthesis of AgNWs

AgNWs were synthesized via a polyol method as described in [38] and [39]. In brief, 5.86 g Polyvinylpolypyrrolidone (PVP) was dissolved in 190 mL glycerol in a flask at 90 °C and then cooled to 50 °C, before adding 1.58 g $AgNO_3$ powder into the solution. A separate solution containing 10 mL glycerol, 59 mg NaCl, and 0.5 mL H_2O was prepared and subsequently decanted into the flask. After gentle stirring (~50 rpm) and heating the resultant mixture to about 210 °C, the reaction was stopped by transferring the final hot grey–green solution into cold Milli-Q water. After allowing the solution to settle for 24 h, the supernatant fluid was removed and the AgNWs sedimented at the bottom were collected. This procedure was repeated three times to remove the excess PVP and Ag nanoparticles from the solution, and the resultant AgNWs were dispersed in ethanol for storage.

2.4. Fabrication of VG/AgNWs modified CFRP laminates

The VG/AgNWs modified CFRP laminates were manufactured by incorporating VG deposited carbon fibre fabrics with AgNWs dispersed epoxy resin. A schematic of the whole manufacturing procedure is

presented in Fig. 1. The carrier solvent of AgNWs, ethanol was evaporated off using a hot-plate prior to dispersion of AgNWs in acetone. The resultant AgNWs/acetone dispersion was then added to the epoxy resin. At last, acetone was removed via simultaneous mechanical mixing (400 rpm) and thermal heating (90 °C) for 1 h. The AgNWs were dispersed uniformly in the epoxy resin assisted by a 30 mins ultrasonication treatment using a digital ultrasonic water bath.

Two VG-grafted carbon fibre fabrics were then used to fabricate composite with the AgNW-modified epoxy using a wet lay-up procedure. Initially, the AgNWs-modified epoxy resin was brushed onto a release film, which was placed on a stainless-steel plate with pre-applied release agent. One layer VG-deposited carbon fibre fabric was laid on the resin layer. The process of brushing the AgNWs-modified epoxy resin was repeated layer-by-layer to ensure full wetting. After both VG deposited carbon fibre fabrics were wet-out completely, a release film was placed on top of the ply stack. The resultant laminate was then compressed using a hot-press with a pressure of 100 kPa, and then left for curing at room temperature for 24 h. Subsequently, the laminate was post cured at 120 °C for 14 h to maximise the CFRP laminate's mechanical properties.

2.5. Characterization

The surface morphologies of the carbon fibre fabrics before and after the deposition of VGs and the synthesized AgNWs were characterized by Scanning Electron Microscope (FEI Nova NanoSEM 450). The Raman spectra of pristine and VG-deposited carbon fibres were collected via a Raman spectroscopy (Renishaw inVia) using a laser wavelength of 514 nm and a spot of ~ 1 μm. An optical microscope (ZEISS Axio Zoom.V16) was used to examine the dispersion of various loadings of AgNWs in the epoxy resin.

Electrical conductivity measurements were conducted on both dry carbon fibre fabrics and cured composite laminates. The dry carbon fabrics were tightly held between two flat stainless-steel plates as electrodes by an electrode clamp that provided a clamp force of approximate 5 N. The electrodes were connected to a Keysight 34465A digital multimeter to measure the electrical resistance. Five samples measuring 16 mm × 16 mm were tested to obtain the mean value of electrical conductivity. For cured composite laminates, samples containing various concentration of AgNWs (ranging from 0.05 wt% to 0.5 wt%) were cut to the size of 10 mm × 10 mm and measured to ascertain the synergistic effect of VGs and AgNWs in terms of the in-plane and through-thickness electrical conductivities. For comparison, the four different material systems of CFRP laminate samples are denoted respectively as CF/Epoxy, CF/Epoxy + AgNWs, CF + VG/Epoxy, and CF + VG/Epoxy + AgNWs. Silver paste was applied on both sides of the cured laminate samples to reduce the contact resistance and the test setups are shown in Fig.S1.

Mechanical properties were measured through tensile and four-point bending tests using an Instron 3369 universal testing machine with 1 kN load cell. Strength and modulus of the four material systems were measured. The strain of the samples during tensile testing was monitored by a dynamic extensometer Instron 2620–601. For the tensile test, 60 mm × 10 mm specimen with two aluminium tabs (15 mm × 10 mm) bonded on each end were prepared, and 5 samples were tested for each material system. For the four-point bending test, 50.8 mm × 12.7 mm specimen were prepared and repeated 7 samples for each group. According to test standards ASTM D3039-17 and ASTM D6272-17, a crosshead speed of 0.5 mm/min was selected for tensile test, while a crosshead speed of 0.9 mm/min and a support span of 25.4 mm were applied for the four-point bending test [40].

3. Results and discussion

3.1. Surface modification of carbon fibres by VG

The surface morphologies of as-received (pristine) carbon fibres and VG deposited/grafted carbon fibres were examined using SEM. Fig. 2a shows a typical pristine carbon fibre surface with several axial grooves clearly visible, which were inherited from the surface morphology of the precursor fibres used for the carbon fibre fabrication [41]. From the magnified micrograph of Fig. 2a, the diameter of the pristine carbon fibre was measured to be ~ 7 μm. Fig. 2b shows the surface morphology of the VG-deposited carbon fibre, where the graphene nanoflakes were radially distributed around the carbon fibre and an open porous network structure of VG is explicitly visible. The diameter of the VG deposited carbon fibre is seen to increase significantly to ~ 24 μm compared to the pristine caron fibre diameters of ~ 7 μm, indicating that deposited VGs were 8.5 μm in height. From the high magnification image of Fig. 2b, the highly porous nanostructure of the VGs can be clearly identified, which is constructed from multiple highly conductive interconnected graphene

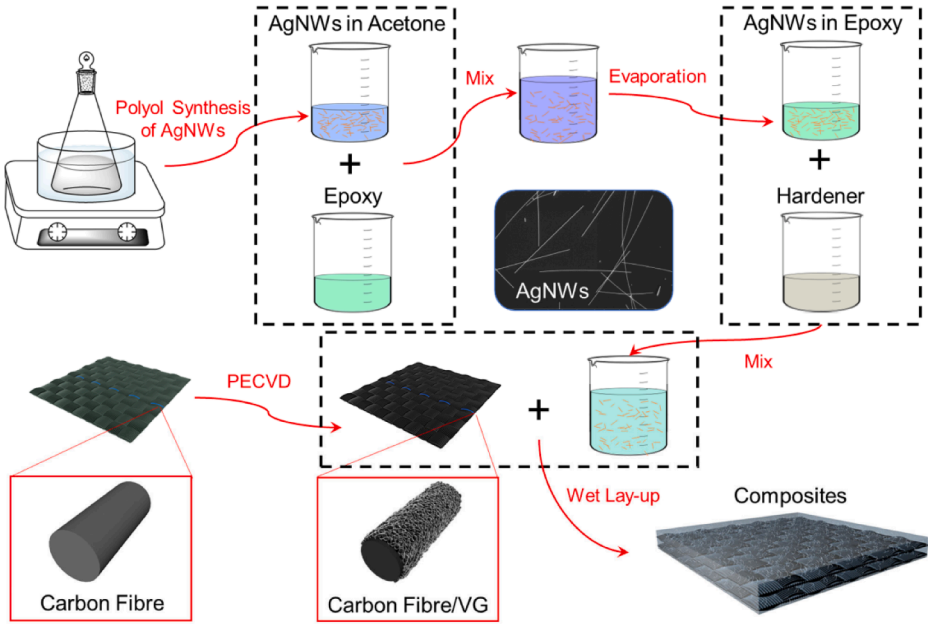

Fig. 1. Hybridization of VG and AgNWs modifications for CFRP laminate.

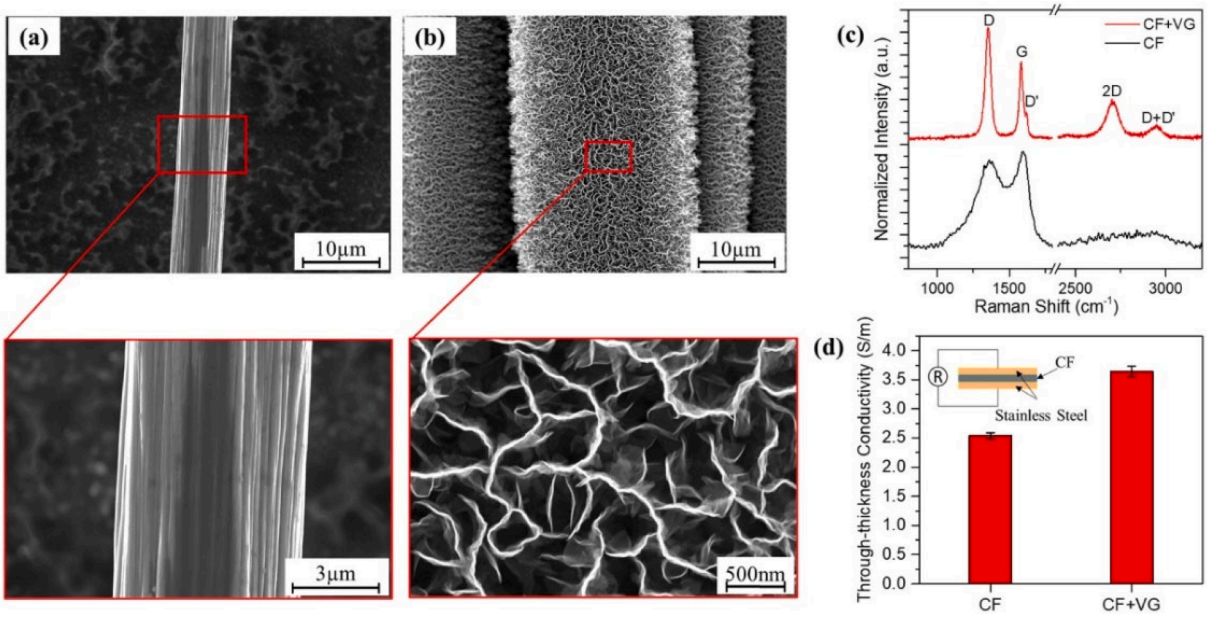

Fig. 2. Functionalization of carbon fibres by VG. (a) SEM image of as-received (pristine) carbon fibre, (b) SEM image of carbon fibre grafted with VG, (c) Raman spectra of pristine carbon fibre and VG functionalised carbon fibre, and (d) through-thickness electrical conductivity of pristine carbon fibre and VG functionalised carbon fibre. The inset sketch shows the setup used for the through-thickness measurement of electrical conductivity.

nanoflakes. The VGs deposited on the carbon fibres not only help to increase the electrical conductivity of the carbon fibre, but also play an important role in improving the carbon fibre/epoxy resin interfacial properties [36].

Fig. 2c shows the Raman spectra of pristine and VG-deposited carbon fibres. Two characteristic peaks, *D* and *G*, which are the superposition of the spectra for the sp^2 carbon layers and the sp^2 carbon clusters [42], can be easily observed from the spectrum of pristine carbon fibre. Correspondingly, except the peak D and peak G, three more peaks D', 2D, and D + D' can be observed from spectrum of the VG-deposited carbon fibres, which are typical characteristic peaks belonging to graphene. Generally, the I_D/I_G and the $I_D/I_{D'}$ ratio can help to determine the defect level and the nature of the defects in graphene, respectively [43]. For the nature of the defects in graphene, an $I_D/I_{D'}$ value of ~ 3.5 indicates boundary-like defects, while this value increases to ~ 7 for vacancy-like defects and to ~ 13 for tetrahedral hybridization associated defects [44,45]. In this work, the I_D/I_G and $I_D/I_{D'}$ ratio of the VG grafted carbon fibre are calculated to be around 1.5 and 6.9 respectively, which indicates a low defect level with a vacancy-like defect type. The I_{2D}/I_G ratio can help determine the type of graphene [46], here the I_{2D}/I_G is calculated to be about 0.5, indicating the samples contain multilayer graphene.

The through-thickness electrical resistances of pristine and VG-deposited carbon fibre fabrics were measured to evaluate the electrical conductivity changes related to the deposition of VGs. During the measurement, a carbon fibre fabric sample (16 mm × 16 mm) was clamped tightly between two flat stainless-steel plates to measure the through-thickness resistance, as shown in the inset of Fig. 2d. The results show that for the pristine carbon fibre fabric sample, the through-thickness electrical resistance is about 0.35 Ω, and it drops to about 0.24 Ω for the VG-deposited carbon fibre fabric. The through-thickness electrical conductivity (σ) of carbon fibre fabric sample can be calculated using the following expression: $\sigma = L/RA$, where L denotes the thickness of carbon fibre fabric (m), R the measured through-thickness electrical resistance (Ω), and A the contact area between carbon fibre fabric and stainless-steel plate (m^2). As shown in Fig. 2d, the deposition of VGs increased the through-thickness electrical conductivity of the carbon fibre-based samples from 2.54 S/m to 3.54 S/m (detail results can be found in Table S1). It is noted that these conductivity results are relatively low, which is mostly due to the specific resistance measuring method applied to the dry fabrics where silver paste was not applied to avoid interfering with the effects of VG deposition. Silver pastes could potentially infuse inside the carbon fabrics and make it challenging to identify the conductivity differences between pristine and VGs deposited carbon fabric. Herein, the conductivity results were measured under the same condition to provide a qualitative comparison between the pristine and VGs deposited carbon fibre fabrics, which indicates that the VGs deposition can improve the through-thickness electrical conductivity of dry carbon fibre fabric by around 39.4 %.

The improvement made by VGs is partly due to the increased volume fraction of conductive filler. However, the VGs deposition is more effective than an equivalent increase in the volume fraction of CF in improving the electrical conductivity, because VG has a highly porous structure with an extremely large specific area, thus electrically connecting carbon fibres at very low concentrations of VG. According to the previous study [40], the mass loading of VGs on carbon fibre fabric is about 0.1 mg/cm^2, which is relatively low compared with the areal density (19.8 mg/cm^2) of the carbon fibre fabric used in this study. However, as can be seen in Fig. 2b, the diameters of the VG-deposited CFs are significantly larger than that of the pristine CFs. The highly porous structure of VG makes it more effective to build conductive networks compared with CFs of the same volume fraction, thus leading to a more significant improvement in the electrical conductivity.

3.2. Matrix modification by AgNWs

The SEM micrographs of the synthesized AgNWs are given in Fig. 3a. The diameter and length of AgNWs are measured to be around 50 ~ 100 nm and 5 ~ 50 μm, respectively. Among the reported methods for using AgNWs to improve the electrical conductivity of composite laminates [35,47,48], directly dispersing AgNWs into the epoxy resin is the simplest and hence was selected for this study. The levels of dispersion of different concentrations of AgNWs in epoxy resin were observed under an optical microscope and are shown in Fig. 3b-3 g. The visible colour of the epoxy resin changed from transparent to grey with the increase in mass loading of AgNWs from 0.05 to 0.5 wt%; the viscosity of the epoxy resin also increased with the mass loading. Another noteworthy phenomenon is that the AgNWs are very difficult to identify in the

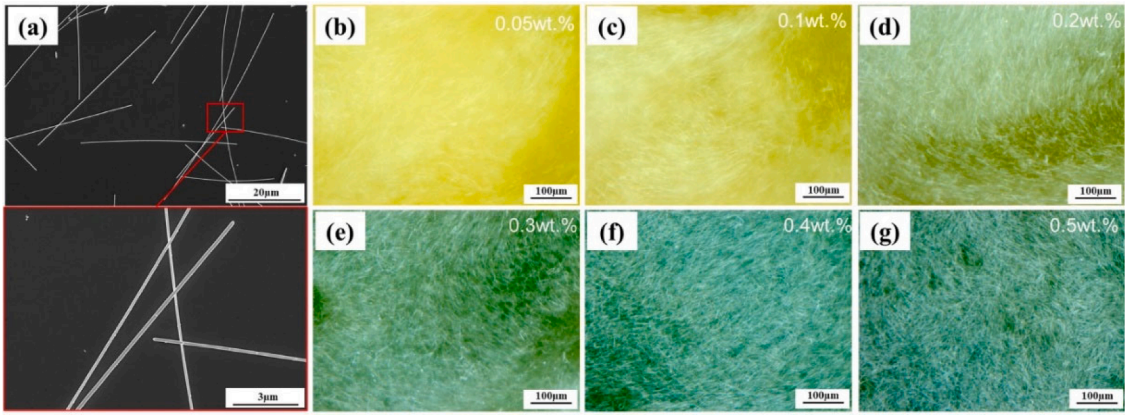

Fig. 3. (a) SEM images of AgNWs; (b-g) Optical microscopic images of the dispersions of AgNWs in epoxy resin with different loadings.

microscopic images of low mass loadings samples, e.g., 0.05 and 0.1 wt %, which is attributed to the excellent optical properties of AgNWs, making AgNWs a favoured candidate to develop transparent conductive components [49]. In high mass loading samples, for example 0.4 and 0.5 wt%, the dispersion of the AgNWs in epoxy resin looks particularly dense, and the individual AgNWs can connect with each other to build conductive networks within the epoxy resin. It is noted that the epoxy resin Raku Tool EL-2203 applied in this work has a relatively high viscosity, and adding concentrations greater than 0.5 wt% AgNWs increases its viscosity beyond what is acceptable for the resin infusion operation. Hence, only composites samples with AgNWs mass loadings of up to 0.5 wt% were investigated, as the resin already exhibited quite a high viscosity at this weight loading.

CFRP laminate samples (10 mm × 10 mm) containing different loadings of AgNWs were fabricated to measure their through-thickness and in-plane electrical resistances, aiming to explore the effect of AgNW content on the electrical conductivity of the composites. The graphic illustrations of the two electrical resistance measurement setups are shown in the inset of Fig. 4a and Fig. 4b. According to the measurements, the through-thickness resistance of the pristine CFRP laminate is around 350.7 Ω. By adding a small amount of AgNWs (0.05 wt%), the through-thickness resistance drops to about 240 Ω. This resistance decreased further to 154.3 Ω when the AgNWs mass loading increased to 0.1 wt%. Generally, with the increase of AgNW loadings, the through-thickness resistance of the CFRP laminate decreases. However, the resistance reduction becomes less significant when the mass loading of AgNWs increases to above 0.4 wt%. For example, the through-thickness electrical conductivity of CFRP laminate containing 0.5 wt% AgNWs is about 77.1 Ω, which is quite close to that of laminates containing 0.4 wt % AgNWs (81 Ω). This trend is similar to the reported results in ref. [50].

Fig. 4a shows the calculated through-thickness electrical conductivity of the CFRP laminate based on the electrical resistance measurements, by simply dispersing 0.5 wt% AgNWs into the epoxy resin, the through-thickness electrical conductivity of the CFRP laminate can be enhanced from 0.0136 S/m to about 0.0618 S/m (detail results can be found in Table S2). Fig. 4b shows the in-plane electrical conductivity calculation results. With the addition of AgNWs to the epoxy, the in-plane electrical conductivity of CFRP laminate can be enhanced as well, though to a less significant extent than when compared with the through-thickness direction. The less noticeable increase of the in-plane conductivity is due to the dominant role of reinforced carbon fibres in determining the in-plane electrical conductivity of the CFRP laminates [51]. The in-plane electrical conductivity of composite laminate rises from roughly 1141 S/m to 1440.7 S/m when 0.5 wt% AgNWs is added to

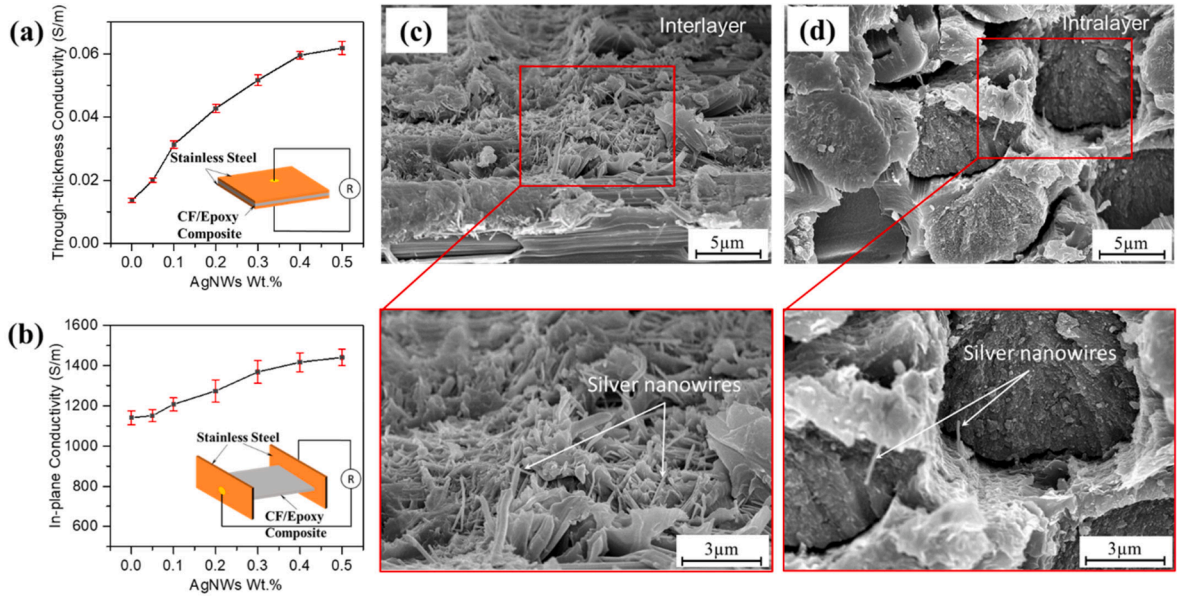

Fig. 4. Carbon fibre reinforced epoxy laminates (CFRPs) with different loadings of AgNWs. (a) through-thickness electrical conductivity, and (b) in-plane electrical conductivity of CFRPs with different loadings of AgNWs. (c-d) SEM micrographs of CFRPs with matrix containing 0.5 wt% AgNWs: (c) the interlayer region, and (d) intralayer region of the CFRPs.

epoxy (detail results can be found in Table S2). Similar to the through-thickness direction, the significance of AgNWs in improving the in-plane electrical conductivity of the CFRP laminate also became less noticeable once its mass loading was over 0.4 wt%.

To examine the distribution of AgNWs in the CFRPs, SEM micrographs were taken of the top and cross-section views. It can be seen in Fig. 4c that most of the AgNWs in epoxy were aggregated in the interlayer region of the CFRP, which is due to the filtration effect of the carbon fibres. For the intralayer region of the CFRP, only a small number of short AgNWs can be identified, as shown in Fig. 4d. Hence, the contribution of AgNWs to the electrical conductivity of CFRPs mainly comes from the interlayer region, which is also the region where resin-rich areas frequently appear, creating conductive paths between layers.

3.3. Performances of hybrid composite laminate modified with VG/AgNWs

3.3.1. Electrical conductivity

The through-thickness and in-plane electrical conductivities of the four material systems of CFRP laminates were measured to evaluate the impact of integrating VGs and AgNWs. Schematics of the microstructures of the four systems are shown in Fig. 5a. As described in section 3.2, adding 0.5 wt% AgNWs into the epoxy can enhance the through-thickness electrical conductivity of the composite laminate from 0.014 to 0.062 S/m, while depositing VGs onto the carbon fibre surface can increase the through-thickness conductivity to about 0.073 S/m. Combining the AgNWs and VG together, however, yields a very dramatic increase in through-thickness electrical conductivity, achieving a through-thickness conductivity of about 0.533 S/m, as shown in Fig. 5b (detail results can be found in Table S3). It is evident that the addition of AgNWs into the epoxy and depositing VGs onto carbon fibres contributed to reductions in the through-thickness electrical resistance of CFRPs.

Importantly, combining these two approaches together can achieve results significantly better than what could be attained by each method individually. When compared with pristine the CFRP laminate, the composite laminate fabricated with 0.5 wt% AgNWs dispersed epoxy resin and VG-grafted carbon fibres has achieved more than 38 times higher through-thickness electrical conductivity. Fig. 5c displays the in-plane electrical conductivity of the four composite material systems (detail results can be found in Table S3). Though not as significant as the improvement in the through-thickness direction, combining AgNWs and VG nevertheless improved the in-plane electrical conductivity of composite laminates by around 39 %. The fundamental explanation for this disparity in conductivity enhancement in the two directions is that the influencing factors that govern conductivity in the two directions are different. The insulating epoxy resins separating carbon fibres put a higher limit in the through-thickness electrical conductivity than the in-plane conductivity. Correspondingly, the carbon fibres dominates the in-plane electrical conductivity of the composite laminates [21], which accounts for a substantial proportion when compared to the loading of VGs and AgNWs. The unmodified CFRP laminate's in-plane electrical conductivity is already tens of thousands of times greater than its through-thickness electrical conductivity [52], making further improvement comparably more difficult. As a result, the electrical conductivity enhancement attributed to the VGs and AgNWs in the in-

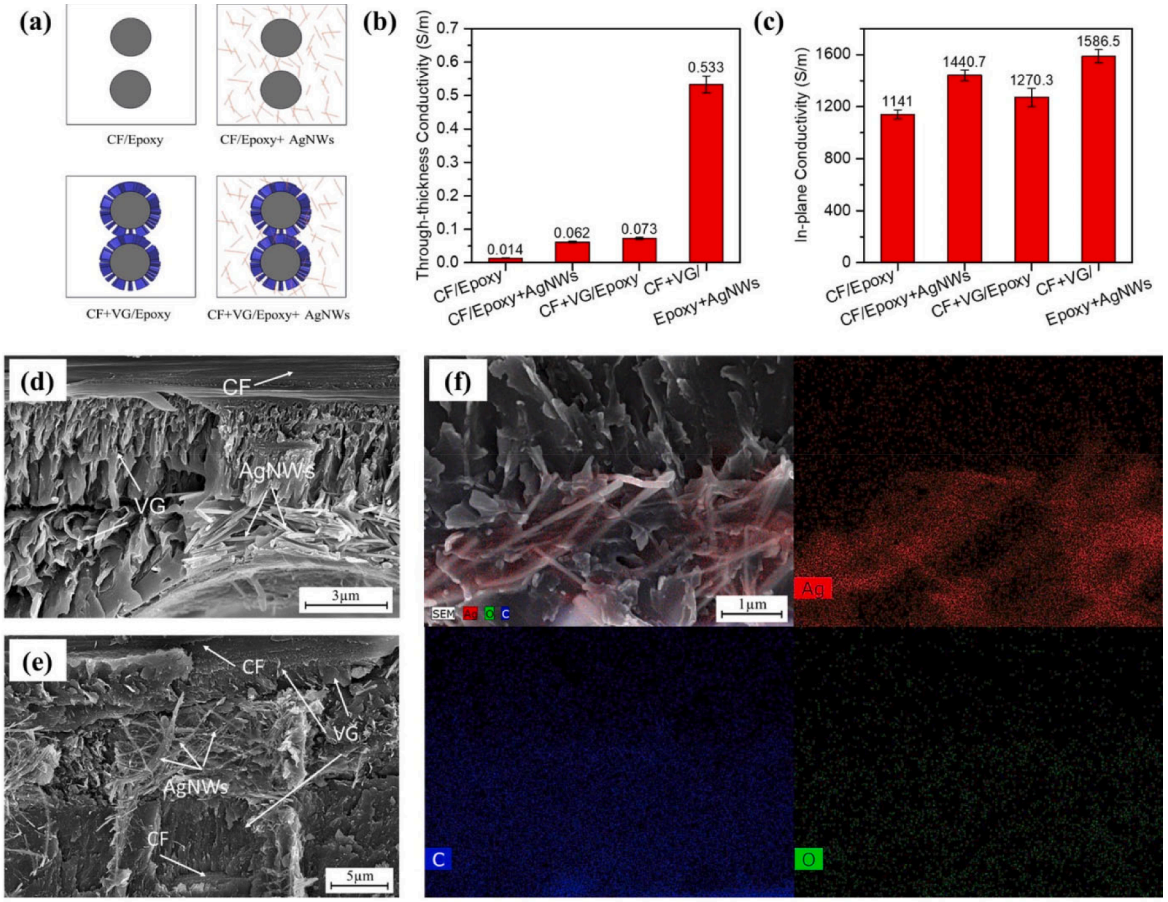

Fig. 5. VG-modified CFRPs containing AgNWs. (a) Schematic of the four composite material systems, (b) through-thickness electrical conductivity, and (c) in-plane electrical conductivity of the four composite material systems. (d-e) SEM micrographs of VG/AgNWs modified CFRP from side view, and (f) energy-dispersive X-ray spectroscopy (EDS) mapping result.

plane direction cannot be as substantial as the one they achieve in the through-thickness direction.

To investigate the mechanism that contributes to the combination of VGs and AgNWs markedly increasing the electrical conductivity of the CFRPs, SEM micrographs were taken from side views, as shown in Fig. 5d and 5e. The VGs and AgNWs together appear to significantly increase the formation of conductive pathways across the layers of carbon fibre fabric, reducing the impact of the insulating epoxy resin on the through-thickness electrical conductivity of CFRP laminate. A similar phenomenon has been reported earlier [35], where both AgNWs and GNPs were dispersed into epoxy resin, and a synergistic effect between the AgNWs and GNPs was observed, enhancing the electrical conductivity of composite laminate. However, the addition of two nanofillers into the resin predictably can increase the resin viscosity, which can create difficulties with the fabrication of the composite laminates at high mass loadings. The elemental composition of the VGs and AgNWs modified CFRPs was determined using energy-dispersive X-ray spectroscopy (EDS), as shown in Fig. 5f. The element of silver is clearly visible among a variety of other elements, confirming the feasibility of the polyol technique used to synthesise the AgNWs in this study.

Finite element analysis (FEA) has been performed using COMSOL Multiphysics to help understand the mechanisms contributing to the increased electrical conductivity by combining VGs and AgNWs. Fig. 6a shows two simplified FEA models of current density within the CF/Epoxy and CF + VG/Epoxy + AgNWs composites, and it is clearly visible that the combination of VG and AgNWs can provide additional current flow paths and higher current densities within the composites. The increase in through-thickness electrical conductivity can be visualised by the rainbow plots of potential distribution in the composites, as shown in Fig. 6b. As the CF/Epoxy composite has a higher through-thickness resistance than the CF + VG/Epoxy + AgNWs composite, the potential difference between its two sides will also be higher for the same current input. In the present study, the epoxy resin containing randomly oriented fillers was treated as uniform solid subject during the FE modelling, and the composite laminate was simplified as two layers of carbon fibre fabric and a thin layer of epoxy resin, where the thin epoxy resin layer in the middle represents the resin rich region, as shown in Fig.S2. With this simplification, the equivalent through-thickness resistance was represented by three resistors in series, and the conductivity of the different materials applied in this FEA were obtained based on the experimental results, as shown in Table S4. It is noted that the fibre distribution in the FEA models were set to achieve a similar conductivity enhancement to the experimental results, to develop a better understanding the cooperation mechanism between the VG and AgNWs.

The following equation can be used to quantify the synergistic effect between the VG and AgNWs in increasing the electrical conductivities of CFRPs.

$$S = \left(\frac{\Delta_{Hybrid}}{\Delta_{VG} + \Delta_{AgNWs}} - 1\right) * 100\% \quad (1)$$

where S represents the synergy ratio, $\Delta_{Hybrid} = \sigma_{Hybrid} - \sigma_{CF}$, $\Delta_{VG} = \sigma_{VG} - \sigma_{CF}$, $\Delta_{AgNWs} = \sigma_{AgNWs} - \sigma_{CF}$. Here σ_{Hybrid} denotes the electrical conductivity of the CF + VG/Epoxy + AgNWs composites, σ_{VG} is the electrical conductivity of the CF + VG/Epoxy composites, σ_{AgNWs} is the electrical conductivity of the CF/Epoxy + AgNWs composites, and σ_{CF} is the electrical conductivity of the pristine CF/Epoxy composites. All these electrical conductivity values are plotted in Fig. 6c and Fig. 6d. It is calculated that the synergy ratio between VG and 0.5 wt% AgNWs in enhancing the through-thickness electrical conductivity of CFRPs is 385.5 %, while in terms of improving the in-plane electrical conductivity, the synergy ratio is 3.84 %. Moreover, it is observed that synergy ratio for through-thickness conductivity increases with the AgNWs mass loading, while for the in-plane conductivity, the synergy ratio remains insignificant with varying AgNWs mass loadings. It is believed that the synergy ratio for the through-thickness conductivity is likely to further increase to some extent when the AgNWs mass loading exceeds 0.5 wt% as there will be more conductive pathways constructed by the VG and AgNWs. While for the in-plane direction, the synergy ratio will remain at a similar level. This significant difference in the synergy ratio between these two measurement directions reflects that the in-plane electrical conductivity is dominated by the continuous carbon fibres.

To further evaluate the through-thickness electrical conductivity

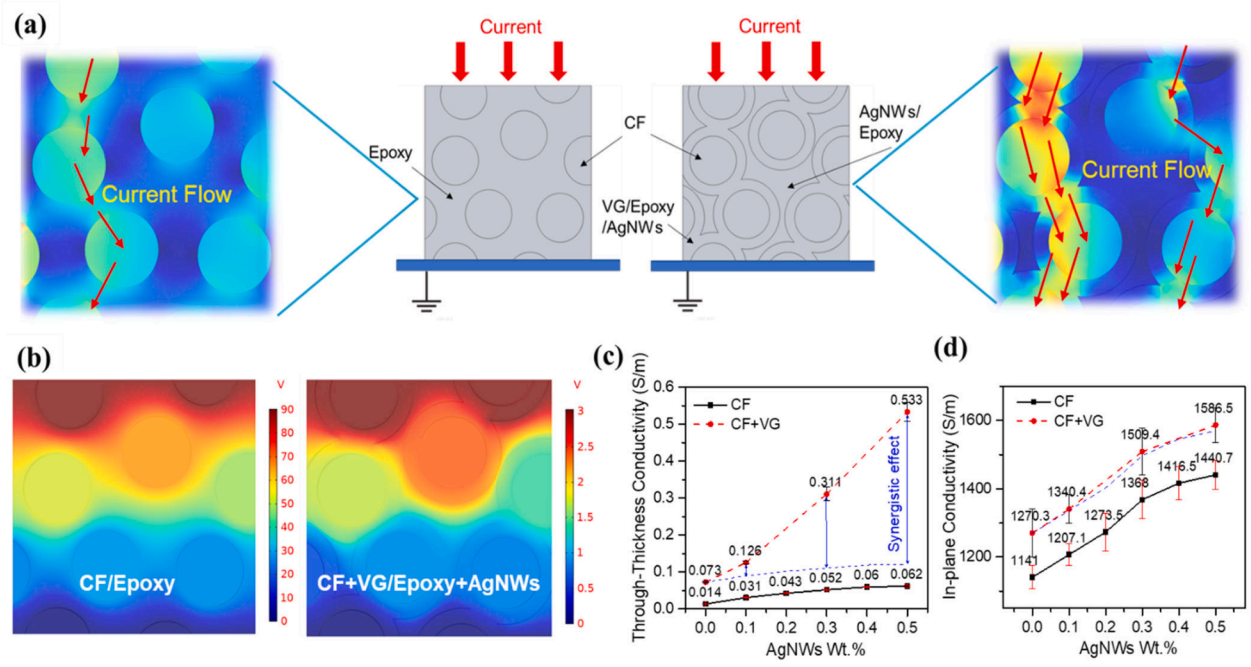

Fig. 6. (a) FEA models of CF/Epoxy and CF + VG/Epoxy + AgNWs composites, and the current flows inside the composites, (b) potential distributions in the composites, noting that the potential value may change with current input; (c-d) Synergistic effect between VG and AgNWs in increasing the through-thickness electrical conductivity, and in-plane electrical conductivity (the blue dash curves refer to the sum of the individual conductivity improvements from VG and AgNWs). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

improvement achieved in this work, the results are compared with previously published studies. Due to the different measurement methods and composites fabrication techniques, the measured absolute conductivity values can vary significantly [35]. Take the pristine CF/Epoxy composites as an example, the different types of carbon fibre fabric and epoxy resin (e.g., conventional carbon fabric or prepreg), different experimental procedures by researchers (e.g., applying or not applying silver paint on the sample surface), and different fabrication techniques (e.g., wet lay-up, vacuum-assisted resin transfer moulding (VARTM), autoclave curing, etc.) can lead to a significant variation of baseline through-thickness electrical conductivity from 0.015 S/m [53] to 13 S/m [56]. Moreover, the measurement method can also significantly influence the conductivity values obtained. Generally, the four-probe method described in ref. [58] is considered more suitable to obtain high precision results. However, limited by the sample size, a simpler two-probe method was employed in this study. Therefore, here we compare the relative improvements in the through-thickness electrical conductivity rather than the absolute conductivity values. Table 1 shows a comparison with literature of the through-thickness electrical conductivity of composites, and it is clearly seen that this work has achieved the largest relative improvement in the through-thickness electrical conductivity of composites. It is also noted that by using VG or AgNWs alone to modify the CFRPs, the through-thickness electrical conductivity can be improved to 4 ~ 5 times of the value of pristine CF/Epoxy composites, and such a degree of conductivity improvement is already comparable to some published literature.

3.3.2. Mechanical performances

Tensile tests were performed on the four material systems: CF/Epoxy, CF/Epoxy + AgNWs, CF + VG/Epoxy, and CF + VG/Epoxy + AgNWs; the test equipment and samples are illustrated in Fig. 7a. The tensile strength and modulus of each test group are shown in Fig. 7b-7c. According to these results, adding AgNWs to epoxy or depositing VG onto carbon fibres did not result in a substantial change in the composite tensile strength. Even when VGs and AgNWs were combined, the tensile strength enhancement was only around 4.3 %, suggesting that these two nanomaterials have a relatively minor impact on the CFRP's tensile properties. It has been reported in previous studies [36,40] that the VG deposition on carbon fibre surface has negligible influence on the tensile properties. This phenomenon may be related to the radially distributed growth morphology of the VG on the carbon fibre surface, which does not provide much strengthening to the carbon fibre along the fibre direction. However, unlike directly mixing graphene nanoplatelets (GNP) with epoxy resin, where the π–π interactions among GNPs can result in multi-layer agglomerates of graphene in epoxy, reducing the composite tensile strength [35,59,60], grafting VG onto the carbon fibre surface prevents the graphene from agglomerating in the epoxy resin, thus avoiding significant degradation to the CFRP's tensile strength. In addition, although not obvious, the PECVD process has the potential to slightly degrade the strength of carbon fibre, however, this degradation may be offset by the nano reinforcing effect of the VG and AgNWs, and hence results in a similar tensile strength value as the pristine CF/Epoxy composite. The tensile modulus results, as shown in Fig. 7c, also showed no significant differences, which is most likely owing to the comparatively modest mass loading of nanomaterial in composites.

Four-point bending tests were conducted to characterise the flexural properties of CFRPs, the test setup and test samples are shown in Fig. 7d. The flexural strength and modulus of each test group can be found in Fig. 7e-7f. There was a small increase in the composite flexural strength (4.8 %) when 0.5 wt% AgNWs were added to the epoxy matrix. However, no substantial variation in the flexural modulus was observed, related to the modest mass loading of AgNWs. Comparatively, grafting VGs onto the carbon fibre surface results in a more noticeable improvement in the composite flexural properties; the composite flexural strength and modulus of the composites increased by roughly 15.4 % and 8.9 %, respectively. The specific growth pattern of the VGs on the carbon fibres, where the graphene nanoflakes are aligned perpendicularly to the carbon fibre surface, is principally responsible for these flexural property improvements [36,61]. During the four-point bending test, this growth pattern means that a substantial number of graphene nanoflakes are aligned with the loading direction. The epoxy resin is efficiently strengthened by these graphene nanoflakes exhibiting high in-plane strength and rigidity, which increases the composite flexural strength and modulus. A slight decrease (~2%) in flexural strength was observed after the further addition of AgNWs into the VG modified CFRPs. These flexural test results were compared with those of the CF + VG/Epoxy composite using an independent samples T-test to determine the statistical significance. According to the T-test results, the flexural strengths of the sample groups of CF + VG/Epoxy ($M = 640.9$, $SD = 49.8$) and CF + VG/Epoxy + AgNWs ($M = 627.8$, $SD = 32.5$) were not statistically different ($P = 0.571$). In comparison to the control group (CF/Epoxy) however, the CFRP's flexural strength and modulus rose by 13.1 % and 11.6 %, respectively, after introducing VGs and AgNWs simultaneously.

3.3.3. Finite element (FE) analyses of lightning strike

Finite element analysis is an effective method to investigate the influence of lightning strike on composite structures [62,63]. Here the FEA was conducted to show the influence of the through-thickness electrical conductivity changes on a composite laminate subjected to lightning strikes. The FE model of composite laminate was designed with dimensions of 50 mm × 50 mm × 0.8 mm, which comprised of three carbon fibre layers and two resin rich layers. Electrical conductivities of each component were acquired from the experimental measurements, as

Table 1
Literature comparison on the through-thickness electrical conductivity of composites.

Modification Method	Nanomaterials	Fabrication Technique	Original Conductivity (S/m)	Improved Conductivity (S/m)	Relative Improvement	Ref.
Matrix Modification	GO	VARTM	5.4	18	233.3 %	[21]
	CNT	Wet Lay-up	2.4	21	775 %	[24]
	GNP/AgNWs	Wet Lay-up	0.077	0.3	290 %	[35]
	CNT	Wet Lay-up	0.015	0.108	620 %	[53]
	CNT	Autoclave Curing	0.76	1.82	140 %	[54]
	Graphene	Hot-press	0.12	0.37	208 %	[55]
Fibre Modification	CNT	VARTM	0.16	0.68	330 %	[29]
	CNT	VARTM	7.6	57.3	654 %	[30]
	RGO	VARTM	0.4	1.4	250 %	[31]
	GNP	Autoclave Curing	2.5	6.6	165 %	[32]
	GO	VARTM	13	32	146 %	[56]
	Cu/CNT	VARTM	0.0065	0.186	2761 %	[57]
Hybrid Modification	GNP, CNT	Wet Lay-up & Autoclave Curing	6	83.3	1288 %	[34]
	VG/AgNWs	Wet Lay-up	0.014	0.533	3707 %	This work

[a] CB/CC is the abbreviation of carbon black / copper chloride.

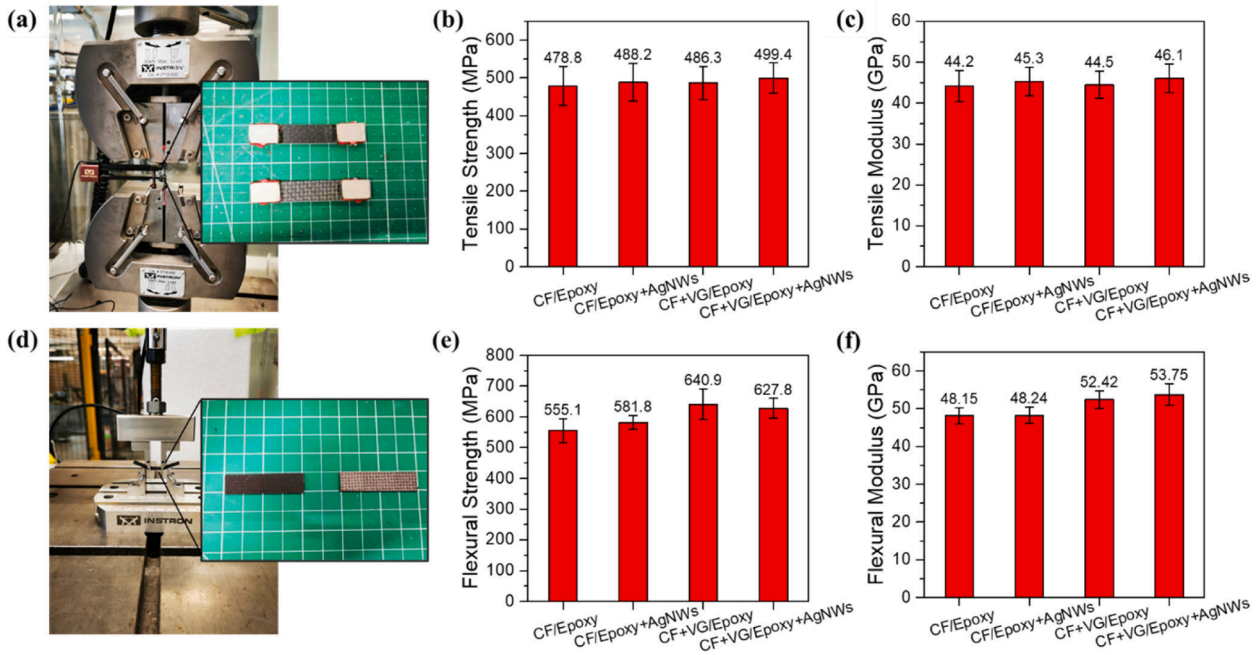

Fig. 7. (a) Tensile test setup and test samples, (b-c) tensile strength and modulus of the different test groups, (d) four-point bending test setup and test samples, (e-f) flexural strength and modulus of the different test groups.

shown in Table S5. The simulations were conducted using the electrostatics module in COMSOL Multiphysics software, and the lightning strike was simulated by applying a 2kA current at the centre point of the composite laminate. It is noted that the FE simulation here is only a qualitative analysis, and the current applied in modelling is much less than that applied during a lightning strike test. During the simulation, the bottom plane of the composite laminate was grounded. As the dimension of laminate in its thickness direction is relatively small, to assist the observation of simulation results, Fig. 8 shows the electric potential distribution and electric energy density of a cross-section close to the centre of model, which was selected to demonstrate the benefits of the electrical conductivity improvement achieved by integrating VG and silver nanowires. It is clearly seen that by adding highly conductive VG and silver nanowires into the composite laminate, the maximum

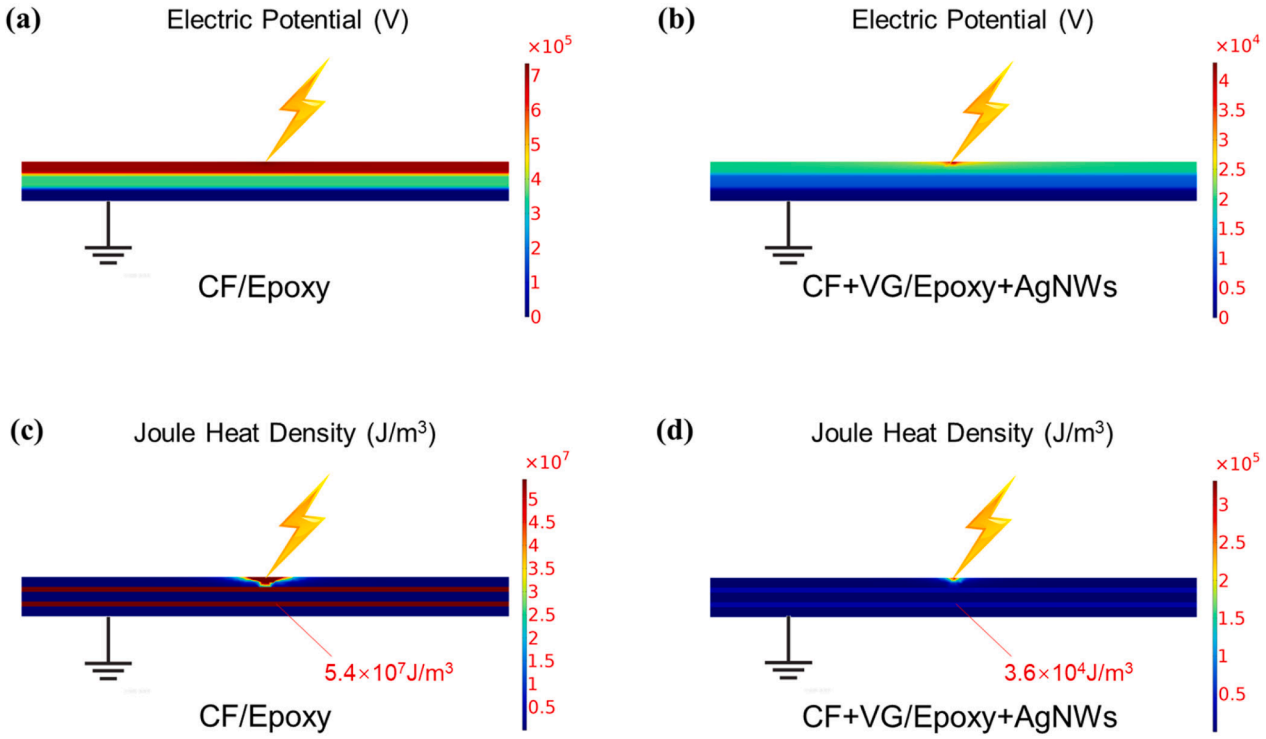

Fig. 8. Cross section of the finite element simulation results of composite laminates subjected to lightning strike: electric potential distribution of the (a) CF/Epoxy composite laminate and (b) CF + VG/Epoxy + AgNWs composite laminate; electric energy density of (c) CF/Epoxy composite laminate and (d) CF + VG/Epoxy + AgNWs composite laminate.

electrical potential between the top and bottom plane generated by a lightning strike can be reduced to around one seventeenth of what it was prior to modification. An even more significant reduction was observed in the simulation results of the electric energy density, which reflects the amount of joule heat generated by a lightning strike on the composite laminate. It is shown that with improved electrical conductivity in the through-thickness direction, the joule heat energy density in the resin rich regions of the composite laminate can be reduced to less than one thousandth of what it was without modification, which is beneficial to the composite structure to maintain its structural integrity after lightning strike.

4. Conclusion

In this paper, we present a novel method for improving the electrical conductivity of CFRPs by combining matrix enhancement with fibre functionalisation. The epoxy resin was specifically modified by AgNWs via direct mixing, while the carbon fibres were modified by grafting VG via the PECVD technique. The electrical conductivity results reveal that combining AgNWs and VG together, the CFRP's through-thickness electrical conductivity can be improved by over 38 times, which is significantly greater than state-of-the-art values in existing reported studies. In a similar manner, the CFRP's in-plane electrical conductivity has been enhanced by 39 %. The addition of VG and AgNWs into the CFRPs does not significantly alter the tensile properties of the composites, however it does improve the flexural strength and modulus by 13.1 % and 11.6 %, respectively. With the improved electrical conductivity in the through-thickness direction, the joule heat energy density on the resin rich region of composite laminate under lightning strike can be reduced by around one thousand times, which is greatly beneficial to the composite structure to maintain its structural integrity after lightning strike. These findings show the benefits of adopting a hybrid modification approach and hybrid conductive nano additives to improve the electrical conductivity of CFRPs.

CRediT authorship contribution statement

Zhao Sha: Conceptualization, Methodology, Investigation, Validation, Writing – original draft. **Xinying Cheng:** Writing – review & editing. **Mohammad S. Islam:** Methodology, Writing – review & editing. **Pichsinee Sangkarat:** Investigation. **Wenkai Chang:** . **Sonya A. Brown:** Writing – review & editing. **Shuying Wu:** Resources. **Jin Zhang:** Writing – review & editing. **Zhaojun Han:** Resources, Methodology, Writing – review & editing. **Shuhua Peng:** Visualization, Writing – review & editing. **Chun H. Wang:** Conceptualization, Methodology, Supervision, Writing – review & editing.

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.

Data availability

Data will be made available on request.

Acknowledgement

The authors are grateful to Electron Microscope Unit of UNSW for the use of facilities. This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.compositesa.2023.107463.

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