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A review of electrical and thermal conductivities of epoxy resin systems reinforced with carbon nanotubes and graphene-based nanoparticles

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ABSTRACT

Epoxy (EP) resins exhibit desirable mechanical and thermal properties, low shrinkage during cuing, and high chemical resistance. Therefore, they are useful for various applications, such as coatings, adhesives, paints, etc. On the other hand, carbon nanotubes (CNT), graphene (Gr), and their derivatives have become reinforcements of choice for EP-based nanocomposites because of their extraordinary mechanical, thermal, and electrical properties. Herein, we provide an overview of the last decade's advances in research on improving the thermal and electrical conductivities of EP resin systems modified with CNT, Gr, their derivatives, and hybrids. We further report on the surface modification of these reinforcements as a means to improve the nanofiller dispersion in the EP resins, thereby enhancing the thermal and electrical conductivities of the resulting nanocomposites.

1. Introduction

Nanofillers are increasingly being used in polymers to tailor their properties for a target application. The resulting polymer nanocomposites (PNCs) have been shown to exhibit much higher mechanical, electrical, thermal, and multifunctional properties than virgin polymers [1–13]. Owing to their desirable physical and chemical properties, epoxy (EP) resins have been extensively used in a wide range of applications, such as surface coatings, adhesives, electronic devices, and as composite matrices in shipbuilding, aerospace, and automotive structures [14–24].

Despite their good mechanical performance, EP resins suffer from poor thermal [25–29] and electrical conductivities [30–34]. Today, the demand for thermally and electrically conductive EP resin systems in applications such as semiconductors is on the rise. The reason behind this is that electronic systems require efficient heat dissipation, which is achieved by using composites with excellent thermal conductivity [35–38]. In general, the main strategy to improve the electrical and thermal properties of EP resins has been to incorporate a second conductive phase.

Due to their inherently excellent electrical and thermal properties, carbon nanotubes (CNT), graphene (Gr), and their derivatives have been promising candidates to increase the electrical and thermal conductivities of EP-based nanocomposites [39–45]. For instance, Fogel et al. [46] prepared EP/multiwalled CNT (MWCNT) nanocomposites using a three-roll mill and reported an enhancement in the DC electrical conductivity of EP by 11 orders of magnitude. In addition, the percolation threshold (PT), which is defined as the minimum content of filler needed to form a three-dimensional conductive network within the polymer matrix, was 0.25 wt% CNT and the maximum electrical conductivity was 0.006 S/m for EP containing 0.75 wt% CNT. In another research by Imran et al. [47], the electrical conductivity of an EP resin was increased by incorporating Gr. They prepared the EP/Gr nanocomposites by a three-roll mill dispersion technique and concluded that the PT of the Gr

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is 1 wt%. The addition of Gr caused an increase in the electrical conductivity of the EP from 4.3×10^{-15} to 2.6×10^{-6} S/m at his weight fraction. Moreover, adding more Gr to EP up to 2 wt% improved the electrical conductivity of EP to a high value of 3.1×10^{-4} S/m. At the same time, the thermal conductivity was increased from 0.14 W/mK for the neat EP to 0.27 W/mK for the EP containing 1 wt% Gr. Wang et al. [48] prepared the EP/CNT/GO nanocomposites, in which GO acted as a non-covalent dispersant for CNT in the EP resin and as a reactive, secondary reinforcing filler to improve the thermal conductivity of EP resin. The thermal conductivity of neat EP was about 0.6 W/mK, which was enhanced to around 0.19 W/mK when 4 wt% hybrid filler was incorporated in the EP.

Several review papers have been written on enhancing the properties of the EP resin systems using CNT and Gr. For instance, Kausar et al. [49] presented a comparative review on the mechanical, thermal, and electrical properties of EP resins reinforced with Gr and its derivatives. Atif et al. [50] provided a review on the effects of key factors such as morphology, surface functionalization, dispersion state, and weight fraction of Gr on the mechanical, thermal, and electrical properties of EP-based nanocomposites. Chen et al. [51] reviewed the fundamental design principles of highly thermally conductive composites. They addressed the critical factors affecting the thermal conductivity of polymers and summarized the properties of thermally conductive fillers such as carbon nanotubes, metal particles, and ceramic particles. Burger et al. [52] presented a theoretical and experimental review on the aspects of thermal conductivity in composites. They studied the fundamental mechanism of thermal conduction, its mathematical aspects, and specific essential parameters, including crystallinity, phonon scattering, or filler/matrix interfaces, to investigate their effect on thermal conductivity. Imtiaz et al. [53] wrote a review paper, discussing the fabrication, properties, and applications of CNT-reinforced polymers and EP-based nanocomposites. Kulkarni et al. [54] reviewed the effects of covalent and non-covalent surface modification of Gr on the mechanical, thermal, and electrical properties of EP/Gr nanocomposites. Kumar et al. [55] presented a review of recent studies on improved thermal, mechanical, and interfacial properties of polymers (including EP, etc.) reinforced with CNT and Gr. In another review paper, Kumar et al. [56] examined the status of the molecular modeling, simulation, and mechanical and thermal characterization of Gr and the properties of polymeric nanocomposites via molecular dynamics. Mousavi et al. [57] reviewed research progress from 2015 to 2021 on the toughening of EP resins using CNT, Gr, and CNT-Gr hybrids.

Although there are some limited review papers investigating the advances in properties of EP resin systems reinforced with CNT and Gr-based nanofillers, they suffer from weak literature reviews regarding the thermal and electrical conductivities of EP resins filled with CNT and Gr-based nanofillers. The present review paper covers most of the published literature on the effects of CNT, Gr, and their derivatives on the electrical and thermal conductivities of the EP resin systems. Such a high-level comparative review of the recent case will help the researchers choose the best CNT or Gr-based nanofillers and optimal possessing conditions to achieve desired electrical and thermal conductivities for the EP-based nanocomposites.

2. Nanocomposites

2.1. Polymer-based nanocomposites

Polymer-based nanocomposites have drawn significant interest due to their outstanding properties compared to neat polymers or even conventional micro or macro composites. To produce a conventional composite, a large amount (>10 wt%) of inorganic fillers is required to achieve favorable properties. Such a large filler content increases the density and consequently deteriorates the final product's properties via interfacial immiscibility between the organic polymer and inorganic filler. Additionally, increasing the filler content has a negative effect on processability. In contrast, nanocomposites exhibit improved properties at low weight fractions (≤5 wt%) of nanofillers. Polymer-based nanocomposites are commonly defined as the polymer matrix reinforced with nanofillers possessing at least one dimension in the nanoscale [58–64].

2.2. Epoxy-based nanocomposites

Epoxy (EP) resins are described as low molecular weight prepolymers that have at least two epoxide groups [65]. They are the most common and extensively utilized thermosetting resins for electronic circuit board materials, semiconductors, hardware components, aerospace structures, coatings, adhesives, encapsulation materials, and composite matrices due to their desirable performance such as high tensile strength, exceptional chemical resistance, good electrical properties, and high stiffness. To improve the electrical and thermal conductivities, mechanical performance, flame retardancy, optical and magnetic properties of EP resins, different types of nanofillers and polymeric additives such as CNT, Gr, iron oxides, nanosilica, nanoclays, titanium oxide, zinc oxide, and polyaniline have been used [66–74]. These properties of the EP-based nanocomposites are affected by many variables such as the filler content, nanoparticle dispersion in the EP resin, shape and size of the nanofillers, and the adhesion of the nanofillers to the EP resin [22,75,76].

3. Carbon nanotubes and graphene-based nanoparticles

Recently, interest in carbonaceous nanomaterials in the form of nanofibers, nanotubes, and nanosheets have been on the rise because they play a key role in different industries such as energy, construction, transportation, aerospace, and environmental science. The reason behind the use of carbonaceous nanomaterials in the mentioned areas of applications is their electron affinity, versatility, high strength, and significant electrical and thermal conductivities [77,78]. Among different carbonaceous nanomaterials, CNT and Gr nanosheets are among the most important ones for improving the electrical and thermal conductivities of EP resins due to the below-mentioned advantages that they offer. CNT display outstanding thermal [79,80], electrical [81,82], and mechanical [83,84] properties, making them promising candidates for a wide variety of applications [85–87]. CNT are carbon sheets that have been rolled into a tube whose properties are directly affected by the way the carbon sheets are wrapped around. It is also worth mentioning that CNT possesses approximately one-dimensional structures due to their high aspect (length-to-diameter) ratios. Moreover, their most important structures include single-walled nanotubes (SWCNT) and multi-walled nanotubes (MWCNT). An SWCNT has a cylindrical shape with only one wrapped graphite sheet, while an MWCNT resembles a collection of concentric SWCNT [88,89].

Gr is a monolayer sheet of graphite and correspondingly has a two-dimensional structure. In addition to its significant mechanical properties, Gr exhibits excellent thermal properties, especially thermal conductivity (5,000 W/mK); hence, it is considered a precious material for thermal management applications in microelectronic and mechanical devices [55,90]. Moreover, categorization of Gr-based nanomaterials can be carried out based on the number of layers stacked in a sheet, i.e., mono-layer, bi-layer, and few-layer. One of the most utilized derivatives of Gr is graphene oxide (GO), which has a lower production cost, larger-scale production, and easier processing [91,92].

The electrical and thermal conductivities of EP resins can be significantly enhanced using CNT and Gr as conductive nanofillers. Most EP resins exhibit a low electrical conductivity of around 10^{-9} S/m [93] and thermal conductivity of 0.17–0.21 W/mK [94], while CNT and Gr have electrical conductivities of approximately 10^{6}-10^{7} [95] and 10^{8} [95] S/m, respectively. Moreover, their thermal conductivities are about 2, 800–6,000 [96] and 1,500–5,000 W/mK [97], respectively. In Fig. 1, the number of published research articles on the electrical and thermal conductivities of EP-based nanocomposites in the last decade are shown.

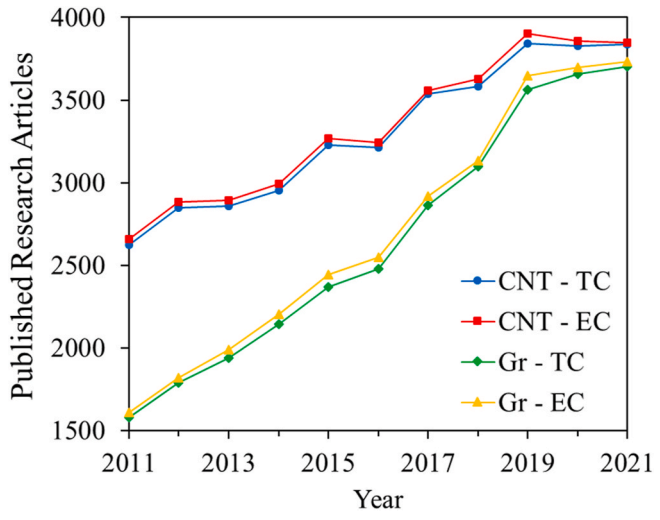

Fig. 1. Published research articles on the thermal conductivity (TC) and electrical conductivity (EC) of EP-based nanocomposites from 2011 to 2021 (based on Web of Science).

Clearly, the interest of researchers in improving the electrical and thermal conductivities of the EP resin systems by incorporating CNT and Gr-based nanoparticles has been increasing year by year. Moreover, a comparison of the number of published works in this field reveals that researchers are more interested in using Gr than CNT to improve the electrical and thermal conductivities of the EP resins.

4. Mechanisms of electrical and thermal conductivities in carbon nanotubes and graphene-based nanoparticles

Incorporating conductive nanofillers into an isolating polymer matrix can induce electrical conductivity and improve thermal properties at very low filler concentrations. The thermal conductivity of polymer composites is one of the vital properties for various applications but still has a long way to be explored thoroughly. Since most polymers indicate a relatively poor thermal conductivity, it is of particular importance to achieve an enhancement for some applications. Because of the consistency of carbon atoms assembled in a graphitic structure/lattice, CNT and Gr, reveal a high thermal conductivity, as their values were mentioned in Section 3. Therefore, by adding CNT and Gr, a certain improvement of the thermal conductivity of polymer based nanocomposites can be obtained [98–100]. It is believed that thermal energy transport in CNT and Gr occurs via a phonon conduction mechanism. As seen in Fig. 2, the thermally conductive nanofillers connect and form a continuous network structure in the polymer matrix. To calculate the overall thermal conductivity of the polymer-based composites, the key factors are dispersity, inherent thermal conductivity, and structure of these nanofillers. The reason is that the thermal energy can be directly transferred through these network structures with minimum phonon scattering [96].

The phonon conduction of thermal energy in materials is affected by scattering effects (interfacial boundary- and defect scattering). Moreover, the damping of the phonons' vibrational amplitude indicates some relevance in the same context. The scattering events probability depends on the defects number and the overall size of the provided interface, defining the efficiency as a thermal conductor. The more the defect density and the larger the provided interface, the lower the efficiency as a thermal conductor. The potential of CNT and other nanofillers can, thus, be assumed as restricted because of the huge surface area, which acts as an interface in polymer nanocomposites [100].

Because of the high disparity of electrical conductivity between the polymer matrix and CNT and Gr, the polymer/CNT or polymer/Gr nanocomposites reveal a percolation-like behavior, and their electrical conductivity enhances abruptly when the CNT and Gr content attains a certain threshold. Two electrical conductivity mechanisms are responsible for this percolation behavior: (1) electron hopping (or quantum tunneling) at the nanoscale and (2) conductive networks at the microscale. Based on the first mechanism, electrons may hop within the tubes or from one CNT to another. This probability is predominantly reliant on the separation distance between the CNT. If the CNT content in the nanocomposite is very low with a high separation distance between the CNT, electron hopping controls the electrical conductivity of the nanocomposite. However, when the separation distance between the CNT diminishes by increasing the CNT content, several adjacent CNT can be electrically connected, leading to the formation of a microscale conductive network. With the CNT content getting larger and larger, the contribution of the conductive network to electrical conductivity outweighs electron hopping [101–103]. Fig. 3 indicates a carrier-transferring model of CNT networks. Two different magnification of conductive paths for doped random CNT can be observed in Fig. 3a and b and for doped 35% stretched CNT in Fig. 3c and d. As can be observed in Fig. 3b and d, among all individual CNT of doped random CNT, the only effective CNT segments that are placed in the conductive path help to provide electrical conductivity. Carriers move via the shortest conductive path with the lowest contact resistance and shortest junction length between the CNT. The adjacent CNTs (dashed circles in Fig. 3b), which are not connected to the conductive path, hardly help the network conductance. Owing to high alignment of individual CNT and dense structures, carriers for the doped 35% stretched CNT move through directly along the CNT with a short junction gap between them. Compared to other samples, the doped 35% stretched CNT exhibits more electrical conductivity, because of the synergistic influences of high alignment and dense structure with shorter junction lengths and high

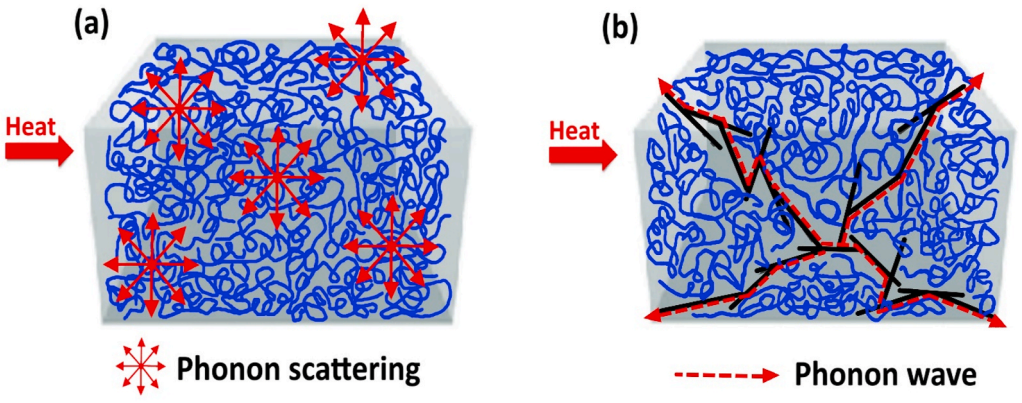

Fig. 2. A polymer filled with a thermally conductive filler.

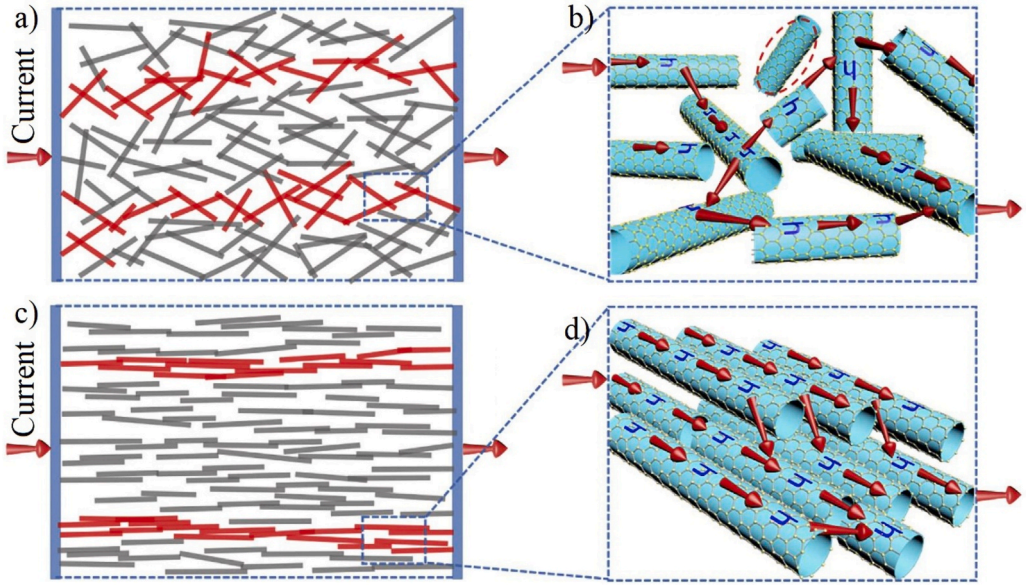

Fig. 3. Schematic of electrical conduction mechanism described by the heterogeneous model. Two examples of electrically conductive pathways for (a) random and (c) 35% stretched CNT are highlighted. (b) and (d) show more details of electrical contribution from CNT alignment, compact structure, and chemical doping for random and 35% stretched ones, respectively [102]. With permission from Elsevier Ltd.

carrier concentration coming from iodine doping.

Both experiments and simulations have indicated that the more the aspect ratio of filler, the less the PT [104,105]. Therefore, Gr is one of the most efficient nanofillers because of its large aspect ratio. Zhang et al. [106] incorporated graphene and graphite into polyethylene terephthalate and determined the PT for each reinforcement. The PT obtained for Gr (~ 0.5 vol%) was remarkably less than that for graphite (~ 4 vol%). These experimental results were in good agreement with simulations carried out by Ambrosetti et al. [104]. The results of simulations revealed that composites containing conductive, plate-like fillers with an aspect ratio of 100 reveal a PT of 26 times lower than composites containing fillers with an aspect ratio of 1. However, the PT enhancement resulted from high aspect ratio fillers can be extremely influenced by the preparation procedure. Du et al. [107] incorporated CNT and GNPs into high-density polyethylene. Their outcomes demonstrated that the nanocomposites containing CNT exhibited a lower PT than those containing GNPs. They stated that this is because the GNPs tend to aggregate and wrinkle, decreasing their effective aspect ratio, and preventing the network formation. Actually, an extremely low PT was obtained for the samples containing CNT (0.0025 vol%) due to preserving their high aspect ratio. On the other hand, Gr has the merit of easier processability than CNT as the viscosity of nanocomposites filled with Gr is remarkably lower than those incorporated with CNT at the same concentrations [108]. This means that a higher content of nanofiller can be added to the matrix, resulting in higher electrical conductivity.

Various theories have been proposed for investigating the influence of the number of interconnections on the electrical conductivity of a random system. However, the variables number influencing the electrical conductivity of these systems makes it challenging. Fournier et al. [109] utilized an analytical model, based on the Fermi–Dirac distribution, for evaluating the transition from insulator to conductor. According to this model, the electrical conductivity can be expressed as follows:

$$\log(\sigma_c) = \log(\sigma_{Gr}) + \frac{\log(\sigma_m) - \log(\sigma_{Gr})}{1 + e^{b(p-p_c)}} \quad (1)$$

where σ_c, σ_{Gr}, and σ_m denote the conductivities of the composite, Gr and matrix respectively, p corresponds to the mass fraction, p_c denotes the PT, and b is an empirical parameter, resulting in the change in electrical conductivity at p_c. The PT can be determined by fitting Equation (1) versus the experimental values.

To model the composites electrical conductivity, most researchers utilize the classical percolation theory as follows:

$$\sigma_c = \sigma_{Gr}(\psi_{Gr} - \psi_c)^t \quad (2)$$

where ψ_{Gr} denotes the volume fraction of Gr, ψ_c corresponds to the percolation volume fraction, and σ_c and σ_{Gr} are the electrical conductivity of the composite and the Gr, respectively. In addition, t denotes the critical power law exponent and relies on the dimensionality of the network, normally taking values of ~ 1.33 for 2D systems (such as when confined in a coating) and ~ 2 for 3D systems [110].

5. Dispersion of nanofillers in polymer matrices

The degree of dispersion of nanofillers in organic polymer matrices is quite poor because of their nano-scale. If homogenous dispersion of nanofillers in polymers is obtained, the final performance of the polymer nanocomposite enhances remarkably because of the improvement in load transfer from the polymer matrix to the nanofillers. Moreover, a higher nanofiller aspect ratio, alignment, and stress transfer improve the polymer performance [111]. To form optimal nanofiller reinforced polymer nanocomposites, surface modification of nanofillers plays a major role in their dispersion in polymer matrices. The organic functionalization of nanofillers makes them more compatible with the organic polymer matrices. Such nanocomposites exhibit enhanced gas barrier, electrical, thermal, and mechanical properties with uniform distribution of nanofillers in the polymer. Also, it is worth mentioning that dispersion of nanofillers relies on factors, such as type of functionalization, presence of polar groups on the surface of nanofillers or polymer, and composite fabrication method [49].

6. Applications of epoxy resins reinforced with carbon nanotubes and graphene-based nanoparticles

CNT and Gr-based nanoparticles have been used in multiple applications due to their outstanding features. A close look at the literature clarifies that adding these nanofillers into EP resins improves their final performance. As EP resins reinforced with CNT or Gr-based nanoparticles exhibit superior properties, they have been extensively used in

aviation, military, aerospace, coating, electrical and electronic devices, etc. In the case of structural components, i.e., fuselage and control surfaces of aircraft, using these nanocomposites has been recommended [112,113]. Because of their excellent mechanical properties, high thermal conductivity, and low electrical conductivity at very low temperatures, EP/GO nanocomposites have been used in encapsulating materials for superconducting magnets [91,114]. On the other hand, EP/Gr nanocomposites have been utilized as conductive materials in a broad spectrum applications, i.e., electromagnetic interference (EMI) shielding [115,116], anticorrosive [117,118], stealth composite coating [119], electrostatic-dissipative [120], and sensors [121,122]. In addition, EP/amino-silane modified GO coatings on mild steel substrate have been found in applications where extraordinary corrosion resistance is needed [123]. For energy storage devices, i.e., conducting electrodes, supercapacitors, and lithium-ion batteries, EP/GNP nanocomposites have been promising candidates [49]. The efficient heat dissipation is a vital factor in high power density communication devices, storage devices, and integrated circuits. Gr has been usually utilized to cope with this issue due to its excellent thermal conductivity [124]. In the case of efficient thermal management, EP/Gr nanocomposites have been applied in thermal interface materials [125]. Also, these types of nanocomposites have been utilized in EMI shielding applications for shielding from the frequency radiations [126]. In addition to the mentioned applications, EP/Gr nanocomposites have been found in other applications, i.e., solar cells, interior and exterior panels, electronic packaging, golf clubs, aircraft flame retardant panels, field emission devices, gas sensors, gas tanks, and tennis and hockey sticks [67,127–129]. Similar applications have been reported for EP/CNT nanocomposites. These nanocomposites have been used in structural applications, coatings, load-bearing applications, electronic packaging applications, erosive wear, thermal interface materials, and so on [130–132].

7. Recent reports on improving the electrical and thermal conductivities of epoxy-based nanocomposites

In this survey, we elaborate on the main results obtained from the last decade's advancements in promoting the electrical and thermal conductivities of EP resins using CNT and Gr-based nanoparticles.

7.1. Carbon nanotube nanofillers

Saw et al. [133] added two types of MWCNT. i.e., long MWCNT (L-MWCNT) and short MWCNT (S-MWCNT) to the EP resin using an ultrasonication technique. They studied the effect of these nanoparticles on the electrical conductivity of EP-based nanocomposites. Their results revealed that except for a large difference in the PT, both types of nanofillers exhibited the same behavior. The PT of the S-MWCNT was 0.1 wt%, much lower than that of L-MWCNT (0.3 wt%). Also, the electrical conductivity of the EP resin was 2.04×10^{-6} S/m, which was increased to 7.74×10^{-3} and 1.11×10^{-2} S/m for EP/L-MWCNT and EP/S-MWCNT nanocomposites containing 0.5 wt% nanofillers, respectively. In another work, Zhang et al. [134] evaluated the electrical conductivity of EP resins containing different amounts of amino-coated MWCNT (NH_2-MWCNT). Their results indicated a positive effect of surface modification on the dispersion of nanofillers in the EP resin. In addition, the PT of the EP/MWCNT nanocomposites was 0.51 wt%, while that of EP/NH_2-MWCNT nanocomposites was 0.13 wt%. Also, the electrical conductivity of the neat EP was approximately 5.2×10^{-15} S/m, which increased to 1.2×10^{-6} S/m for the EP containing 5 wt% MWCNT, and 9.4×10^{-6} S/m for the EP containing 0.5 wt% NH_2-MWCNT. The research team claimed that the lower PT of the EP/NH_2-MWCNT is due to the fact that surface modification of MWCNT using amino groups has a remarkable positive effect on the dispersion of nanofillers, leading nanoparticles to form an electrically conductive network to increase the electrical conductivity.

To break down long CNT (L-CNT) to short CNT (S-CNT), Russ et al. [135] utilized a sonication-induced scission. They incorporated up to 1 wt% of nanofillers in the EP resin and investigated the electrical and thermal conductivities of the samples. Compared to the neat EP, the EP/S-CNT and EP/L-CNT nanocomposites demonstrated eight and 12 orders of magnitude increase in the electrical conductivity, respectively. Both nanofillers had a modest improvement in the thermal conductivity of the nanocomposites. The thermal conductivity of the neat EP was approximately 0.13 W/mK, which was enhanced up to around 0.16 and 0.23 W/mK when 1 wt% S-CNT and L-CNT were added to the EP, respectively. The work of Gradea et al. [136] focused on the influence of pristine, oxidized, and fluorinated CNT (CNT, O-CNT, and F-CNT, respectively) on the electrical and thermal conductivities of the EP-based nanocomposites. Based on their results, the greatest improvements in the electrical conductivity of the EP/CNT and EP/O-CNT nanocomposites were observed at 0.3 wt% nanofiller, which were approximately 10 and 7 orders of magnitude larger than that of the neat EP, respectively. In contrast, incorporation of F-CNT in the EP revealed almost no change in the electrical conductivity. Moreover, slight enhancements of around 5.6, 4.4, and 2.6% in the thermal conductivity were observed for the EP/CNT, EP/O-CNT, and EP/F-CNT nanocomposites, respectively, compared to that of the pure EP.

Vahedi et al. [137] studied the influence of different contents (0.05, 0.1, 0.25, 0.5, 1, and 2 wt%) of MWCNT on the electrical conductivity of an EP resin. They reported that MWCNT amounts of lower than 0.1 wt% had no effect on the electrical conductivity of the EP resin. Moreover, the optimum nanofiller content for obtaining desirable electrical conductivity was approximately 0.5 wt%, which was the PT for the nanocomposites. Beyond this weight percentage, electrical conductivity exhibited only a slight change. The electrical conductivity was observed to increase from 5×10^{-13} S/cm for the neat EP to 1.2×10^{-5} S/cm for the EP containing 0.5 wt% MWCNT and to 7.9×10^{-5} S/cm for the EP containing 2 wt% MWCNT. A low magnetic field (MF) of 0.4 T was applied by Ma et al. [30] to align the CNT in the EP resin. They measured the MF alignment both along and transverse to the MF direction. Their research demonstrated that the degree of CNT alignment was decreased when a high content of CNT was incorporated in the resin. By increasing the CNT amount, the electrical conductivity of samples showed improvement, indicating an insulating-to-conducting transition with a remarkable electrical conductivity enhancement of 6-8 orders of magnitude. At and beyond 3 wt% CNT content, the plateau electrical conductivity of all samples, regardless of how they were oriented, was the same (10^{-5} S/cm). The authors claimed that because of inadequate electrical conductivity data in the range of 1–3 wt% CNT, the PT could not be achieved. However, they argued that it should be less than 3 wt%. In another research work, Schilde et al. [138] introduced a novel production technique, a modification of a generally utilized solvent-based method, for highly loaded EP-based nanocomposites. In this technique, which is presented schematically in Fig. 4, a high CNT content between 10 and 80 wt% was realized. The electrical conductivity of the corresponding nanocomposite showed remarkable improvement, attaining a maximum of 838 S/m when 60 wt% CNT was incorporated.

Fogel et al. [46] fabricated EP/MWCNT nanocomposites by a three-roll mill process and investigated their electrical properties. An improvement in electrical conductivity of EP by 11 orders of magnitude was observed in the DC mode. Moreover, the PT was determined to be 0.25 wt% CNT and the maximum electrical conductivity was 0.006 S/m for EP containing 0.75 wt% CNT. Pereira et al. [139] carried out non-covalent functionalization of MWCNT using an imidazolium-based ionic liquid (IL) to achieve better dispersion of MWCNT in an EP resin and achieve higher electrical and thermal properties. They used 1 phr functionalized MWCNT with different MWCNT/IL ratios (1:0, 1:5, 1:10, and 1:15). Their results indicated that incorporating functionalized MWCNT with a mass ratio of 1:5 increased the electrical conductivity of the EP by approximately three orders of magnitude. In addition, the

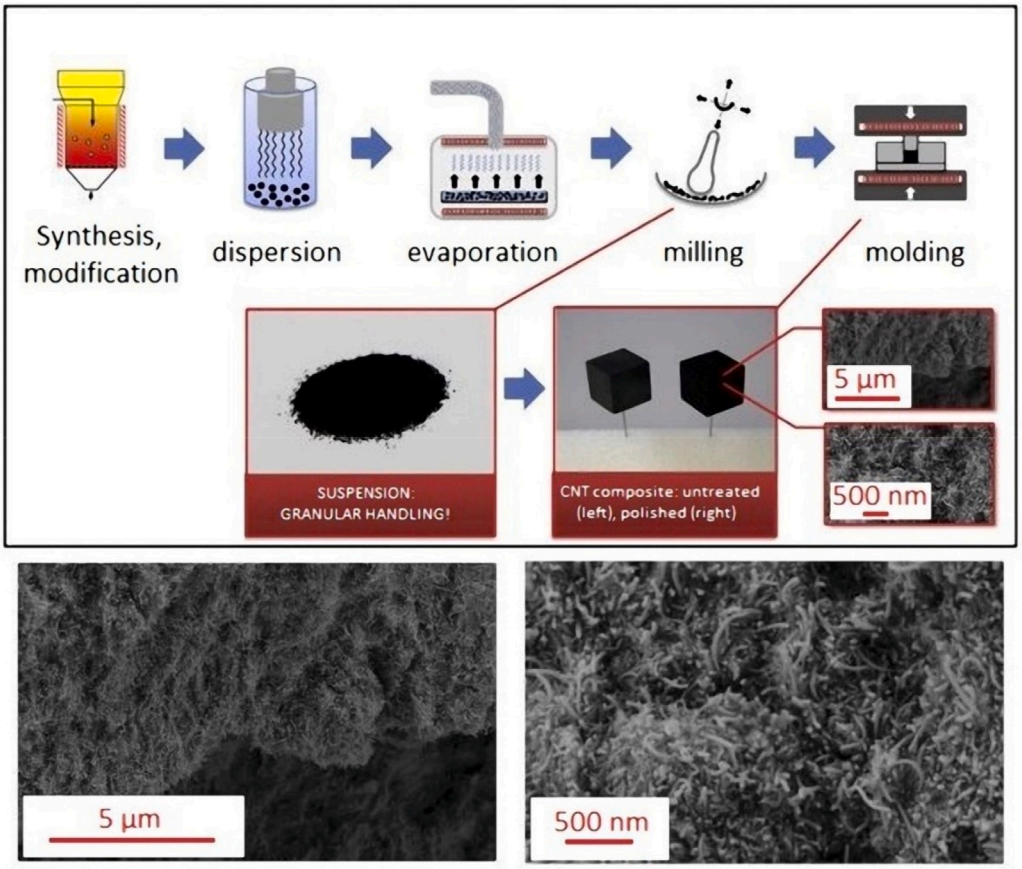

Fig. 4. The production technique of highly loaded EP-based nanocomposites (top). Milled CNT-EP resin suspension, prepared nanocomposite cubes, and high-resolution SEM micrographs of the nanocomposite fragment with 50 wt% CNT (below) [138]. With permission from Elsevier Ltd.

greatest improvement in thermal conductivity (98%) was observed when functionalized MWCNT with a mass ratio of 1:15 were incorporated in the EP. Li et al. [140] incorporated functionalized MWCNT (f-MWCNT) in an EP resin in the presence of alpha-zirconium phosphate (ZrP) to assist with the nanofiller dispersion. Using this procedure, they prepared EP/f-MWCNT nanocomposite foams through a supercritical CO_2 foaming technique. The solid EP/f-MWCNT and foamed EP/f-MWCNT nanocomposites exhibited an enhancement of around 8–9 and 12 orders of magnitude, respectively, compared to the electrical conductivity of the neat EP resins.

Chen and Yan [141] investigated the effect of MWCNT aspect ratio on the electrical and thermal conductivities of an EP resin. For this purpose, they used L-MWCNT with an aspect ratio of 800 and S-MWCNT with an aspect ratio of 80. The outcome of their research revealed that PT for both types of MWCNT in the resin is lower than 0.1 wt%. In addition, the electrical conductivity of the EP/L-MWCNT and EP/S-MWCNT nanocomposites were increased by 8 and 5 orders of magnitude, respectively, compared to that of neat EP. Also, the thermal conductivity of EP exhibited approximately 120 and 26% enhancements when 1 wt% L-MWCNT and S-MWCNT were incorporated in the EP resin, respectively. Tanabi and Edral [142] evaluated the influence of the shear mixing parameters and CNT amounts on the electrical conductivity of EP/CNT nanocomposites. For this purpose, they applied mixing speeds of 500 and 2,000 rpm, mixing times of 10 and 60 min, and CNT contents of 0.2 and 0.5 wt%. Their results indicated that when samples were mixed at 2,000 rpm for 60 min, the electrical conductivity was approximately six times higher than those of the other samples with the same CNT content. In addition, subjecting an EP/CNT suspension to a magnetic field (MF) of 0.2 T during the curing cycle resulted in a two-fold enhancement of the electrical conductivity.

The effect of CNT purification on the thermal conductivity and EC of an EP resin was studied by Chen et al. [41]. The thermal conductivity of the pure EP was found to be around 0.22 W/mK and an enhancement of up to four times was observed for the EP/purified CNT nanocomposite, signifying the positive effect of CNT purification. Also, the PT of the EP containing purified CNT was much higher than that of the EP containing pristine CNT. Additionally, the electrical conductivity of the EP resin reinforced with pristine CNT was almost three orders of magnitude higher than that of the EP/purified CNT nanocomposite. In Fig. 5, the electrical and thermal conductivities of the EP resins containing different amounts of pristine CNT and purified CNT are illustrated.

7.2. Graphene-based nanofillers

Kim et al. [143] incorporated aluminum hydroxide-functionalized GO (AH-GO) in an EP resin and studied the electrical and thermal properties of the final nanocomposites. The thermal conductivity of EP/GO nanocomposite was observed to increase by approximately 91% compared to that of the neat EP. In contrast, incorporating AH-GO in the EP led to a lower enhancement (35%). Finally, the electrical conductivity of EP/GO and EP/AH-GO was improved by 113 and 77%, respectively, compared to that of the pure EP. Tang et al. [144] studied the impact of the dispersion state of thermally reduced GO (RGO) on the electrical conductivity of an EP. As their results suggested, EP incorporated with highly dispersed RGO exhibited an electrical conductivity of one or two orders of magnitude greater than that incorporated with poorly dispersed RGO. Monti et al. [145] introduced two solvent-assisted methods to enhance the electrical conductivity of the EP/graphene nanoplatlets (GNP) nanocomposites containing 0.5–3 wt% nanofiller. In the first method, they used chloroform and EP monomer,

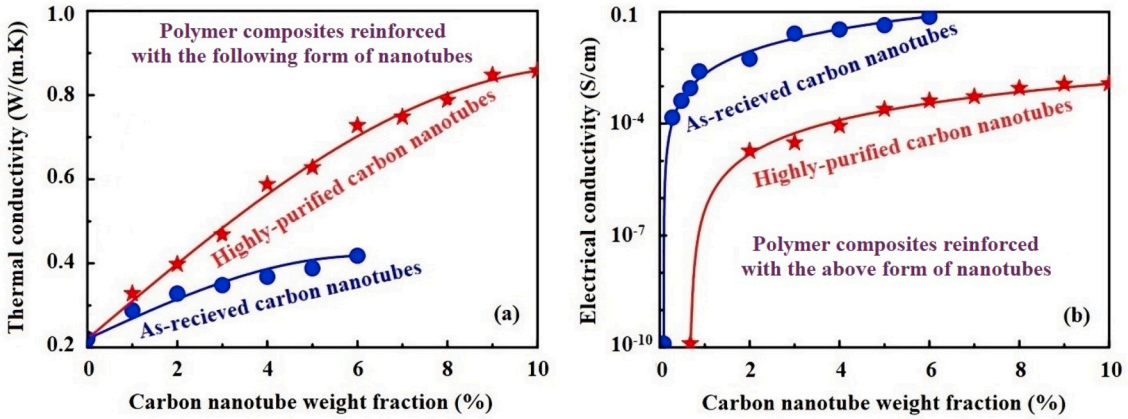

Fig. 5. (a) Thermal and (b) electrical conductivities of the EP resin containing different amounts of as-received CNT and highly-purified CNT [41]. With permission from Elsevier Ltd.

while in the second method, they utilized tetrahydrofuran and an aromatic hardener. Although both methods indicated good dispersion of GNP in the EP resin, the first method led to a better GNP exfoliation. Incorporating 3 wt% GNP in the EP resin resulted in an increase of almost 10 orders of magnitude in the DC electrical conductivity of nanocomposites prepared by both methods. Additionally, the first method did not lead to a remarkable change in the AC electrical conductivity of the EP resins containing GNP. In contrast, the second method resulted in an enhancement of approximately five orders of magnitude in the AC electrical conductivity of the EP containing 3 wt% GNP in the frequency of 10 Hz and around one order of magnitude in the frequency of 10^6 Hz. Wajid et al. [146] introduced two different procedures, freeze-drying/mixing and solution processing, for the addition of Gr to an EP resin to produce multifunctional nanocomposites. Specifically, for better dispersion of the nanofiller in the EP matrix, they used an efficient method of dispersing Gr in an extensive range of solvents, utilizing a stabilizing polymer, i.e., polyvinylpyrrolidone (PVP). They found that the freeze-drying/mixing procedure was more promising and versatile. The electrical conductivity of the EP/PVP/Gr containing 0.5 wt% nanofiller prepared by this method was approximately one order of magnitude higher than the other method. Song et al. [147] improved the thermal conductivity of an EP resin using non-covalently functionalized Gr flakes (f-GFs) prepared by a novel method. According to this technique, f-GFs possessing remarkably low oxygen content (2.9%) were fabricated using a ternary eutectic system of alkali salts. Their results revealed that the thermal conductivity of the neat EP was 0.18 W/mK and increased up to 1.53 W/mK when 10 wt% f-GFs was added to the EP. Guo et al. [148] fabricated nanocomposites of thermally conductive EP resin, containing 5–25 wt% GNP, using ball milling. Their results showed that higher thermal conductivity could be obtained at higher GNP contents. Compared to the neat EP, the thermal conductivity of the sample containing 25 wt% GNP was enhanced by approximately 1,364%.

Li et al. [149] report a simple and scalable technique for the preparation of EP-based nanocomposites, utilizing aligned and interconnected multilayer Gr (MLG). They incorporated 11.8 wt% MLG in the EP and found that thermal conductivity reached 33.54 W/mK, a dramatic enhancement of 16,670% compared to that of the neat EP. Different contents (7, 14, and 35 wt%) of a combination of Gr and graphite were used by Mahanta et al. [150] to improve the thermal conductivity of an EP resin. With the addition of 35 wt% mixed fillers (a 6:1 ratio of Gr: graphite), the thermal conductivity of 42.4 W/mK was obtained, which was approximately 250 times higher than the thermal conductivity of the neat EP. In another work, Wang et al. [151] studied the effect of GNP size on the thermal properties of EP-based nanocomposites. Two types of GNP, i.e., small GNP (S-GNP with a diameter of less than 1 μm) and large GNP (L-GNP with a diameter of 5 μm), were used. Their results demonstrated that the highest improvement (115%) in thermal conductivity was obtained when 5 wt% L-GNP were added to the EP.

Ribeiro et al. [152] modified a few-layer GO with tetraethylenepentamine (TEPA) using a reaction assisted by microwave radiation. They further fabricated EP/TEPA-GO nanocomposites. The thermal conductivity of the neat EP was 0.35 W/mK and increased up to 70 and 103% for the EP/GO and EP/TEPA-GO nanocomposites, respectively, when 0.5 wt% nanofillers were added to the resin. They claimed that such an enhancement in the thermal conductivity of the EP containing modified GO indicates the important role of surface modification in inhibiting nanofiller aggregation. In the work of Lian et al. [153], novel vertically aligned and interconnected Gr networks (VAIGNs) were incorporated in an EP resin, and the thermal conductivity of the nanocomposites was investigated. Top-view and side-view images of VAIGNs is presented in Fig. 6. As this team highlighted, incorporating an ultra-low Gr content (0.92 vol%) in the EP resin resulted in a high thermal conductivity of 2.13 W/mK, equivalent to a significant improvement of approximately 1,230% compared to the neat EP.

Olowojoba et al. [154] investigated the effect of dispersing freeze-dried RGO in an EP resin using shear mixing on the thermal conductivity of EP-based nanocomposites. They incorporated different contents of RGO in the EP resin and recorded the thermal conductivity of nanocomposites in a temperature range of 30–60 °C. Their results demonstrated that the maximum thermal conductivity was 0.264 W/mK for the EP resin containing 2 wt% RGO at 60 °C, an enhancement of about 43% compared to the neat EP at the same temperature. Yao et al. [155] introduced the chemical functionalization of Gr using 4-nitrobenzenediazonium salt as a new method to homogeneously disperse Gr nanosheets in the EP resin. The outcome of their research revealed that the thermal conductivity of the EP-based nanocomposites with 5 wt% modified Gr was increased up to 0.56 W/mK, almost 2.5 times over that of the pure EP. In another study conducted by Imran et al. [47], the electrical conductivity of an EP resin (EPON 828) was enhanced by adding Gr. They fabricated EP/Gr nanocomposites by three-roll milling as the nanofiller dispersion method and found that the PT of the Gr was 1 wt%. The electrical conductivity of the EP was observed to increase from 4.3×10^{-15} for the neat EP to 2.6×10^{-6} S/m for the nanocomposite. In addition, incorporating a higher Gr content up to 2 wt% led to a further increase in the electrical conductivity to of 3.1×10^{-4} S/m. Also, the thermal conductivity was enhanced from 0.14 W/mK for the neat EP to 0.27 W/mK for the EP containing 1 wt% Gr. In another work, Bustero et al. [156] introduced a novel technique for the addition of Gr in an EP, through the incorporation of Gr as a free-standing film in EP. Their outcomes showed that the thermal conductivity of EP

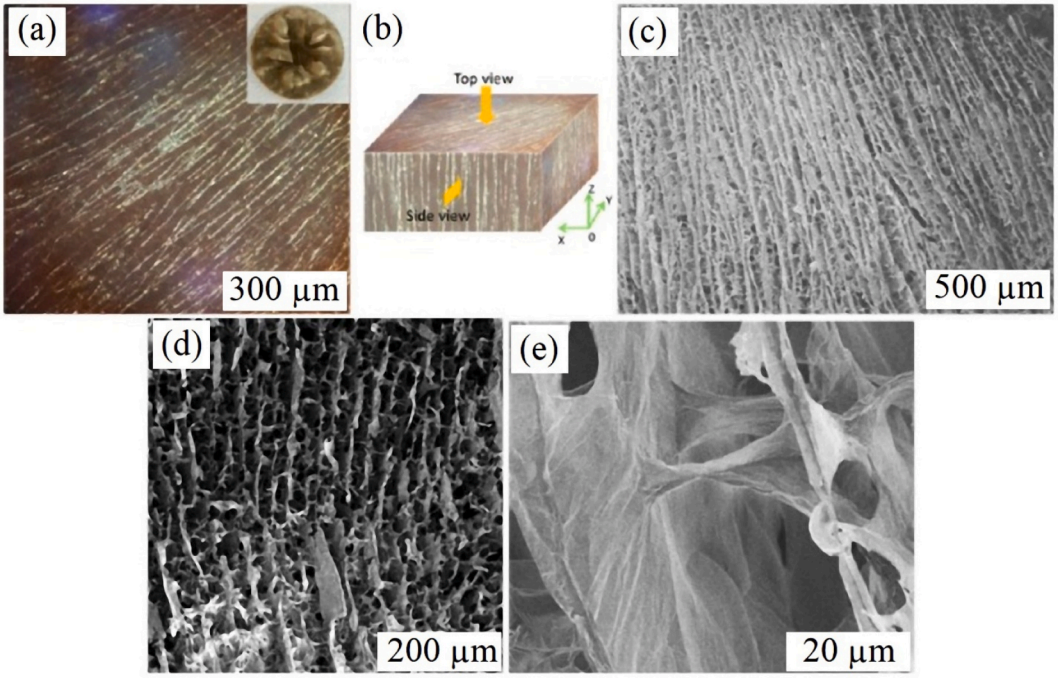

Fig. 6. (a) Optical top-view image and (b) Schematic illustration of vertically aligned GO network. Inset in (a) is the corresponding GO foam. (c) SEM image of GO network (top view). (d) SEM image of GO network (side view). (e) SEM image of the nanojunctions between the GO walls [153]. With permission from ACS publications.

enhanced from 0.2 to 20 W/mK, when 30 wt% Gr film is added.

7.3. Hybrid nanofillers

These days, attention has shifted more to the simultaneous utilization of various nanofillers in polymer matrices, especially EP, and their combined effects on the thermal and electrical conductivities of polymer nanocomposites. The strategy of using hybrid nanofillers with various dimensions and shapes has been indicated to result in improved thermal and electrical conductivities of the final nanocomposite [157,158].

7.3.1. Hybrid carbon nanotubes and graphene-based nanofillers

Yue et al. [159] studied the influence of different CNT:GNP ratios on the electrical conductivity of the EP-based nanocomposites. At a content of 4 wt% CNT, the EP/CNT nanocomposites indicated an electrical conductivity of 3.9×10^{-3} S/m, around nine orders of magnitude higher than that of the neat EP. Approximately the same improvement was observed when 4 wt% hybrid nanofillers with a CNT:GNP ratio of 8:2 was incorporated in the EP. Moreover, with the addition of 4 wt% GNP to the EP, the best performance, i.e., an electrical conductivity of 1.68×10^{-5} S/m (seven orders of magnitude increase over the neat EP), was obtained for the EP/GNP nanocomposite. In a similar work, Moosa et al. [160] prepared EP/CNT/GNP hybrid nanocomposites using shear mixing followed by sonication. Their results showed that the EP containing 0.4 wt% hybrid nanofillers with a CNT:GNP ratio of 1:1 improved the electrical conductivity of the EP by almost eight orders of magnitude. Wang et al. [48] fabricated EP/CNT/GO nanocomposites, in which GO behaved as a non-covalent dispersant for CNT in the EP resin and as a reactive secondary reinforcing filler to enhance the thermal conductivity of EP resin. The thermal conductivity of neat EP was around 0.06 W/mK and increased up to approximately 0.19 W/mK when 4 wt% hybrid nanofillers were added to the EP. In another attempt to increase the thermal conductivity of EP resins, Liang et al. [161] applied three different methods, i.e., combined hydrothermal reduction, vacuum-assisted infiltration, and ice-templated assembly, to fabricate well-aligned 3D networks of RGO walls bridged by f-SWCNT in the EP resin. Their results revealed that the 3D RGO/f-SWCNT aerogel significantly increased the thermal conductivity without sacrificing the electrical insulating behavior of the aerogel. When 3.65 vol% mixed RGO/f-SWCNT (1:2.5) was added to the EP resin, the thermal conductivity of the nanocomposite reached 0.63 and 0.69 W/mK at 300 and 390K, respectively. This is four times higher than the thermal conductivity of the EP.

7.3.2. Hybrid carbon nanotubes or graphene-based nanofillers with inorganic nanoreinforcements

Teng et al. [162] studied the effect of simultaneous use of MWCNT and micro-sized aluminum nitride (AlN) on the thermal conductivity of an EP resin. They functionalized MWCNT using poly(glycidyl methacrylate) (PGMA) by free radical polymerization and modified AlN using a zirconate coupling agent to form covalent bonds between the EP and the reinforcements. The SEM micrographs of the resulting nanocomposites confirmed that the functionalized fillers were well dispersed in the EP. Also, the thermal conductivity of the EP resin containing 25 vol% modified AlN and 1 vol% functionalized MWCNT was 1.21 W/mK, approximately equal to that of the EP resin containing 50 vol% unmodified AlN (1.25 W/mK). Hsiao et al. [163] prepared hybrid nanosheets possessing sandwich structures consisting of thermally reduced GO (TRGO) and silica. The thermal conductivity of the EP was found to increase by around 19, 27, 37, and 61% by incorporating 1 wt% silica nanoparticles, silica nanosheets, TRGO, and TRGO/silica nanosheets in the EP resin. Also, the electrical resistivity exhibited almost no change when 1 wt% silica nanoparticles and silica nanosheets were added to the resin, while it decreased by approximately seven and four orders of magnitude for the EP containing the same amounts of TRGO and TRGO/silica nanosheets. The reason behind the observed thermal and electrical properties of the TRGO/silica nanosheets was claimed to be the coverage of the TRGO surface by the silica layer, which prevents electrical conduction and, therefore, efficiently forms a 3D phonon transport channel. Consequently, the electrical and thermal properties of the EP resin is dramatically influenced.

Zakaria et al. [164] used different contents of hybrid alumina/CNT

fillers (1–5 wt%) in an EP resin to investigate the thermal conductivity of the resulting nanocomposites. Their materials consisted of a physically mixed alumina/CNT filler (MIX) and a synthesized hybrid alumina/CNT filler (HYB). The HYB was prepared by chemical vapor decomposition. Fig. 7 reveals the SEM images of MIX and HYB. The HYB is formed by the attachment of CNT to the alumina particles and, hence, the CNT dispersion is dependent on alumina dispersion. On the other hand, the CNT and alumina particles in MIX are not physically attached to each other. Therefore, the CNT are not uniformly distributed on the surface of alumina particles and their dispersion is independent of the dispersion of the alumina particles. The thermal test indicated that the EP/HYB nanocomposites had greater thermal conductivity than EP/MIX nanocomposites. The thermal conductivity of EP resin was enhanced by approximately 13 and 16% by incorporating 5 wt% MIX and HYB in the resin, respectively.

Wang et al. [165] prepared a hybrid nanofiller, consisting of GO and nanosilica (NS) using sol-gel and surface treatment processes. They added the hybrid nanofiller to the EP resin and evaluated the nanofiller effect on the thermal conductivity of the EP resin. Their results demonstrated that the thermal conductivity of the EP containing 1.5 wt % hybrid filler was 1.38 times that of the neat EP resin. Gao et al. [166] synthesized aluminum nitride (AlN), AlN/Gr, and AlN/CNT hybrid nanostructures. They studied the influence of these synthesized hybrid nanofillers on the thermal conductivity of the EP-based nanocomposites containing alumina microparticles. Based on their results, the largest improvement in the thermal conductivity was achieved when 5 wt% AlN/Gr and 45 wt% alumina were incorporated in the resin, i.e., an increase of 185% compared to the neat EP. Kandare et al. [167] added GNP, silver nanoparticles (SNPs), and silver nanowires (SNWs) to carbon fiber (CF, 45 vol%)-reinforced EP to achieve enhanced through-thickness thermal and electrical conductivities. The thermal conductivity of EP/CF/GNP nanocomposite increased up to 9% with the addition of 1 vol% GNP to the EP resin. Also, incorporating 0.05 vol% SNPs and SNWs in EP/CF led to thermal conductivity enhancements of 18 and 8%, respectively. In addition, the largest enhancement in electrical conductivity (around 70%) was observed when 0.05 wt% SNWs was added to the EP/CF nanocomposite.

It is accepted that although Gr can significantly increase the thermal conductivity of EP resins because of its high aspect ratio and excellent inherent thermal conductivity, it leads to a serious decrease in electrical insulation, thereby limiting the widespread application of EP/Gr nanocomposites in thermal management systems. To overcome this drawback, Sun et al. [168] decorated GNP with electrically insulating alumina using a supercritical carbon dioxide ($scCO_2$)-assisted technique. Their results revealed that the EP containing 12 wt% GNP/alumina hybrid nanofiller indicated a high thermal conductivity of 1.49 W/mK, which was approximately 677% higher than that of the neat EP. Rybak et al. [169] added a novel class of hybrid nanofiller, i.e., a masterbatch of GNP and NS, in an EP resin. They used two methods, a standard (ST) method and a modified ultrasonic (US) method, for preparing the samples, as shown in Fig. 8. Their results demonstrated the thermal conductivity of the nanocomposite containing 2 wt% GNP and 63 wt% silica prepared by the ST method was improved by 56% compared to the sample containing 65 wt% silica fabricated by the ST method. Similarly, the thermal conductivity of the sample prepared by the US method was increased by 58%. These improvements were achieved because of the bridges formed by the GNP between the silica grains, as shown in Fig. 9. It was claimed that such a structure would lead to an enhanced thermal conductivity without sacrificing electrical insulation.

Akhtar et al. [170] reported the influence of surface modification of alumina/Gr hybrid reinforcement by silane moieties on the thermal conductivity of an EP resin. Based on their results, surface modification of the hybrid filler resulted in good bonding between the EP and the hybrid filler, causing an excellent dispersion of the hybrid filler in EP resin. The thermal conductivity for the EP containing 50 vol% alumina and 1 wt% Gr was approximately 10 times greater than that of the neat EP. In another study, a novel strategy was applied by Ren et al. [171] to increase the thermal conductivity of an EP resin by utilizing a hybrid filler of Gr and boron nitride (BN) with different ratios (1:1, 1:10, and 1:50). The BN nanosheets in the hybrid filler were claimed to tightly adsorb onto the Gr surface through π-π interactions and act as bridges to help convey photons. Moreover, they would act as obstacles to electron transport between the adjoining Gr nanoparticles. The thermal conductivity of the EP containing 5 wt% Gr/BN (1:1) hybrid filler was increased by around 140%, compared to that of the neat EP. Feng et al. [172] used a two-step hydrothermal technique to synthesize a

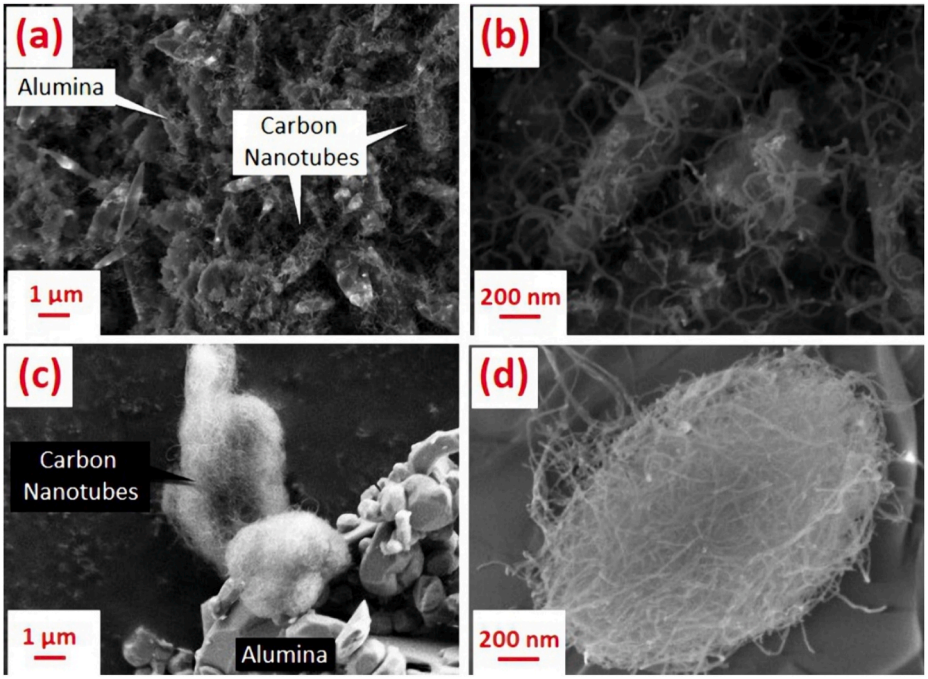

Fig. 7. SEM images of HYB (a, b) and MIX (c, d) [164]. With permission from Elsevier Ltd.

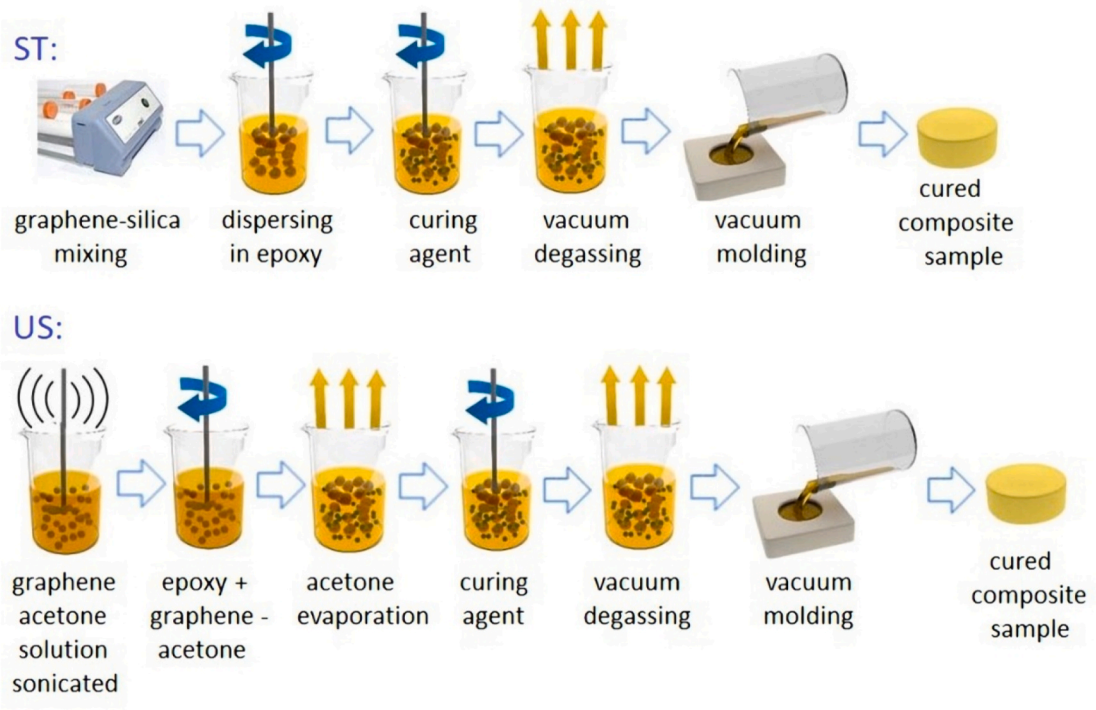

Fig. 8. Schemes of standard and modified ultrasonic techniques of GNP/silica hybrid composite preparation [169]. With permission from John Wiley & Sons, Inc.

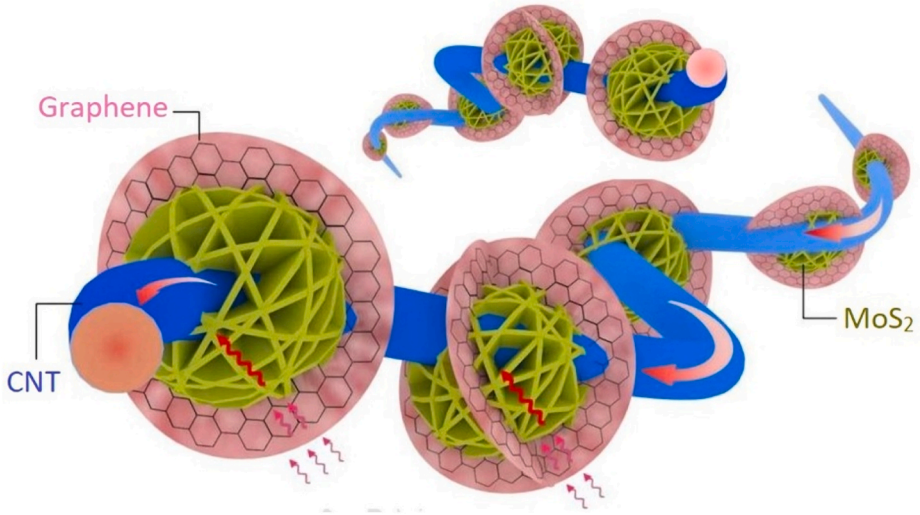

Fig. 9. Structural diagram of CNT/Gr/MoS_2 nanostructure and heat transfer process in the EP matrix [175]. With permission from Elsevier Ltd.

multifunctional hydrophilic Gr-based hybrid containing $Ni(OH_2)$ nanoribbons and RGO. They added hexagonal BN (hBN) and RGO@$Ni(OH)_2$ hybrid filler into the EP resin. Their results revealed that, with the incorporation of 20 wt% hBN and 2 wt% RGO@$Ni(OH)_2$ in the EP resin, an enhancement of thermal conductivity up to 851% over that of the neat EP was achieved. Various ratios of RGO/alumina hybrid filler with a fixed content of 1 wt% were incorporated in an EP resin by Osman et al. [173]. The greatest thermal conductivity was obtained for the nanocomposite containing only RGO (10:0). In contrast, the insulation properties of the EP-based nanocomposites were retained compared to the EP/RGO nanocomposites.

7.3.3. Ternary hybrid nanofillers

Yao et al. [174] reported a simple strategy to enhance the thermal conductivity of an EP resin using a magnetic composite nanofluid as reinforcement. The magnetic nanofluid consisted of a multicomponent core of GO/MWCNT/Fe_3O_4 and a shell of polyether and was synthesized by a combination of ultrasonic-assisted chemical coprecipitation technique and post-modification. Finally, the EP/GO/MWCNT/Fe_3O_4 nanocomposites were fabricated under a magnetic field and the thermal conductivity was measured in the oriented and vertical directions. The thermal conductivity of the neat EP was measured at 0.2345 W/mK. In the oriented direction, the thermal conductivity was slightly enhanced up to 0.2458 W/mK for the EP containing 2 wt% ternary hybrid filler. On the other hand, in the vertical direction, incorporating 0.5 wt% filler decreased the thermal conductivity to 0.1139 W/mK. They concluded that the EP-based nanocomposites exhibited a clear anisotropic thermal conductivity. A low interfacial thermal resistance CNT/Gr/molybdenum disulfide (MoS_2) heterostructure was designed and synthesized by Ji et al. [175]. In the synthesized nanostructure, the heat was collected

from the EP by the Gr and MoS_2 nanosheets and transferred to the thermally conductive CNT channel, which is interconnected in three dimensions in the EP (Fig. 9). These researchers incorporated the ternary hybrid filler in the EP resin and found that 2,300 and 247% enhancements were obtained in the in-plane and through-plane thermal conductivities, respectively, compared to those of the neat EP.

Owais et al. [176] combined the chemical exfoliation and mechanical mixing to disperse the hybrid nanofillers of GNP and BN inside EP/short CF. Compared to the pure EP, incorporating 5 wt% GNP/BN (1:1) gave an increase of approximately 211% in the thermal conductivity. The BN nanoplatelets with smaller sizes than those of the GNP were found to adsorb on the GNP surfaces via π-π stacking, resulting in a good dispersion of GNP in the EP resin and, hence, enhanced thermal conductivity.

8. Similarities and differences in improving the electrical and thermal conductivities of epoxy resins using carbon nanotubes and graphene-based nanoparticles

In this section, we explain the similarities and differences between electrical and thermal conductivities of EP resins reinforced with CNT and Gr-based nanoparticles.

To better understand the influence of CNT, Gr, and their hybrid on EPs' thermal and electrical conductivities, Table S1 provides a summary of previous research studies on EPs' thermal and electrical conductivities reinforced with CNT, Gr, and their hybrid. As expected, CNT, Gr, and their hybrid improved the electrical and thermal conductivities of EP resins in all research studies conducted in the last decade.

A close look at Table S1 reveals that the most improvement in electrical conductivity of EP resins has been achieved by incorporating 3 wt% Gr in EP (EPON 828) [47]. A three-roll mill was used to mix EP and Gr in this study. The significant enhancement in the electrical conductivity of EP in this work indicates that three-roll milling can be an efficient dispersion technique for mixing Gr into EP resin. In addition, the most improvement in electrical conductivity of EP/CNT nanocomposites has been more than that of EP reinforced with hybrid reinforcements. The highest increment in electrical conductivity of EP resins has been obtained by adding 0.92 vol% f-MWCNT in EP [140]. The incorporation of f-MWCNT in EP resin was performed in the presence of alpha-zirconium phosphate (ZrP) to assist with the nanofiller dispersion. The improvement in the electrical conductivity of EP in this study, obtained by MWCNT functionalization and the way to disperse nanoparticles into EP, demonstrate again the importance of state of dispersion.

The highest enhancement (1,364%) in thermal conductivity of EP resins was obtained by adding 25 wt% GNP in EP [148]. This study utilized a ball mill to disperse GNP into EP resin. Moreover, a comparison between the thermal conductivities of EP reinforced with CNT and hybrid nanofillers reveals that EP resins reinforced with hybrid reinforcements had higher thermal conductivity than that of EP/CNT nanocomposites. The maximum improvement in thermal conductivity of EP resins (851%) was achieved when 20 wt% hBN and 2 wt% RGO@Ni $(OH)_2$ were added into EP resin [172].

As it is clear, an investigation of the last decade's studies demonstrates the importance of the state of dispersion in improving the electrical and thermal conductivities, as it can be obtained using efficient dispersion techniques and also nanoparticles functionalization. Therefore, choosing an efficient dispersion method and the best procedure to functionalize CNT and Gr-based nanoparticles are the most important scientific issues in this field to be solved.

9. Conclusion and future challenges

Outstanding properties offered by epoxy (EP) resins have made them one of the most widely utilized matrices in polymer-based nanocomposites. With the addition of high-performance carbon nanotubes (CNT) and graphene (Gr), into EP resins, high-performance nanocomposites can be developed for different applications. Constructing a continuous three-dimensional network of these nanofillers is a promising method for preparing thermally and electrically conductive EP-based nanocomposites. Several scientists have demonstrated that nanofiller functionalization is the foremost factor in achieving the desired thermal and electrical conductivities in the nanocomposites. This is due to the fact that covalent and non-covalent functionalization of CNT and Gr have dramatic effects on the nanofiller dispersion in EP resins. However, two potential disadvantages have been mentioned for the covalent functionalization of CNT and Gr nanofillers: (1) CNT rupture, which leads to a diminishing in the nanofiller aspect ratio, and (2) disturbance of the graphitic structure of Gr. In addition, non-covalent functionalization generally results in poorly bonded functional groups. The electrical and thermal conductivities of CNT and Gr are mainly affected by these features because of the changes in their intrinsic electrical and thermal conductivities. These changes, along with different states of dispersion caused by different functionalization levels, lead to complicated effects on the electrical and thermal conductivities of the EP resins filled with CNT and Gr-based nanoparticles. Therefore, the reported electrical and thermal conductivities of the EP resins filled with CNT and Gr-based nanoparticles are very scattered.

Although the CNT and Gr-based nanoparticles possess ultrahigh thermal conductivity, their utilization in different electronic parts has been restricted due to their high electrical conductivity. Hence, some recently published research papers have developed thermally conductive, yet electrically insulating, nanofillers. Coating and hybridizing the CNT and Gr-based nanoparticles with other additives and morphological control of these nanofillers in polymer-based nanocomposites have been performed to achieve this purpose. Generally, the hybridization of CNT and Gr-based nanoparticles has been carried out using inorganic materials such as silica, alumina, boron nitride, etc., to hinder the formation of electrically conductive networks. Although these works have been carried out in recent years, EP-based nanocomposite materials possessing significant electrical and thermal conductivities have been developed using different processing methods, such as ultrasonication, supercritical carbon dioxide (scCO2)-assisted modification, sol-gel modification, surface treatment, and directional freeze-drying. Some electronic devices need heat dissipation through the thickness rather than in the surface direction. However, the thermally conductive polymer-based nanocomposites generally possess much lower through-plane thermal conductivity than in-plane. Thus, researchers have recently tried to control the oriented heat conduction and vertical alignment of CNT and Gr-based nanoparticles by incorporating ternary hybrid fillers such as graphene oxide/multi-walled CNT/Fe_3O_4 and CNT/Gr/Molybdenum disulfide in an EP resin. Despite the great promise of CNT and Gr-based nanoparticles-reinforced EP nanocomposites as thermally and/or electrically conductive materials, more research is needed to address some pending problems, such as simpler and more cost-effective production of these nanocomposites, before they are fully commercialized. Solutions are further needed to lessen the thermal and electrical interface resistance between the CNT and Gr-based nanoparticles and EP resin, prepare thermally conductive yet electrically insulating fillers, improve the weak interfacial adhesion between the EP and nanofillers, control Gr stacking, prevent CNT bundling, etc. Research must be undertaken to provide a full understanding of the properties of CNT and Gr-based nanoparticles. Additionally, studies on the structural control and alignment of the electrically insulating CNT and Gr-based nanoparticles should be continued so that better thermal management systems can be designed for electronic devices in the future.

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Hossein Ali Khonakdar: investigation, resources, writing—reviewing, Supervision.

All authors read and approved the final manuscript.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.polymertesting.2022.107645.

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Abbreviations

Abbreviation	Explanation
3D:	Three-dimensional
AH-GO:	Aluminum hydroxide-functionalized graphene oxide
AlN:	Aluminum nitride
AN:	Aluminum nitride
BN:	Boron nitride
CF:	Carbon fiber
CNF:	Carbon nanofiber
CNT:	Carbon nanotubes
EP:	Epoxy
F-CNT:	Fluorinated carbon nanotubes
f-GF:	Non-covalently functionalized Gr flakes
f-SWCNT:	Functionalized single-walled carbon nanotubes
GNP:	Graphene nanoplatelets
GO:	Graphene oxide
Gr:	Graphene
hBN:	Hexagonal boron nitride
IL:	Ionic liquid
L-CNT:	Long carbon nanotubes
L-MWCNT:	Long multi-walled carbon nanotubes
MF:	Magnetic field
MLG:	Multilayer graphene
MO:	Metal oxide
MoS_2:	Molybdenum disulfide
MWCNT:	Multi-walled carbon nanotubes
NH_2-MWCNT:	Amino-coated multi-walled carbon nanotubes
NP:	Nanoparticle
NS:	Nanosilica
O-CNT:	Oxidized carbon nanotubes
PGMA:	Poly(glycidyl methacrylate)
PT:	Percolation threshold
PVP:	Polyvinylpyrrolidone
RGO:	Reduced graphene oxide
SCF:	Short carbon fiber
S-CNT:	Short carbon nanotubes
SNP:	Silver nanoparticle
SNW:	Silver nanowire
SWCNT:	Single-walled carbon nanotubes
S-SWCNT:	Short single-walled carbon nanotubes
TEPA:	Tetraethylenepentamine
TRGO:	Thermally reduced graphene oxide