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Article

Epoxy nanocomposites with three-dimensional graphene reinforcements: Specifics and technical forge

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Abstract: Epoxy resins or polyepoxides are basically reactive prepolymers having epoxide functionalities and can be cured using hardening agents to form high-performance materials. Graphene is the most widely adopted nanofiller for thermosets, and similarly modified graphene forms, like three-dimensional graphene, were investigated as a valuable reinforcing agent. Looking at the significance of three-dimensional graphene for epoxy nanomaterials, this innovative review highlights all essential aspects of these materials from basics/synthesis to properties/applications. Consequently, literature reports were observed for facile designs and processing of epoxy/three-dimensional graphene nanocomposites via solution casting, infiltration, or melt blending methods. These nanomaterials have been investigated for microstructures, thermal/mechanical stability, anticorrosion, tribology, electrical/thermal conduction, and other properties. Among applied zones, epoxy/three-dimensional graphene nanocomposites' physical attributes revealed enormous worth for electromagnetic and gamma ray shielding, corrosion protective coatings, and lubricants. Nevertheless, research seems restricted to design variations and applications of epoxy/three-dimensional graphene nanocomposites. Similarly, scientific reports lack sufficient information on interfacial and property enhancement mechanisms and foremost challenges limiting research margins on these hybrids. Hence, this article unveils the state-of-the-art of epoxy/three-dimensional graphene nanocomposites and can serve as a valuable guide for field researchers.

Keywords: epoxy; three-dimensional graphene; nanocomposites; synthesis; properties; radiation shielding; anticorrosion

1. Introduction

With the nanotechnological advancements, different nanocarbon nanofillers have been designed as polymeric reinforcements to form high-performance nanomaterials [1]. Like graphene, three-dimensional graphene has also been used as a nano-additive to form advanced hybrids [2]. Three-dimensional graphene is simply a modified hierarchically grown nano-assembly of graphene [3,4]. Due to the three-dimensionally interlinked nanoarchitecture, the hierarchical graphene exhibited several advantages related to structure, characteristics, and overall physical performance, compared with two-dimensional parent graphene nanosheets [5]. In thermosetting matrices, like epoxies, three-dimensional graphene has been applied as an effective reinforcement [6–8]. Consequently, epoxy/three-dimensional graphene nanocomposites have been designed using facile solution casting, curing, freeze drying, infiltration, and other facile techniques [9]. These nanocomposites have been explored for a range of morphological, conducting, thermal, mechanical, barrier, and other valued properties [10]. With fine electrical conductivity, unique porous architecture, and mechanical stability, practical fields of epoxy/three-dimensional graphene were noted for electromagnetic interference shielding and anticorrosion coatings [11–13]. Owing to fine interfacial interactions, load transfer, and antifriction and anti-wear properties, these nanocomposites have been applied as lubricating additives for engineering applications [14].

In addition to experimental studies so far on designs and performance of epoxy/three-dimensional graphene, theoretical and computational investigations must be performed to overcome the field challenges and further advances [15]. Furthermore, large-scale production, environmental safety, and sustainability aspects of epoxy/three-dimensional graphene hybrids have not been focused up till now.

With reference to previous research, this review article valuably surveys the current research state of an important thermosetting polymer (i.e., epoxy) and three-dimensional graphene-based nanocomposites. In due course, fundamentals, manufacturing, and physical performance of epoxy/three-dimensional graphene nanocomposites have been reviewed. These nanomaterials can be applied for radiation defense, corrosion resistance, and lubrication materials. This valuable review also attempts to highlight principal field challenges that limit the research evolution of epoxy/three-dimensional graphene hybrids. According to the potential of three-dimensional graphene-based hybrids, future research can be expanded to the environmental remediation and biomedical zones of epoxy/three-dimensional graphene nanocomposites.

2. Thermosetting polymers: Epoxies

Thermosets are polymeric materials that have irreversible crosslinking between the macromolecular chains and permanent hardening [16]. These polymers are tough, rigid, and hard in nature [17]. Epoxies or epoxy prepolymers or polyepoxide are the most common types of thermosetting polymers with epoxide functionalities in their backbones [18,19]. Generally, bifunctional compounds (diamines, diols, dianhydrides, etc.) have been used for curing, setting, or hardening of epoxies [20–23]. After setting, epoxies usually exhibit superior heat stability, mechanical properties, thermal conductivity, anticorrosion, wear, and other physical properties [24–26]. Diglycidyl ether of bisphenol A (DGEBA) is the most commonly used bifunctional epoxy resin in pristine, blended, or composite form for applied purposes [27,28]. Novolac has been generally studied as a multifunctional epoxy resin [29,30]. Relative to bifunctional epoxies, multifunctional epoxy resins have abundant crosslinking between macromolecular chains [31,32]. Some polyurethane backbones have also been designed with irreversible crosslinking and to be applied as thermosetting resins [33]. Applications of epoxy resins have been noted for wide-ranging technical industries, such as devices, anticorrosion/antifriction coatings, aerospace, automobiles, civil engineering, etc. [34–36] With the advancements in materials sciences, epoxy resins have been reinforced with numerous inorganic or carbon nanofillers to form high-tech nanocomposites for advanced industries [37,38].

3. Three-dimensional graphene

Graphene is a two-dimensional nanosheet-like nanocarbon, which is made up of sp2 hybridized carbon atoms [39,40]. In other words, it is one layer out of a stacked graphite structure [41]. To date, numerous sophisticated and facile top-down/bottom-up techniques have been devised to form graphene, namely exfoliation (mechanical/thermal), chemical vapor deposition, laser ablation, plasma, pyrolysis, electrochemical, and chemical routes [42,43]. Consequently, graphene has been synthesized with superior surface area, mechanical strength, heat stability, optical transparency, thermal conductivity, electrical conductivity, and other beneficial physical properties [44,45]. Subsequently, applied areas of graphene include aerospace/automotive structures, energy storage/production devices, electronic devices, and medical sectors (**Figure 1**) [46–48]. The increasing scientific curiosities in the field of graphene also led to the formation of modified graphene nanostructures and hybrids [49].

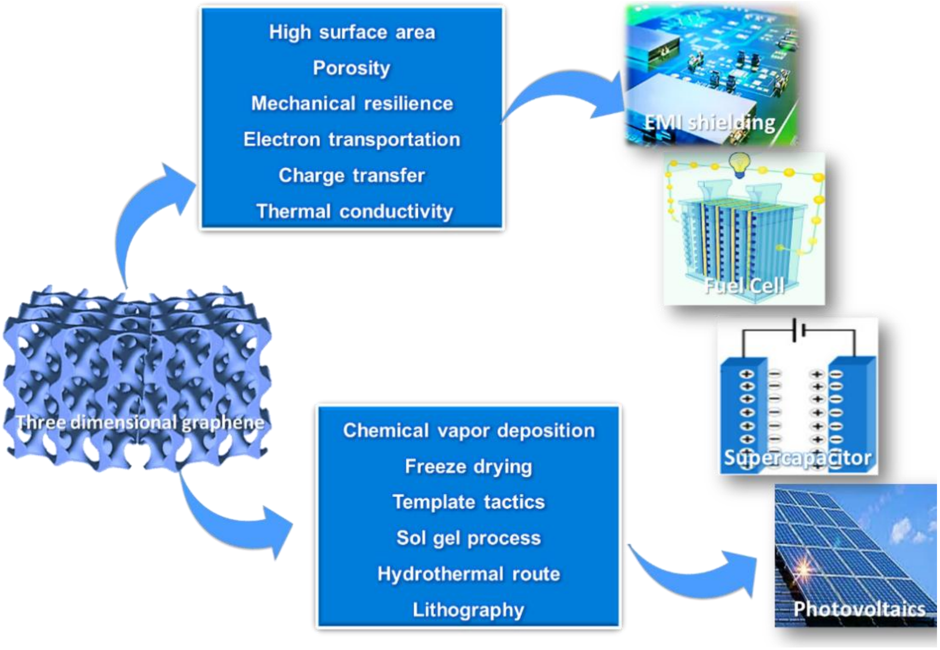

Figure 1. Three-dimensional graphene—Synthesis, properties, applications.

Three-dimensional graphene has appeared as one of the unique advancements in the field of graphene (**Figure 2**). These are hierarchical graphene nanostructures, which have also been reported as graphene nanofoams, aerogels or sponges in the past research [50]. In these nano-assemblies, two-dimensional graphene nano-sheets usually link via weak forces to form hierarchical nano-structures [51]. For synthesizing three-dimensional graphene, several techniques have been adopted so far, including freeze drying, hydrothermal, solvothermal, templating, direct deposition, sol-gel, and chemical methods [52–54]. Numerous experimental and theoretical studies have been reported so far, disclosing the structure of three-dimensional graphene [51]. Three-dimensional graphene also exhibited enormously high surface area, porosity, mechanical resilience, electron transportation, thermal conductivity, and other valuable physical properties [55,56]. In addition, applications of these hierarchical graphene nanostructures have been observed in sectors such as energy, environmental remediation, engineering, biomedical appliances, etc. [57–59].

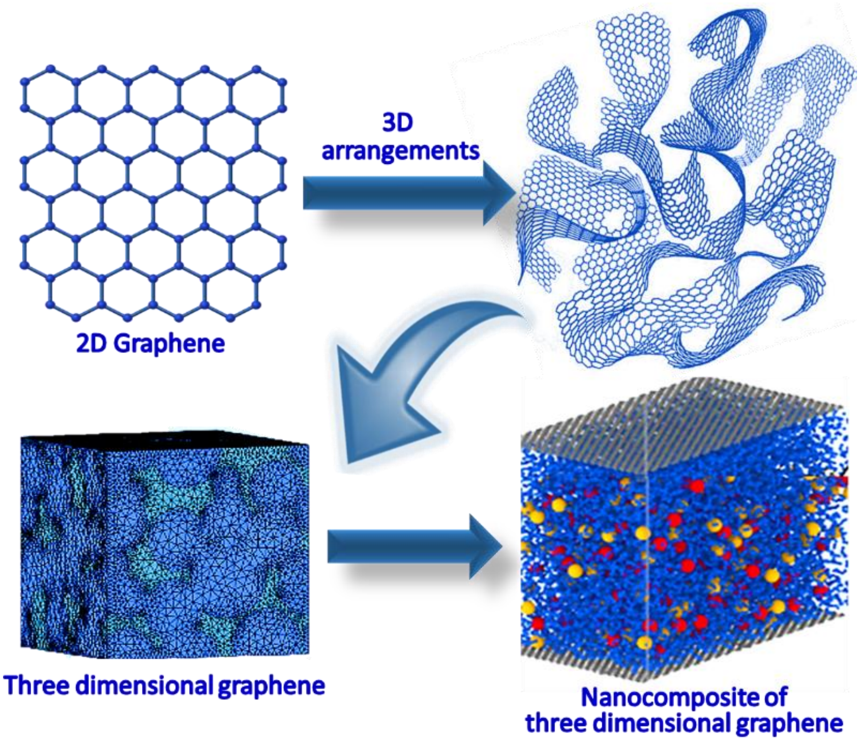

Figure 2. From three-dimensional graphene to derived nanomaterials.

4. Epoxy matrix reinforced three-dimensional graphene nanocomposites

Epoxy resins have been identified as ideal thermosets for nanocarbon reinforcements, such as graphene [60,61]. Similar to graphene, three-dimensional graphene has also been filled in epoxies like diglyceryl ether of bisphenol A [62]. Among earlier attempts, Gao et al. designed three-dimensional graphene by the freeze-drying method and applied it to reinforce epoxy resin using a simple curing technique [63]. The samples were then analyzed for morphological and thermal properties. **Figure 3A** shows the formation of graphene oxide using Hummer's method and then conversion to three-dimensional graphene oxide by the freeze-drying technique. Different amounts of three-dimensional graphene oxide were later added to epoxy resin and cured using methylhexahydrophthalic anhydride. The samples were thermally cured at high temperatures (>130 °C). **Figure 3B** shows scanning electron microscopy images of pristine epoxy, 0.3, and 1.3 wt.% three-dimensional graphene oxide nanofiller, respectively. The epoxy matrix had a smooth layered surface, while wrinkled surfaces were observed in the nanofiller-loaded samples. Due to the brittleness of the fractured surfaces of epoxy/three-dimensional graphene oxide hybrids, graphene oxide nanosheets can be seen pulled out of the matrix. **Figure 3C** illustrates storage modulus vs. temperature trends for epoxy/three-dimensional graphene oxide hybrids with 1.3 wt.% nanofiller at different frequencies. It was observed that the maxima of the storage modulus, tan δ, curve were increased with rising frequencies. These results showed the dependence of the glass transition of epoxy/three-dimensional graphene oxide on the frequency changes. **Figure 3D** presents thermal conductivity and thermal diffusivity plots of epoxy nanocomposites with three-dimensional graphene oxide loading. Higher thermal conductivity was observed for the 1.3 wt.% three-dimensional graphene oxide loaded sample (0.62 $Wm^{-1}K^{-1}$), relative to neat epoxy (0.25 $Wm^{-1}K^{-1}$) due to better phonon transfer tendency of the nanofiller. The thermal diffusivity of nanocomposites did not exhibit any remarkable improvement in the property.

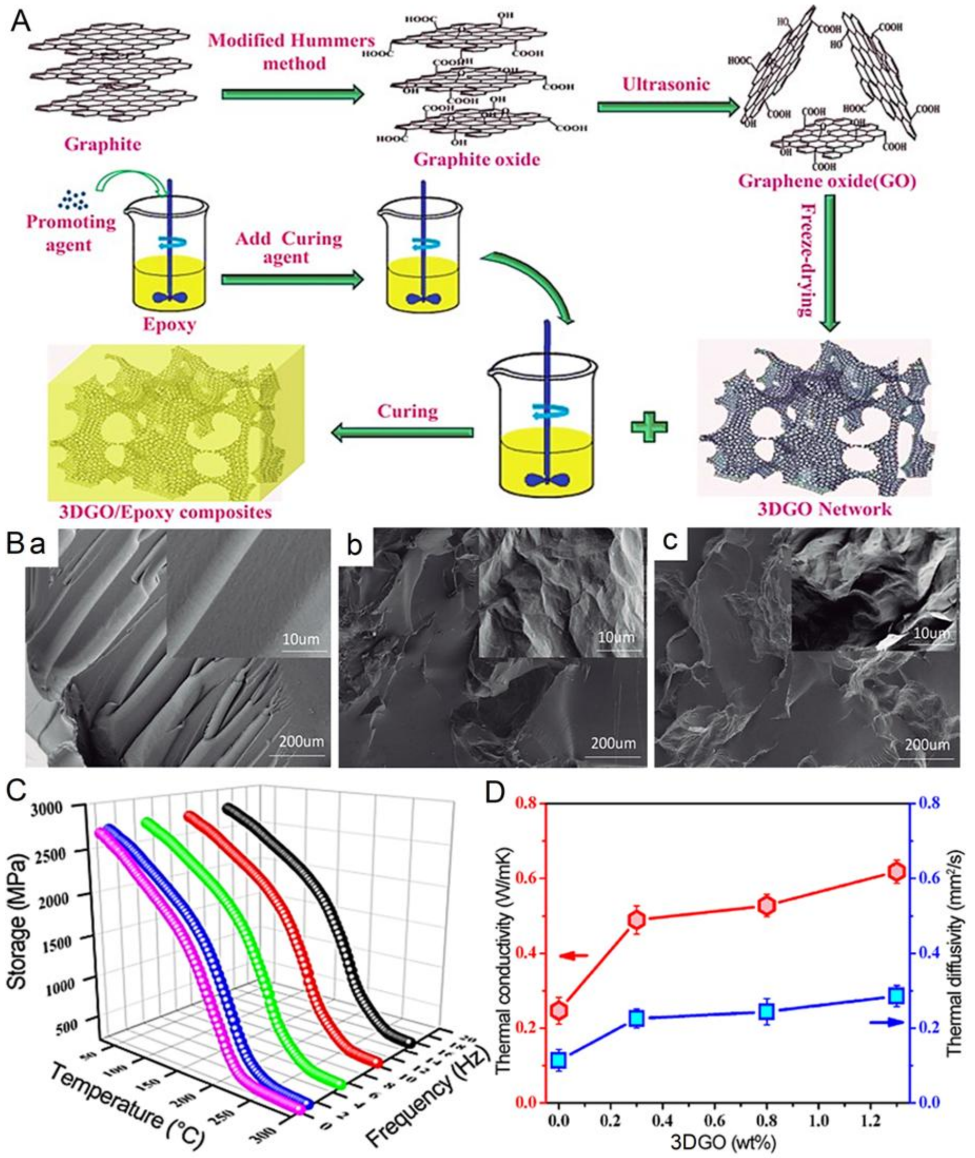

Figure 3. (**A**) Schematic for the synthesis of three-dimensional graphene oxide (3DGO) and epoxy nanocomposites; (**B**) Scanning electron microscopy images of: (a) neat epoxy; (b) epoxy nanocomposite with 0.3 wt.% 3DGO; (c) epoxy nanocomposite with 1.3 wt.% 3DGO, Insets: high magnification images of the samples; (**C**) storage modulus vs. temperature scans for 1.3 wt.% nanocomposite at different frequencies; (**D**) thermal conductivity and thermal diffusivity profiles of neat epoxy and nanocomposites with rising nanofiller contents [63]. Reproduced with permission from Springer.

Jia et al. designed three-dimensional graphene on a nickel template and then impregnated it with epoxy resin and cured it to form epoxy/three-dimensional graphene nanocomposites [64]. **Figure 4A** displays an optical image of an epoxy/three-dimensional graphene nanocomposite with pores of different shapes. Scanning electron microscopy micrograph of nanocomposite (0.2 wt.%) demonstrated a large block of matrix having consistent pores due to the insertion of nickel foam templated three-dimensional graphene (**Figure 4B**). Adding 0.2 wt.% or higher three-dimensional graphene contents increased the glass transition temperature to >126 ℃, relative to neat epoxy (95 ℃), due to the reinforcement effect of graphene-based nanofiller (**Figure 4C**).

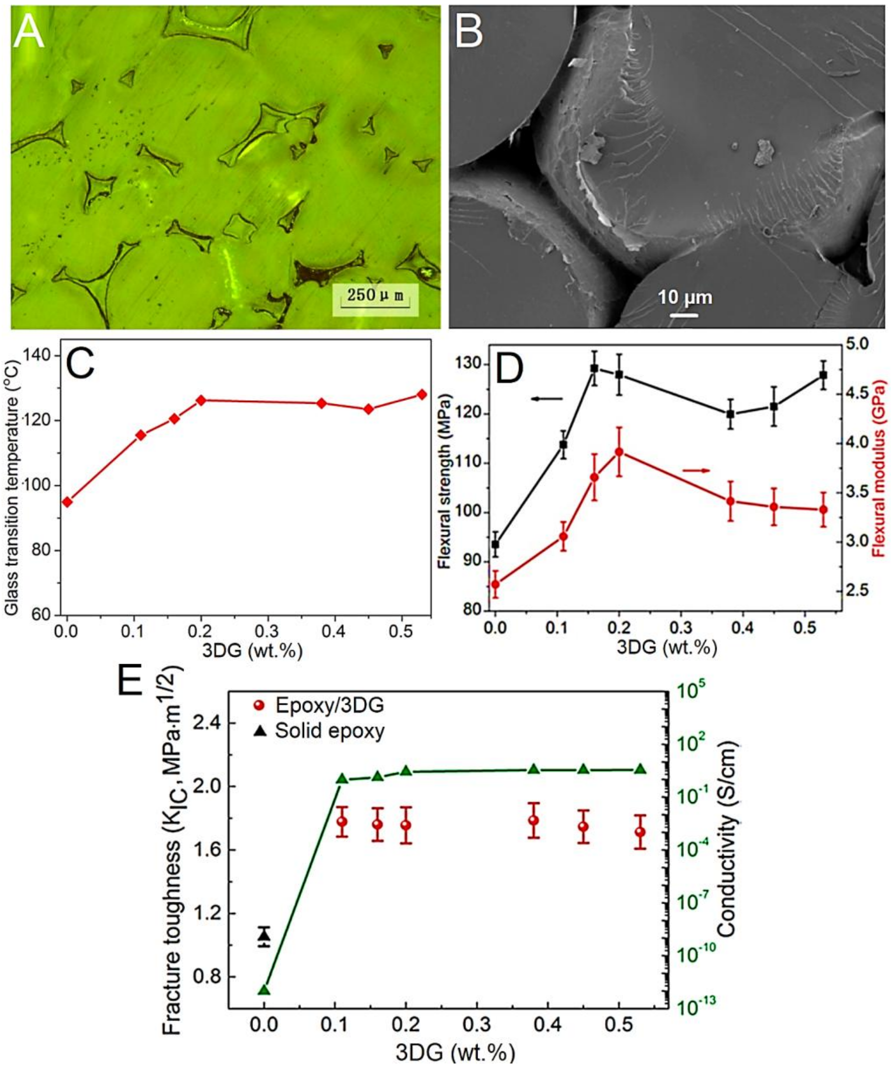

Figure 4. (**A**) Optical image of epoxy/three-dimensional graphene nanocomposite surface with pore distribution; (**B**) scanning electron microscope images of nanocomposites with 0.2 wt.% nanofiller; (**C**) glass transition temperature of nanocomposites vs. three-dimensional graphene contents; (**D**) flexural strength and modulus of epoxy/three-dimensional graphene nanocomposites vs. nanofiller contents; (**E**) fracture toughness and electrical conductivity of pristine epoxy and epoxy/three-dimensional graphene nanocomposites with rising nanofiller contents [64]. 3DG = three-dimensional graphene. Reproduced with permission from ACS.

Figure 4D shows flexural strength and modulus trends of epoxy/three-dimensional graphene nanocomposites with rising nanofiller contents. Addition of 0.2 wt.% three-dimensional graphene led to an increase of 38% and 53%, enhancements in flexural strength and modulus of hybrids, respectively. The increase in mechanical properties can be attributed to robust matrix-nanofiller interfacial adhesion. **Figure 4E** depicts fracture toughness and thermal conductivity profiles of pristine epoxy and epoxy/three-dimensional graphene nanocomposites with increasing nanofiller contents. The 0.1 wt.% loading of three-dimensional graphene enhanced fracture toughness by 70%, as compared to neat epoxy, due to strong interfacial linkages. However, the property decreased at higher loading levels, possibly due to weakening of interfacial bonding against applied load. The maximum value of electrical conductivity for the 0.1 wt.% hybrid obtained was around 6.4 Sm^{-1}, and beyond that loading level, the property showed a constant trend. The results imply that 0.1 wt.% was the optimal amount to form effective percolation networks in these hybrids.

Embrey et al. used commercial three-dimensional graphene and formed epoxy/three-dimensional graphene and epoxy/graphene hybrids using dip coating and curing approaches [65]. **Figure 5A** shows a scanning electron microscopy image of an epoxy/three-dimensional graphene hybrid. The micrograph exhibited an interfacially interlinked matrix-nanofiller microstructure responsible for the overall compatibility effects of the nanocomposites.

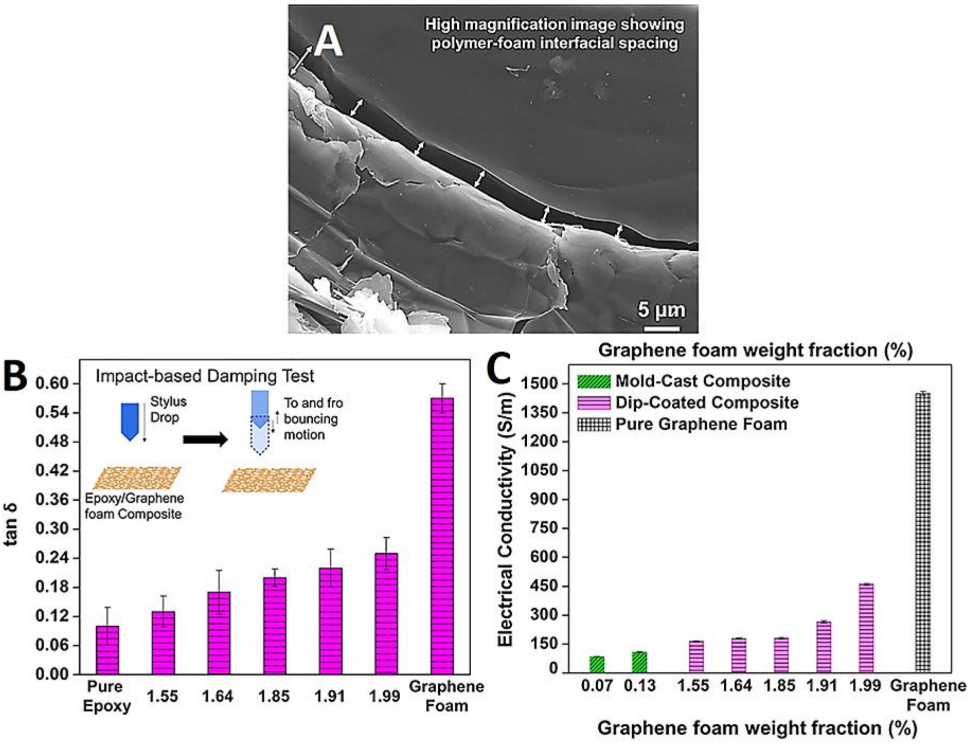

Figure 5. (**A**) High resolution field emission scanning electron microscopy micrograph showing epoxy/three-dimensional graphene nanocomposite; (**B**) loss tangent values for pure epoxy, three-dimensional graphene, and epoxy/three-dimensional graphene nanocomposite; and (**C**) electrical conductivity values for pristine three-dimensional graphene and epoxy/three-dimensional graphene nanocomposite fabricated by dip coating and mold casting vs. three-dimensional graphene weight fraction [65]. Graphene foam = three-dimensional graphene. Reproduced with permission from ACS.

Figure 5B demonstrates loss tangent plots of neat epoxy, three-dimensional graphene, and epoxy/three-dimensional graphene nanocomposite with varying nanofiller contents. tan δ was observed to be 140% higher for 1.99 wt.% loaded nanocomposite, relative to neat epoxy resin. In **Figure 5C**, addition of three-dimensional graphene caused a considerable improvement in the electrical conductivity (461 Sm^{-1}), as compared to the pristine epoxy resin (10^{-9} Sm^{-1}). This can be due to the formation of well-interconnected percolation network throughout the epoxy matrix, responsible for continuous electron conduction. In addition, the electrical conductivity of the epoxy/three-dimensional graphene hybrid prepared via dip coating was found to be higher than the casted samples, showing the effectiveness of this technique to form compatible nanocomposites.

Han et al. developed an ultra-stiff three-dimensional graphene by the chemical vapor deposition route [66]. Later, they fabricated an epoxy/ultra-stiff three-dimensional graphene hybrid through vacuum-based resin infiltration. **Figure 6A** shows photographs of as-prepared pristine epoxy and nanocomposite samples.

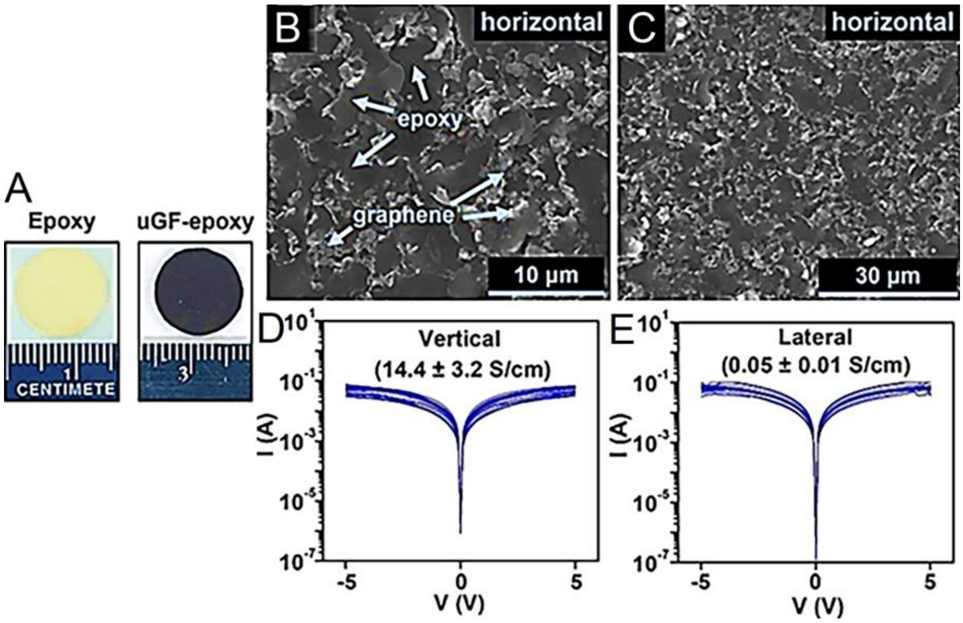

Figure 6. Characterization of epoxy/three-dimensional graphene nanocomposite; (**A**) photographs of neat epoxy and nanocomposite; (**B&C**) scanning electron microscopy images (horizontal cross section); and current voltage (I V) curves in (**D**) vertical direction and (**E**) lateral direction [66]. uGFs-epoxy = ultra stiff three-dimensional graphene filled epoxy nanocomposite. Reproduce with permission from ACS.

Scanning electron microscopy micrographs in **Figure 6B**,**C** exhibited regular arrangement, consistent dispersion, and network formation of ultra-stiff three-dimensional graphene in the matrix. According to current voltage scans of epoxy/ultra-stiff three-dimensional graphene nanocomposite in vertical and lateral directions, the electrical conductivity was found to be 14 and 0.05 Scm^{-1}, respectively (**Figure 6D**,**E**). The superior electrical conductivity of the hybrid in the vertical direction depicted better alignment and network formation of three-dimensional graphene in that direction. Similarly, Wang et al. worked on hybrids of epoxy/polyaniline reinforced with three-dimensional graphene obtained via *in situ* and casting techniques [67]. These hybrids revealed fine percolation and electrical conductivity properties (0.04 Scm^{-1}). The porous hierarchical nanostructures of three-dimensional graphene and polyaniline conducting phase seemed to be responsible for superior electron conduction through the system [68].

5. Promising technical aspects of epoxy/three-dimensional graphene hybrids

Similar to thermosetting/graphene nanocomposites, three-dimensional graphene-based hybrids have gained enormous scientific interest for technological industries [69]. There are significant utilizations of thermosetting polymers and three-dimensional graphene-based nanomaterials have been noted for the fields of radiation protection, anticorrosion, and tribological or lubricant materials [70,71].

Electromagnetic radiation pollution has become a serious threat to our entire ecosystem due to the growing use of electronics and modern devices, and equipment [72,73]. To prevent the rising environmental and health hazards of undesirable electromagnetic radiations in the atmosphere, efficient electromagnetic interference shielding materials have been developed [74,75]. Graphene and derivative nanostructures have been inspected for nuclear, gamma, or cosmic ray shielding [76–78]. In recent years, three-dimensional graphene has been considered as an advanced form of graphene used for gamma-ray electromagnetic interference shielding [79–81]. For instance, Guo et al. fabricated three-dimensional graphene oxide via freeze drying technique to study radiation shielding effectiveness [82]. The three-dimensional graphene oxide-based shield (3 mm thick) has electrical conductivity and shielding effectiveness of >700 Sm^{-1} and 69 dB, respectively. In addition, epoxy/three-dimensional graphene nanocomposites have been studied as high-tech radiation shields [83,84]. Wang et al. manufactured epoxy/three-dimensional graphene nanocomposites via resin transfer molding [85]. These nanocomposites showed superior compression and flexural strength (84 and 56%, respectively), relative to pristine systems. Most importantly, epoxy/three-dimensional graphene hybrids revealed gamma ray shielding efficiency and > 90% retention of mechanical properties even over prolonged exposure. Liang et al. filled graphene nanoplatelets along with three-dimensional graphene in the epoxy matrix to form multiphase nanomaterials [86]. The synergistic effects of graphene nanofillers led to electrical conductivity of approximately 180 Sm^{-1} and a radiation shielding effectiveness of approximately 51 dB was observed. Wang et al. developed epoxy, polyaniline, and three-dimensional graphene-derived nanocomposites using an *in situ* and solution curing route [87]. These nanomaterials were tested for durable radiation shielding applications. **Figure 7A** shows hardness and reduced modulus plots of pristine epoxy and epoxy/polyaniline/three-dimensional graphene nanocomposites with different loadings of three-dimensional graphene nanofiller. Consequently, the nanocomposite had superior hardness and reduced modulus of 0.16 GPa and 3.84 GPa, respectively, relative to neat epoxy (0.09 GPa and 2.74 GPa, correspondingly). It seems that polyaniline acted as a bridge to interlink and strengthen the interfacial effects in epoxy/polyaniline/three-dimensional graphene nanocomposites. **Figure 7B** presents the electromagnetic interference shielding effectiveness of neat epoxy, and epoxy hybrids with three-dimensional graphene, polyaniline/three-dimensional graphene, and p-phenylenediamine grafted three-dimensional graphene nanofillers. The epoxy/polyaniline/three-dimensional graphene nanocomposite had higher electromagnetic interference shielding effectiveness (>36 dB), relative to neat epoxy (3.1 dB) as well as epoxy/three-dimensional graphene nanocomposite (31 dB). These results depicted that the presence of conducting polyaniline phase and percolation network formation in epoxy/polyaniline/three-dimensional graphene nanocomposite, supporting the electron flow and radiation absorption properties. **Figure 7C** illustrates the probable mechanism of electromagnetic interference shielding in these nanocomposites. The radiation shielding phenomenon was proposed to be due to reflection at the interfaces of epoxy/polyaniline/three-dimensional graphene nanocomposite.

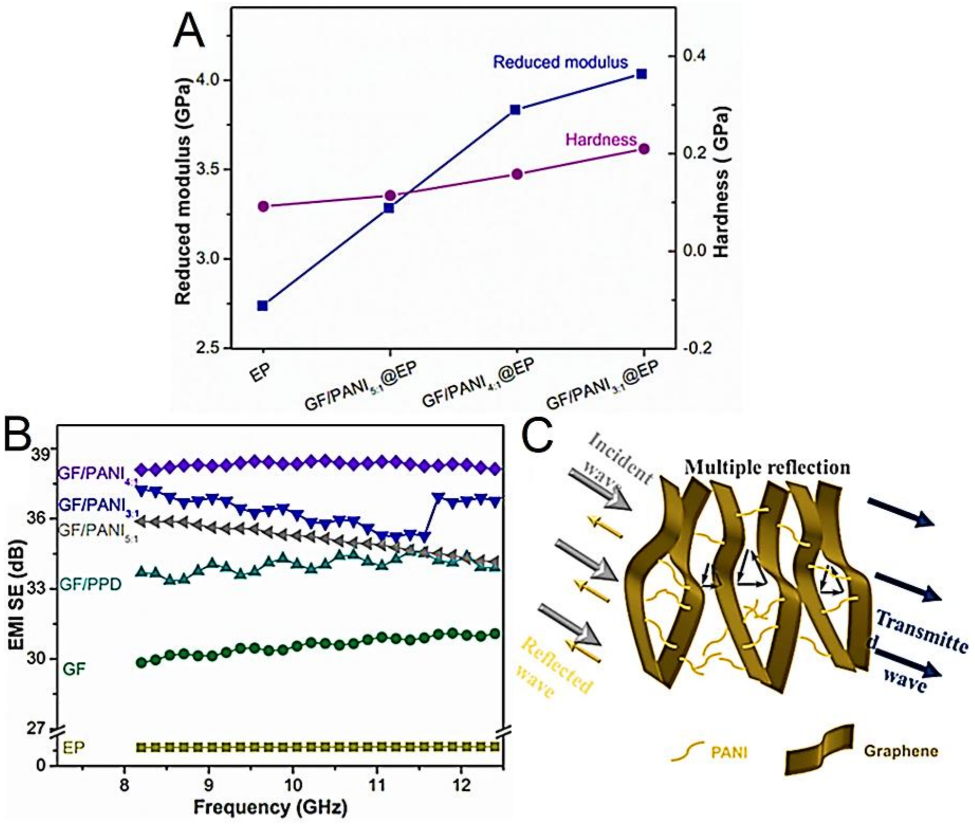

Figure 7. (**A**) Hardness and reduced modulus of the EP, GF/PANI5:1@EP, GF/PANI4:1@EP, GF/PANI3:1@EP nanocomposites; (**B**) Electromagnetic interference shielding effectiveness (EMI SE) of neat epoxy, three-dimensional graphene, polyaniline and three-dimensional graphene-based nanocomposite, and p-phenylenediamine grafted three-dimensional graphene hybrid; (**C**) Schematic of electromagnetic interference shielding mechanism [87]. GF/PANI = three-dimensional graphene/polyaniline; GF = three-dimensional graphene; GF/PPD = three-dimensional graphene grafted p-phenylenediamine; PANI = polyaniline; EP = epoxy. Reproduced with permission from MDPI.

In the field of corrosion-resistant materials, graphene has been studied as a barrier to resist corrosive media [88]. Besides pristine graphene, high-performance polymer/graphene nanocomposite coatings have been industrialized and studied for corrosion-resistant applications [89]. Traditionally, epoxy/graphene-based anticorrosion coatings have been reported [90]. Research progresses on graphene and hybrids led to the formation of corrosion-resistant three-dimensional graphene-based nanomaterials [91]. The epoxy/three-dimensional graphene nanocomposites with anticorrosion potential have been reported [92,93]. Guan et al. explored epoxy/zinc and epoxy/zinc reinforced with graphene or three-dimensional graphene nanocomposites for anticorrosion purposes [94]. Three-dimensional graphene was synthesized by the flame method, and template and dip coating approaches were used to form the epoxy-based hybrids. As compared to graphene-filled epoxy/zinc hybrids, the epoxy/zinc/three-dimensional graphene system revealed percolation properties and electrical conductivity due to the formation of a hierarchical porous nanostructure. The epoxy/zinc/three-dimensional graphene hybrids had valuable corrosion resistance and barrier properties of the coating hybrid coatings. Qin et al. also formed zinc-rich epoxy/graphene nanocomposites and zinc-rich epoxy/three-dimensional graphene nanocomposites and applied them as coating materials on Q235 steel (**Figure 8A**) [95]. Graphene or three-dimensional graphene was prepared by using chemical vapor deposition. Epoxy was filled with zinc powder and then graphene or three-dimensional graphene through a solution curing approach with polyamine curing agent. For corrosion resistance analysis, Tafel polarization studies were performed for neat zinc-rich epoxy, zinc-rich epoxy/graphene, and zinc-rich epoxy/three-dimensional graphene coatings after prolonged immersion in seawater (**Figure 8B**). Zinc-rich epoxy/three-dimensional graphene with 0.1 wt.% loading had notably higher corrosion potential than pristine and graphene-filled coatings. These outcomes can be attributed to the development of a consistent nanoporous framework of three-dimensional graphene in the epoxy matrix, thereby promoting the electron transfer through the system. **Figure 8C**,**D** illustrate scanning electron microscopy micrographs of epoxy/three-dimensional graphene nanocomposites before and after immersion in seawater. After the corrosion test, the nanocomposite had a porous morphology, possibly due to the oxidation of zinc particles in the matrix. Thus, the addition of inorganic nanoparticles, like zinc, along with three-dimensional graphene opened a valuable route to next-level anticorrosion coatings.

According to the literature, tribological properties and lubrication potential of epoxy resins and derivative materials have been studied [96]. Similarly, graphene and its polymeric nanocomposites have gained scientific worth for tribological applications [97]. However, recent research has focused on the effectiveness of porous three-dimensional graphene nanoarchitectures for lubrication, rather than pristine graphene [98]. The epoxy/three-dimensional graphene hybrids have been used as lubricating materials [99–101]. Despite a large amount of research done in the past, future attempts must be made to design high-performance epoxy/three-dimensional graphene-based lubricants.

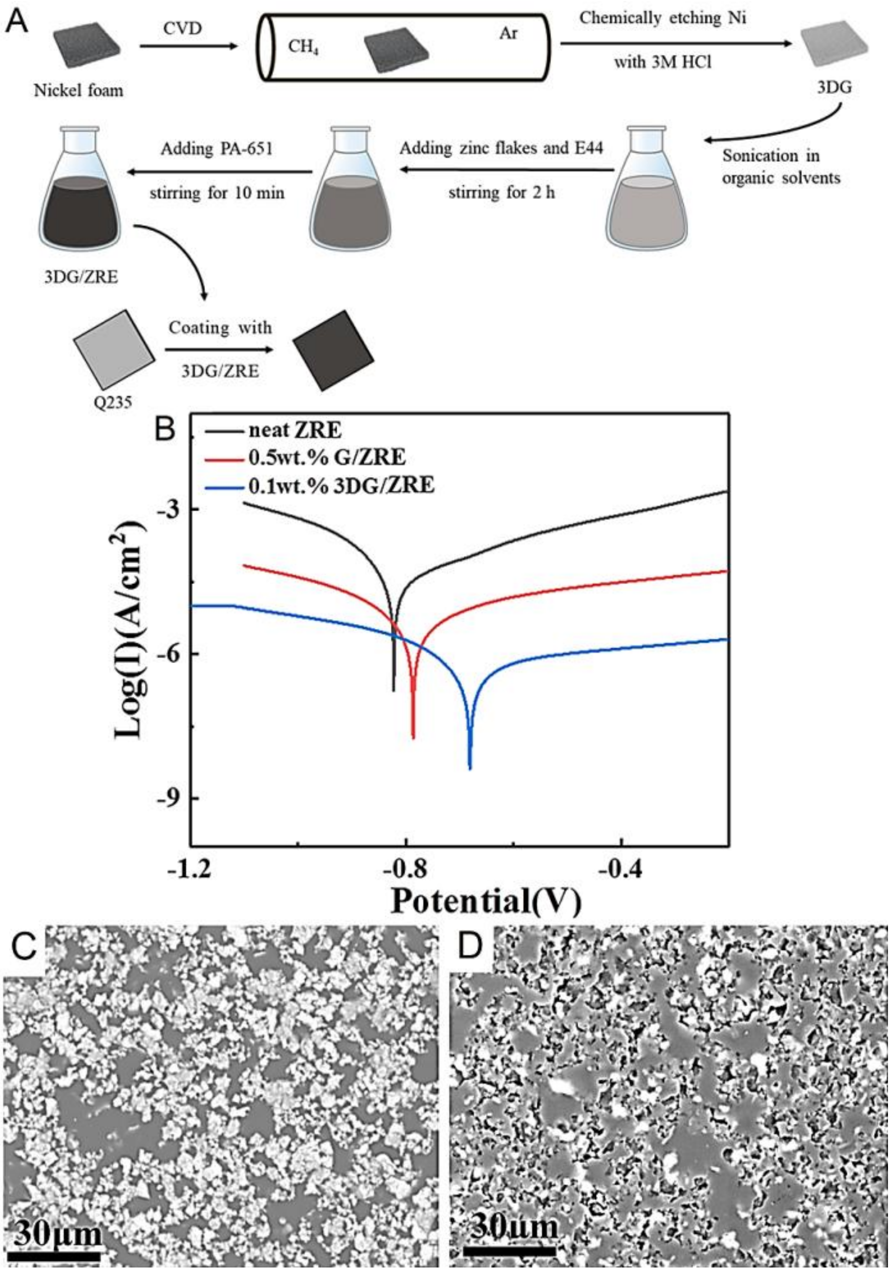

Figure 8. (**A**) Schematic of the preparation of zinc rich epoxy/three-dimensional graphene nanocomposite and its coating on Q235 steel plate; (**B**) Tafel curve of neat zinc rich epoxy; zinc rich epoxy/graphene, and zinc rich epoxy/three-dimensional graphene coated Q235 steel plates after 300 h immersion in simulated seawater; scanning electron microscopy images of (**C**) epoxy/three-dimensional graphene before and (**D**) after 300 h immersion in sea water [95]. 3DG/ZRE = three-dimensional graphene filled zinc rich epoxy; G/zinc rich epoxy = graphene filled zinc rich epoxy; ZRE = zinc rich epoxy; CVD = chemical vapor deposition; PA-651= polyamine curing agent. Reproduced with permission from MDPI.

Table 1 shows an inclusive overview of literature surveys so far on epoxy/three-dimensional graphene nanocomposites to highlight their design specifications, fabrication routes, indispensable physical properties, and applications. In summary, research to date discloses some important systems of thermosetting hybrids reinforced with three-dimensional graphene nanofiller. As argued in this novel manuscript, it looks evident that three-dimensional graphene acted as an advantageous nanofiller for epoxy matrices, relative to pristine two-dimensional graphene nanosheet. The most obvious reason can be attributed to the formation of porous, hierarchical, and interfacially compatible epoxy-three-dimensional graphene nanostructures. Three-dimensionally networked nano-level consistency has been found to support the electrical conductivity, mechanical resilience, heat stability, rusting/chemical resistance, and countless other properties of epoxy-based nanocomposites. Additionally, these nanomaterials have notable structure-property links supporting their practical applications for electromagnetic interference or gamma shields, corrosion-resistant coatings, and lubricants (**Figure 9**).

Table 1. Design, physical features, fabrication, and application areas of epoxy/three-dimensional graphene nanocomposites.

Matrix	Nanofiller/additive	Fabrication strategies	Properties/Applications	Ref
Epoxy	Three-dimensional graphene	Nickel foam template; infiltration technique	Electrical conductivity 6.4 Scm^{-1}; fracture toughness 1.78 MPa·m$^{1/2}$ (70% higher than unfilled system); 0.2 wt.% nanofiller caused 38 and 53% enhancements in flexural strength and modulus, respectively, glass transition temperature > 126 °C	[64]
Epoxy; methylhexahydrophthalic anhydride	Three-dimensional graphene oxide	Freeze drying; thermal curing technique (>130 °C)	Storage modulus; thermal conductivity 0.62 Wm^{-1}K^{-1}; thermal diffusivity	[63]
Epoxy	Three-dimensional graphene	Dip coating; curing	1.99 wt.% nanofiller caused 140% higher tan δ than the neat matrix; higher electrical conductivity (461 S m^{-1}), as compared to the pristine epoxy resin (10^{-9} S m^{-1}); interfacially interlinked	[65]
Epoxy	Ultra-stiff three-dimensional graphene	Vacuum-based resin infiltration	Electrical conductivity 14 and 0.05 Scm^{-1} for vertical and lateral directions, respectively	[66]
Epoxy/polyaniline	Three-dimensional graphene	In situ method; casting techniques	Percolation network; electrical conductivity 0.04 Scm^{-1}	[67]
Epoxy	Three-dimensional graphene	Freeze-drying technique	Electrical conductivity and shielding effectiveness >700 Sm^{-1} and 69 dB, respectively	[82]
Epoxy	Three-dimensional graphene	Resin transfer molding	Compression and flexural strength of 84 and 56%, respectively; >90 % retention of mechanical properties in radiation	[85]
Epoxy	Three-dimensional graphene; graphene nanoplatelets	Curing; casting	Electrical conductivity approximately 180 Sm^{-1}; radiation shielding effectiveness approximately 51 dB	[86]
Epoxy/polyaniline	Three-dimensional graphene	*In situ* method; solution curing	Hardness and reduced modulus of 0.16 GPa and 3.84 GPa, respectively; electromagnetic interference shielding effectiveness >36 dB	[87]
Epoxy	Three-dimensional graphene	*In situ* method; solution curing	Electromagnetic interference shielding effectiveness >31 dB, higher than neat resin 3.1 dB	[87]
Epoxy	Three-dimensional graphene; zinc particles	Flame method; template technique; dip coating	Hierarchically porous nanostructures; percolation properties; electrical conductivity; anticorrosion	[94]
Epoxy; polyamine	Three-dimensional graphene; zinc particles	Chemical vapor deposition; solution casting	Anticorrosion coatings; coating materials on Q235 steel; Tafel polarization studies; seawater immersion tests	[95]

Epoxy	Three-dimensional graphene; graphene nanoplatelets	Curing; casting	Tribological properties; lubrication	[98–101]

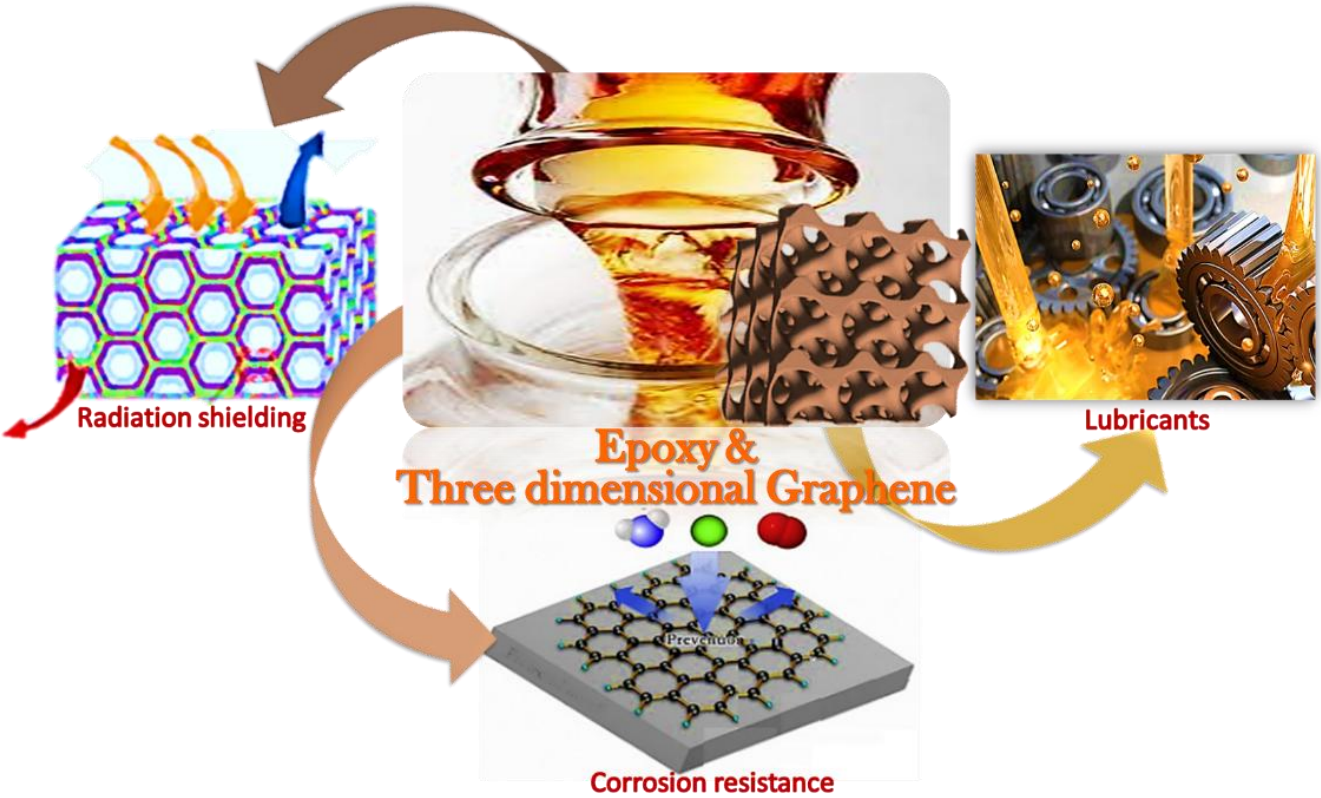

Figure 9. Applications of epoxy/three-dimensional graphene-derived nanocomposites.

6. Conclusions and viewpoints

Nevertheless, future progress in all the mentioned functional sectors of epoxy/three-dimensional graphene nanocomposites demands concentrated research examinations. According to literature reports so far, three-dimensional graphene has been commonly produced using freeze-drying and chemical vapor deposition routes, while template methods have been used in a few cases. For nanocomposite formation with polymers, mostly solution curing and infiltration methods have been applied. Moreover, further applied opportunities of these hybrids can be extended by controlling design, processing parameters, and underlying mechanisms. Advances in ecological epoxy/three-dimensional nanomaterials may lead to sustainable solutions for several high-tech industries. For instance, sustainable air/water environmental remediation systems can be developed for green ecosystems. Future applications of green epoxy and three-dimensional graphene can be seen in the fields of durable and inexpensive renewable energy systems and flexible electronics (e.g., wearable sensors). In the future, the performance of petroleum products can be remarkably enhanced by using environmentally friendly bio-based epoxies. Furthermore, green epoxy/three-dimensional nanocomposites can be used to design sustainable automobile parts, space structures, marine units, and defense applications. Last but not least, green and sustainable coatings/adhesives can be developed for further applications in the environment, energy, and space-related sectors. Herein, green manufacturing and scalability of epoxy/three-dimensional graphene yet need to be achieved by overcoming the challenges observed for processing/property control of lab-scale modules. Henceforth, future-focused studies by the field scientists can overcome the complexities in scaling up the epoxy/three-dimensional graphene for high-performance end uses.

Conflict of interest: The author declare no conflict of interest.

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