Journal of
Composites Science

Review

Unveiling the Influential Factors and Heavy Industrial Applications of Graphene Hybrid Polymer Composites

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Abstract: Graphene hybrid-filler polymer composites have emerged as prominent materials that revolutionize heavy industries. This review paper encapsulates an in-depth analysis of different influential factors, such as filler/graphene type, aspect ratios, dispersion methods, filler-matrix compatibility, fiber orientation, synergistic effects, different processing techniques, and post-curing conditions, which affect the processing and properties of graphene hybrid polymer composites, as well as their resultant applications. Additionally, it discusses the substantial role of graphene reinforcement with other fillers, such as carbon nanotubes, silica, nano-clays, and metal oxides, to produce functionalized hybrid polymer composites with synergistically enhanced tailored properties, offering solutions for heavy industries, including aerospace, automotive, electronics, and energy harvesting. This review concludes with some suggestions and an outlook on the future of these composite materials by emphasizing the need for continued research to fully optimize their potential.

Keywords: graphene; polymers; hybrid composites; influencing factors; applications

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Citation: Ali, Z.; Yaqoob, S.; Yu, J.; D'Amore, A. Unveiling the Influential Factors and Heavy Industrial Applications of Graphene Hybrid Polymer Composites. *J. Compos. Sci.* **2024**, *8*, 183. https://doi.org/10.3390/jcs8050183

Academic Editor: Francesco Tornabene

Received: 6 April 2024
Revised: 4 May 2024
Accepted: 9 May 2024
Published: 13 May 2024

1. Introduction

Graphene hybrid-filler polymer composites are a strong candidate in material science due to their exceptional properties and transformative implications in heavy industries. The remarkable mechanical, thermal, and electrical properties and lightweight of graphene composites are due to graphene, a two-dimensional material with sp^2 bonding of carbon atoms. The integration of graphene in polymer matrices with various fillers leads to multifunctional composites with magical properties that offer durability and competence under rigorous engineering conditions. Due to its high tensile strength ($\approx$1 TPa) [1], intrinsic mobility of charge carriers ($\approx$200,000 $cm^2\ V^{-1}\ s^{-1}$) [2], and intrinsic thermal conductivity ($\approx$5000 $W\ m^{-1}\ K^{-1}$ at room temperature) [3], graphene is ideal for producing polymer composites with substantial increase in strength, conductivity, and thermal stability. Moreover, the presence of graphene with multi-fillers, e.g., carbon nanotubes (CNTs), nano-clays, metal oxides, carbon black, silica, and one-dimensional nanowires plays a critical role in producing polymer composites with diverse properties [4]. The introduction of graphene with these fillers results in complex systems with synergistic effects [5] to produce hybrid, ternary, and quadra polymer composites, which open avenues for advanced applications in heavy industries [6]. For instance, graphene combined with CNTs creates an entangled structure and increases mechanical strength and electrical conductivity [7]. However, when combined with nano-clays, graphene promotes thermal stability and barrier properties [8]. Combining graphene with metal (e.g., silver and copper) nanoparticles offers higher electrical conductivity and antimicrobial properties [9,10]. Furthermore, when combined with boron nitride microparticles (micro-BN), graphene nanoplatelets (GNPs) significantly increase the thermal conductivity of epoxy composites [2].

However, the fabrication and performance of these polymer composites is highly influenced by several factors [11], including the type of polymer matrix, types and morphologies of multiple fillers, their concentrations, quality, and aspect ratio of graphene, as well as the processing methods of composite preparation [12]. The production and performance of graphene hybrid polymer composites are influenced by several key factors. The type and proportions of graphene used can have a significant impact on the mechanical, thermal, and electrical properties of the composite. The compatibility of the filler matrix, including the interfacial interaction between the graphene and the polymer, is essential to ensure effective load transfer and property improvement. The optimal concentrations of graphene and other fillers must be balanced to achieve improved properties without compromising processability. The uniform dispersion of graphene throughout the polymer matrix is critical to avoid agglomeration, which could reduce benefits. The orientation of graphene fibers or flakes, randomly distributed or aligned, can tailor the anisotropic properties of the composite. Synergistic effects between graphene and other nano-reinforcements, such as MXene, can lead to superior mechanical, thermal, and electrical performance. The processing techniques employed, such as solution mixing, in situ polymerization, or melt mixing, can influence dispersion, alignment, and interfacial interactions. Finally, post-treatment and functionalization of graphene can further improve compatibility with the polymer matrix and the overall properties of the composite [13,14].

The ability of graphene hybrid-filler polymer composites to withstand extreme conditions while maintaining their durability and efficiency makes them vital for fulfilling industrial demands. In heavy industries, graphene hybrid-filler polymer composites are utilized in manufacturing lightweight structural components, thermal management systems for electronic devices, and conductive elements in energy storage devices [15,16]. Moreover, graphene hybrid polymer composites have been extensively studied for crucial applications in the aerospace, automotive, electronics, and energy storage industries. These composites improve the stability and longevity of graphene in lithium-ion batteries (LIBs) by expanding layer spacing, preventing restacking, and providing additional accommodation sites for Li^+. The addition of carbon nanotubes (CNTs) or fullerenes into graphene nanosheets further increases the specific capacitance of LIBs [17]. Additionally, highly conductive 3D printable polymer-graphene hybrid composites have been developed for micro-supercapacitors and stretchable light-emitting displays. Fabricating highly conductive flexible graphene circuits via 3D printing provides excellent shielding from electromagnetic interference. These advances in graphene-based hybrid-filler polymer composites have the potential to significantly improve the performance and efficiency of several industries, such as aerospace, electronics, energy storage, and automotive [18]. Figure 1 represents global research trends on graphene hybrid polymer composites, including factors affecting processing and properties, as well as applications in heavy industries.

Thus, in this paper, we summarized the key factors that play a decisive role in the development and performance evaluation of graphene hybrid polymer composites, and the applications of these polymer composites in various industrial sectors were critically reviewed. In the end, a few suggestions and prospects will be proposed to overcome the structural flaws and fully harness the potential of these polymer composites.

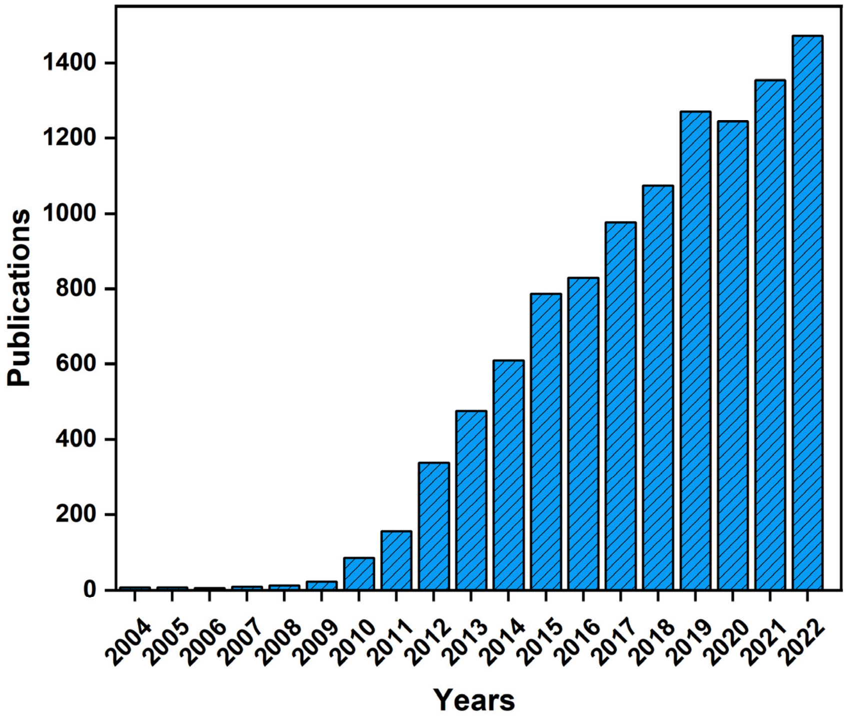


Figure 1. The number of articles on graphene hybrid polymer composites, applications in heavy industries, and affecting factors published over the years were reported in the Scopus database.

2. Influencing Factors of Graphene Hybrid Polymer Composites

2.1. Graphene Type and Aspect Ratio

Filler type and aspect ratio are the backbone of the fabrication of smart composites [19]. While dealing with graphene hybrid polymer composites, the choice of graphene type plays a decisive role in portraying the structural and functional properties of the final composite. Graphene comes in several forms, each with unique properties and applications. Here are the primary types: pristine graphene, graphene oxide (GO), and reduced graphene oxide (rGO), as shown in Figure 2. However, each type has its unique structural characteristics, which significantly influence the performance and tailor the properties of the resultant composites. For example, pristine graphene, a single layer of carbon atoms arranged in a hexagonal lattice, greatly boosts mechanical features such as tensile strength and Young's modulus of polymer composites due to its intrinsic structural organization and atomic-level thickness. Moreover, with a high aspect ratio, pristine graphene sheets present effective load transfer and stress distribution properties [20]. On the other hand, GO keeps oxygen-containing functional groups, which highly facilitate the dispersibility and strong interfacial interactions through polymer matrices, directing them to enhanced thermal stability and mechanical properties [21]. Reduced graphene oxide (rGO) is obtained by the reduction of GO and owns several oxygen functionalities. However, the exceptional structure of rGO supports effective charge transport in the composite compared to GO. It thus shows higher electrical conductivity and mechanical strength, making it an ideal candidate for applications involving electrical conductivity, e.g., sensors and conductive coatings [22].

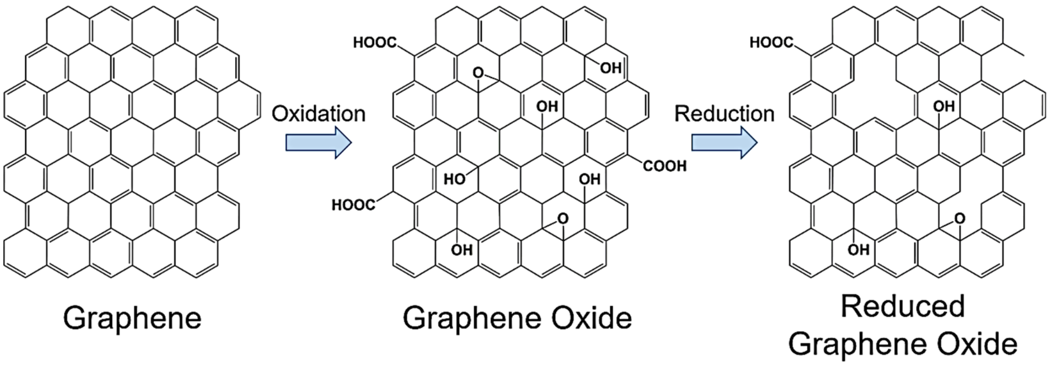


Figure 2. Different types of graphene. The chemical structures of graphene, graphene oxide, and reduced graphene oxide, along with their respective synthesis processes. Adopted with permission from ref. [23].

In recent years, various graphene derivatives and nanostructures have been employed to manufacture polymer composites for desired applications. Graphene nanoplatelets (GNPs), with their plate-like shape and high aspect ratio, improve the strength and toughness of polymer composites [24]. Graphene and graphene oxide quantum dots, with their quantum confinement effects and tunable electronic properties, optimize composite materials' electrical conductivity and optical characteristics [25]. Graphene nanoribbons (GNRs), with their one-dimensional structure, offer exceptional mechanical strength and superior thermal conductivity [26]. Ultralight and porous graphene aerogels (GAs) provide thermal stability and energy absorption properties [27]. Finally, graphene masterbatches simplify the incorporation of graphene into composites, ensuring uniform distribution for consistent performance [28]. These innovations enable the design of advanced composite materials with tailored properties for various industrial applications, opening new perspectives in materials engineering.

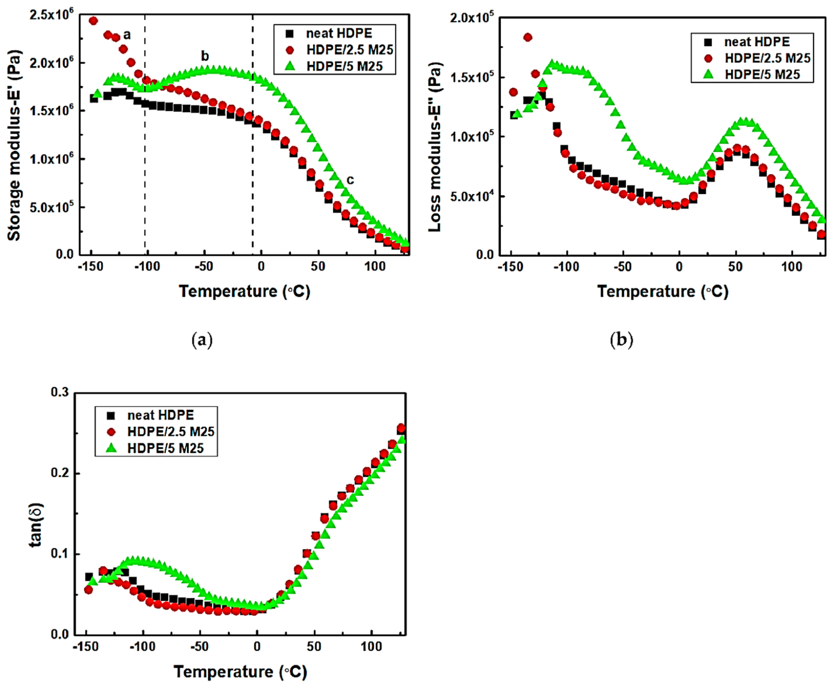
Additionally, graphene holds great promise due to its discrete dimensions, which allow it to combine with other fillers. For instance, combining graphene with CNTs maintains micrometer lengths and 10–30 nm diameters. This scheme establishes a synergistically infiltrated network through polymers, producing materials with superb electrical conductivity (1000–3000 S m^{-1}), mechanical strength, and thermal conductivity [29,30]. Furthermore, graphene combined with metal nanoparticles such as gold (Au), silver (Ag), and copper (Cu), offers nanoscale dimensions (<100 nm in diameter) and produces a highly connected network structure in the polymer matrix, presenting a higher electrical conductivity of 10^7 S m^{-1} [31,32]. Besides the dimension, the aspect ratio of graphene drastically impacts the structural features and properties of the polymer composites. The higher aspect ratio promotes better dispersion and interfacial adhesion and thus helps to achieve interconnected network structures in polymer matrices, resulting in increased mechanical strength and electrical conduction [33,34]. Long fibers, e.g., CNTs and CFs, can also make a continuous network in a polymer matrix, even with low loading, which precedes excellent stress distribution, heat dissipation, tensile strength, and resistance to structural deformation [32].

Conversely, the low aspect ratio of short nanoparticles results in gaps and irregular networks, which hinder the overall performance of the composite. Nevertheless, filler type, size, and aspect ratio are crucial to producing composite materials with tailored properties for specific applications [35]. Table 1 summarizes the role of different types of graphene with various aspect ratios reported in published research.

Table 1. Graphene hybrid polymer composites with diverse aspect ratios and different types of graphene types and major findings.

Composite Name	Graphene Type/Derivatives	Graphene Aspect Ratio (wt.%)	Findings	Ref.
Short carbon fiber-reinforced polyether sulfone composites by graphene oxide (GO) coating	GO	0.5	31.7% enhanced Young's modulus and 12.1% tensile strength	[36]
GO-coated short glass fiber-reinforced polyether sulfone composites	GO	0.5	25.4% enhanced Young's modulus and 10.2% tensile strength	[37]
Multi-graphene platelets (MGPs) and carbon nanotubes epoxy composites	MGPs	0.9	146.9% increase in thermal conductivity	[38]
Graphene and copper nanoparticles-based epoxy composites	Pristine graphene	40	6650% increase in thermal conductivity	[39]
Reduced graphene oxide sheets (RGO)/cellulose nanocomposite	RGO	10	71.8 S m^{-1} Electrical conductivity	[40]

Moreover, Tarani et al. [41] examined the influence of graphene nanoplatelets (GNPs) with an average platelet diameter of 25 µm (M25) aspect ratio on the mechanical properties of high-density polyethylene (HDPE) nanocomposites. Figure 3 shows that adding GNPs improves both the storage and loss modulus of the nanocomposite compared to pure HDPE, indicating an enhancement of the mechanical properties. This enhancement is more evident in nanocomposites with higher proportions of GNPs, suggesting that the aspect ratio of GNPs plays a significant role in the reinforcement of the HDPE nanocomposite. Tan (δ) is the ratio of the loss modulus to the storage modulus and is used to identify the glass transition temperature of the material. A sharp peak in neat HDPE shows its glass transition temperature. The broader and shifted peaks in the nanocomposites suggest that the GNPs increase the thermal stability and have a significant impact on the mechanical damping and thermal properties of HDPE, potentially extending its application range.



(a) (b)

(c)

Figure 3. The storage modulus (**a**), loss modulus (**b**), and tan (δ) values (**c**) of HDPE nanocomposites reinforced by graphene nanoplatelets (GNPs) with an average platelet diameter of 25 µm (M25), containing 2.5 wt.% and 5 wt.% GNPs. Adopted with permission from ref. [41].

2.2. Filler-Matrix Compatibility

Filler-matrix compatibility is a crucial factor in achieving robust and flawless composites. Graphene's 2D structure, nanometer size, and high surface area aid in the infusion capability to fabricate polymer composites with interlinked structures and expanded properties [42]. A well-maintained filler-matrix compatibility aids in realizing a uniform dispersion of the filler and an ordered structure with tailored properties [43–45]. Lu et al. [46] conducted molecular dynamics simulations to compare the elastic properties of graphene- and fullerene-reinforced polymer composites. They found that the size and morphology of graphene fillers strongly influence stiffness improvement, with smaller graphene flakes (2–4 nm radius) providing higher reinforcement than larger flakes or spherical fullerene fillers. This effect was attributed to the improved ordering and densification of polymer chains at the graphene-matrix interface. In another study, Li et al. [47] developed dopamine (DA)-modified graphene oxide (GO) and copper nanowires (Cu NWs) hybrid epoxy composites, and with the proportion of Cu-NWs @PDA:GO@PDA 7:3, a strong filler matrix compatibility was observed, which resulted in higher thermal conductivity of 0.36 W m^{-1} K^{-1} along with outstanding electrical insulation properties.

Conversely, due to strong van der Walls forces, high aspect ratios of graphene lead to agglomeration and non-uniform dispersion through the polymer matrix [48]. Graphene is surface-functionalized using different chemical groups or polymers to break the barrier, increasing the interfacial bonding and interaction between the filler and matrix. Romasanta et al. [49] successfully produced poly(dimethyl)siloxane nanocomposites filled with functionalized graphene sheets (FGSs) at a concentration of 2 wt.% FGS. A high dielectric constant was achieved, with high mechanical properties. Likewise, D. R. Son et al. [50] examined the compatibility of thermally reduced graphene (TRG) with multiblock copolyesters composed of poly (butylene terephthalate) (PBT) and poly(tetramethylene ether) glycol segments. The results showed that compatibility improved with an increasing content of the PBT segment in the polyester. This compatibility behavior was quantitatively analyzed using the electrical conductivity percolation threshold and Hansen solubility parameters. The results suggest that the compatibility of TRG with a polymer increases as the polymer's total solubility parameter (dT) value approaches 24.0 $(\mathrm{MPa})^{1/2}$. Several other studies [4,51] have also reported using hybrid fillers, such as graphene/carbon nanotubes or graphene/ceramic composites, to achieve synergistic improvements in the polymer composites' thermal, electrical, and mechanical properties. The compatibility between the filler and the polymer matrix components plays a crucial role in determining the overall performance of these hybrid systems.

2.3. Homogeneity of Dispersion

In the fabrication of excellent hybrid polymer composites, homogeneous dispersion is crucial for determining performance and properties. In graphene-based hybrid composites [52], graphene and other fillers are first evenly dispersed with each other and then uniformly distributed in a polymer matrix. Good dispersion results in a symmetrical structure and significantly boosts mechanical properties, i.e., fracture resistance, high tensile strength [53], and load-bearing facilities. Electrical and thermal conductivities are also improved, facilitating continuous charge transport, sensors, and heat management applications [47]. Tuichai et al. [54] prepared well-dispersed Ag-rGO/poly (vinylidene fluoride) (PVDF) composites using a seed-mediated growing technique and achieved a highly connected microstructure with a high dielectric constant. However, poor dispersion [55] results in agglomeration and uneven distribution within hybrid fillers and the polymer matrix, severely compromising the composite's capabilities. Such composite presents an irregular and non-uniform structure, leading to mechanical disruptions and ineffective electrical and thermal conductivities due to interrupted pathways [56]. Gong et al. [57] used the chemical vapor deposition (CVD) method to deposit graphene on the surface of nickel meshes, followed by creating graphene woven fabrics (GWFs) by removing the nickel template. Next, they stacked these graphene fabrics layer by layer to obtain an anisotropic graphene structure, which gave the polyimide (PI) composite an in-plane thermal conductivity of 3.73 W m^{-1} K^{-1} with 12 wt.% of graphene, as shown in (Figure 4a). On the other hand, Dai et al. [58] employed a roller equipment to extend and roll the porous polyurethane (PU)-graphene film, resulting in a large-scale monolith with a vertically aligned dual-assembled graphene framework (DAGF), as shown in (Figure 4b). After the graphitization process and epoxy resin infiltration, the resulting composite demonstrated an unprecedented thermal conductivity of 62.4 W m^{-1} K^{-1}.

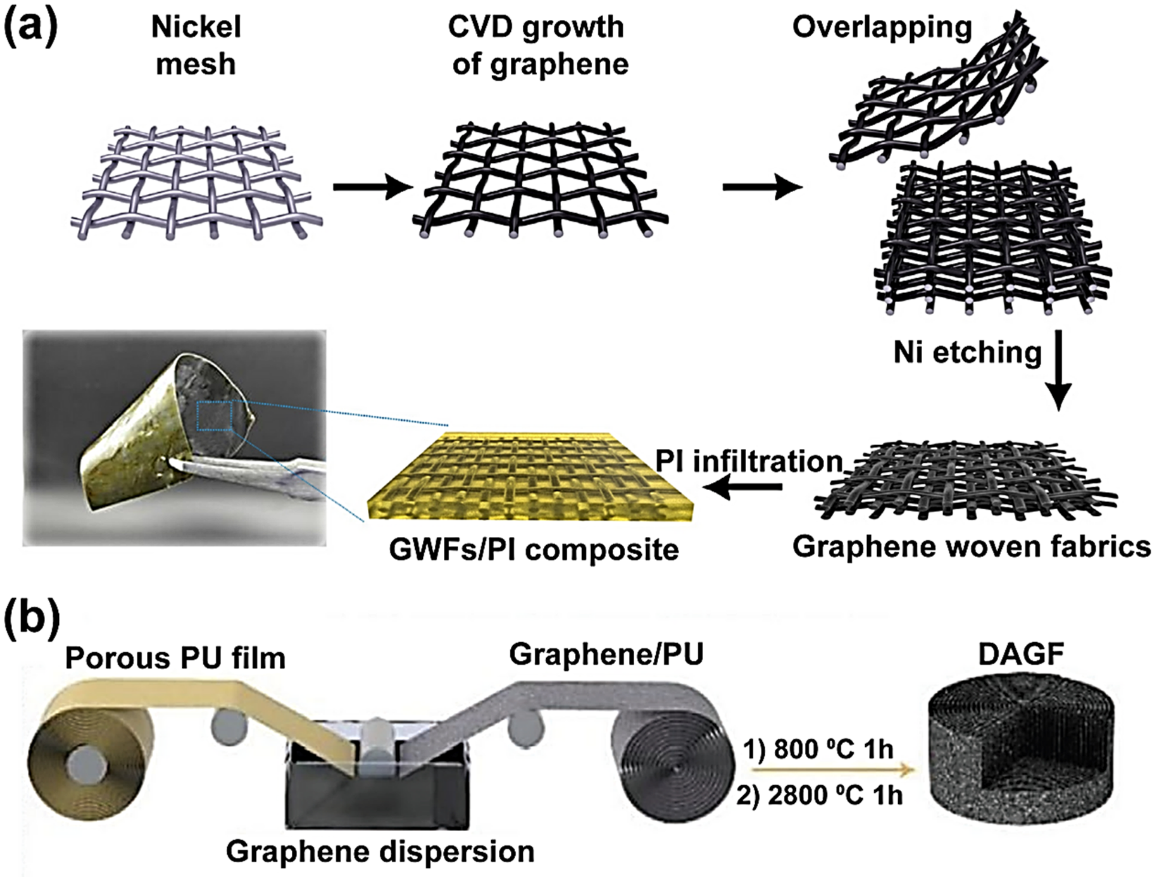


Figure 4. (**a**) Preparation process of GWFs/PI composite films and (**b**) preparation of dual-assembled graphene framework (DAGF) and composites by rolling process. Reprinted with permission from refs. [57,58].

2.4. Fiber Orientation

Fiber orientation highly influences the performance of polymer composites. In fiber-reinforced composites, unidirectional alignment offers high strength along the fibers but weaker properties in perpendicular directions. Bidirectional laminates offer a balance of properties, while graphene can improve different mechanical, thermal, and electrical characteristics depending on its orientation. Controlling the orientation of multiple fibers and graphene during processing allows the customization of composite properties for specific applications. Liu et al. [59] prepared anisotropic graphene aerogels by reducing GO with ascorbic acid at 70 °C for four hours, followed by freeze-drying (Figure 5a). Later, the examination showed that these anisotropic graphene aerogel materials offer high compressive strength in the axial direction and good compressibility in both directions, together with sensitive electrical conductivity upon deformation. Additionally, they feature ultra-low density, high porosity, fire resistance, and flexibility in organic liquids. After the absorption of liquids, they can be easily recycled by combustion, distillation, and pressing. Moreover, Zhang et al. [60] developed a vertically aligned graphene sheet membrane (VA-GSM) using a freezing approach (Figure 5b). VA-GSM demonstrated water evaporation rates of up to 6.25 kg m^{-2} h^{-1} under four times solar illumination, with a solar thermal conversion efficiency of up to 94.2%. This performance makes it possible to efficiently produce clean water from sources such as seawater and wastewater, as well as concentrated acid and alkaline solutions.

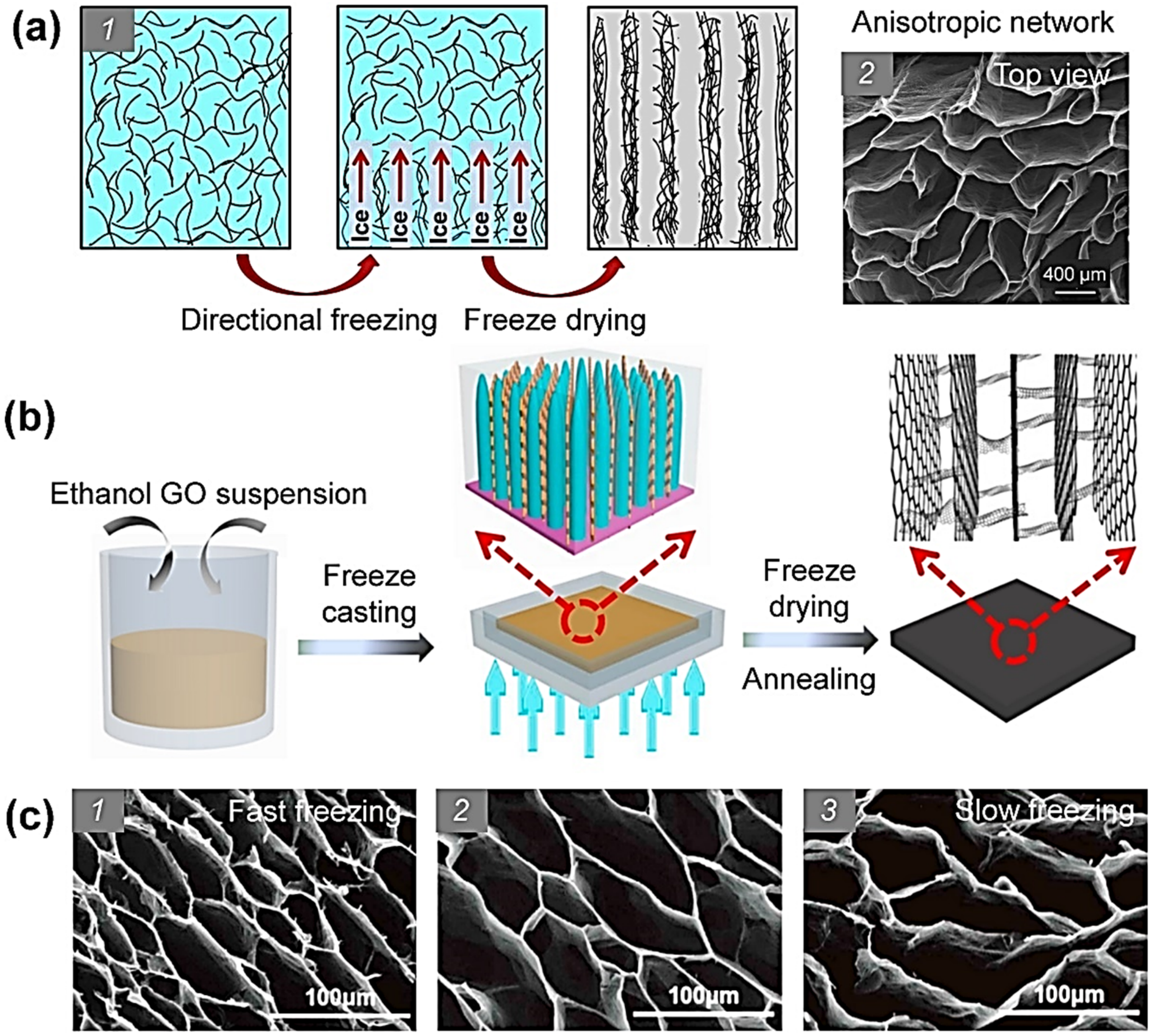


Figure 5. (**a**) Schematic of the microscopic sequences of directional freezing and top view of the final structure [59]. (**b**) Schematic of directional freezing of graphene oxide (GO) suspension and aligned graphene (GA) structure [60]. (**c**) SEM images showing vertically aligned graphene networks, fabricated through freeze-drying at different freezing rates, as indicated, and subsequent freeze-drying, captured from a top view [61]. Reproduced with permissions from refs. [59–61].

In another study, thermally conductive polymer composites were obtained from graphene oxide hydrogels via directional freezing and graphitization at 2800 °C. It was observed that these aerogels exhibit excellent thermal and electrical conduction properties due to the highly aligned network of graphitized graphene sheets. An epoxy composite containing 0.75% high-quality graphene exhibits a vertical thermal conductivity of 6.57 W m^{-1} K^{-1}, along with improved mechanical strength and compressive deformation. Figure 5c shows SEM images of vertically aligned graphene networks at different freezing rates, captured from a top view [61].

An et al. [62] fabricated vertically aligned graphene hybrid foams (GHFs) via the hydrothermal reduction process of GNPs, resulting in high-density foams. The integration of GNPs improves the thermal conduction network's density and prevents excessive foam shrinkage during manufacturing. By graphitization at 2800 °C, the oxygen-containing groups are eliminated, and the defects are healed, obtaining high-quality graphene foams (Figure 6a). It was observed that GHF/epoxy composites exhibit an extraordinary through-plane thermal conductivity of 35.5 W m^{-1} K^{-1} at a graphene content of 19 vol.%, making them compatible for thermal management applications (Figure 6b).

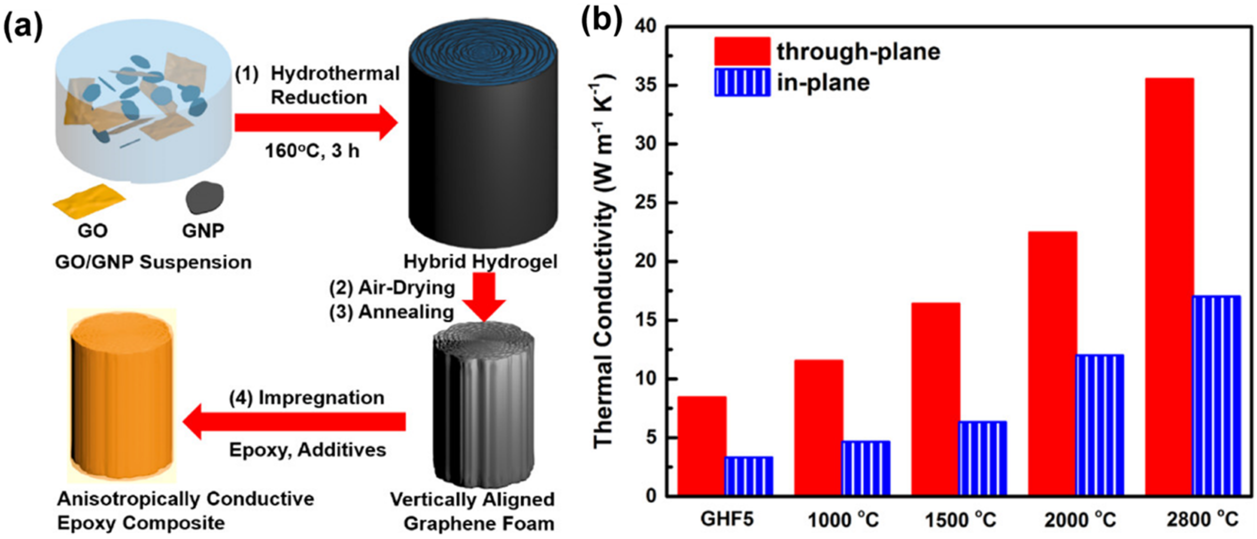


Figure 6. (**a**) Representative diagram of a conductive epoxy composite production enriched with vertically oriented RGO/GNP hybrid foam. (**b**) Thermal conductivity variation in the in-plane direction (red column) and in the transverse direction (blue column) of the epoxy composite incorporating the RGO/GNP hybrid foam aligned vertically at different temperatures. Adopted with permission from ref. [62].

2.5. Synergistic Effects

The presence of graphene in hybrid polymer composites results in synergistically enhanced network structures, making them ideal for structural applications [63]. These composites exhibit superior mechanical properties, such as increased tensile strength, modulus, and toughness. Their high electrical and thermal conductivity makes them suitable for flexible electronics, thermal management, and electromagnetic interference shielding. Xiao et al. [64] developed PVDF composites with various combinations of CNTs and GNPs (Figure 7a). The addition of 2 wt.% CNTs to PVDF/GNP composites generated a three-dimensional hybrid network with improved thermal and electrical conductivity, and the incorporation of 2 wt.% GNPs into the PVDF/CNT composites did not change the percolated structure. Both CNTs and GNPs promoted the crystallization of PVDF without significantly altering its crystallinity. Ternary composites demonstrated increased electrical conductivity due to a three-dimensional conductive path. Particularly, PVDF/CNT/GNP composites showed higher thermal conductivity and synergistic efficiency compared to PVDF/GNP/CNT composites. In addition, Liang et al. [65] developed an epoxy nanocomposite with four times the thermal conductivity of pure epoxy, using reduced graphene oxide (RGO)/functionalized single-walled carbon nanotube (f-SWCNT) filler loading at 3.65 vol%. This result was achieved due to a rigid and well-aligned 3D skeleton of RGO walls connected by f-SWCNTs (Figure 7b), which not only improved the alignment of the SWCNTs but also allowed a significant reduction in the amount of SWCNT used, helping to reduce costs. The 3D networks improved both the dimensional and thermal stability of the nanocomposite without compromising its electrical insulation properties. Shtein et al. [66] explored the effective integration of GNPs into a polymer matrix to improve thermal conductivity (TC) significantly. Through a scalable and straightforward dispersion process, the resulting composite demonstrated an ultrahigh TC of 12.4 W m^{-1} K^{-1}, compared to 0.2 W m^{-1} K^{-1} for the pure polymer, achieved by applying high compressive forces to close the spaces between adjacent graphene nanoplatelets. It was found that the addition of boron nitride nanoparticles synergistically improved TC while reducing electrical conductivity, preventing short circuits (Figure 7c). This advancement facilitates progress toward large-scale production of thermally conductive composites based on GNPs. Xue et al. [67] developed novel composite phase change materials (PCMs) by incorporating melamine foam with GO and GNPs (Figure 7d). The resulting aerogels exhibited high structural stability and high load-bearing capacity. The composite PCMs exhibited high-temperature shape stability and high phase change enthalpy retention capacity, as well as significantly improved thermal and electrical conductivities. These results indicate considerable potential for applications in energy efficiency and thermal protection of microelectronic devices.

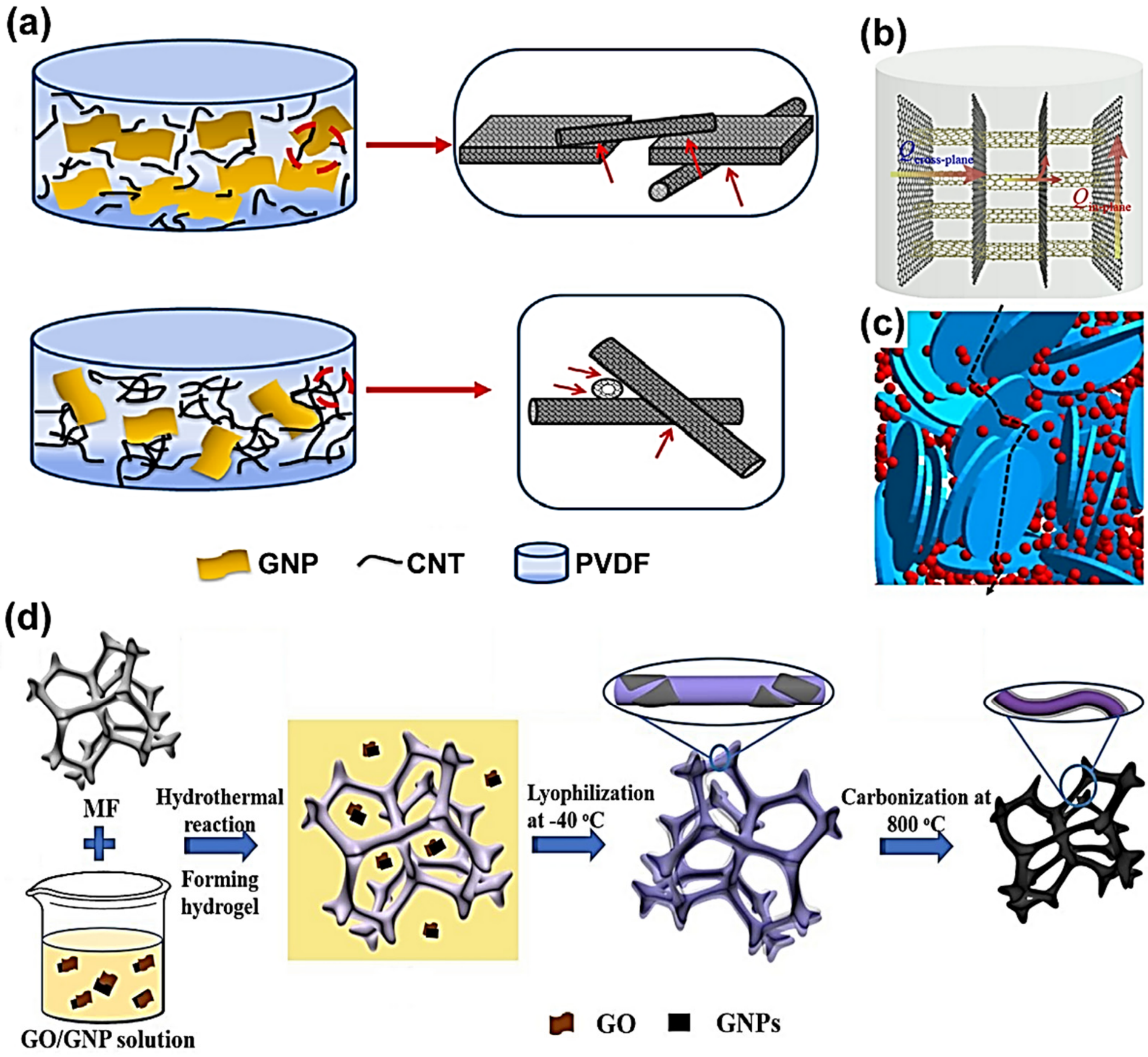


Figure 7. (**a**,**b**) Diagrams illustrating the synergies between GNPs and CNTs in thermal conduction [64,65]. (**c**) Schematics illustrating the synergies between GNPs (polymeric nanoalloys, represented by the blue slices) and BN (boron nitrate, represented by the red spheres) [66]. (**d**) Schematic illustration of the preparation process of both the graphene/MF foam (mixed fiber graphene foam) and the derived graphene/carbon foam [67]. Reprinted with permission from refs. [64–67].

2.6. Processing Techniques

Composite processing methods are pillars to achieve a uniformly distributed microstructure with expanded properties. Graphene-based polymer composites can be prepared using several methods, each with specific advantages that influence the final properties and applications of the composites. The most common techniques include solution mixing, melt mixing, and in situ polymerization. In solution mixing, graphene is dispersed in a solvent, then the polymer is added, and finally, the solvent is removed to ensure good dispersion of graphene in the polymer matrix. Melt blending mixes graphene and polymer at high temperatures and shear, affecting the size and morphology of graphene, and is suitable for large-scale production. In situ polymerization involves growing the polymer around graphene, creating strong interfacial interactions but requiring more complex processing conditions. The choice of preparation method affects graphene dispersion, interfacial interactions, and mechanical, electrical, and thermal properties of the composite. Effective dispersion of graphene with strong polymer interactions usually leads to improved mechanical, electrical, and thermal performance of composite materials. These graphene-polymer composites are used in industries such as electronics, energy storage, and personal protective equipment [68–71]. Moreover, the fabrication techniques involving compression mold, 3D printing, and filament winding affect the filler placement inside the matrix. Fabrication of graphene-based hybrid polymer composites with uniform distribution of graphene highly aids in fully harnessing graphene's unique properties and synergistic relation with other fillers [16,72]. However, the tendency of graphene layers to attach precedes clustering issues. Thus, efficient processing strategies, e.g., sonication, high shear mixing, and extrusion methods, aid homogeneous graphene dispersion and increase its reinforcing effect on the composite [73,74]. Moreover, curing conditions, e.g., time or temperature, significantly impact the interfacial bonding and load transfer ability between filler and matrix, improving the composite's mechanical properties and durability. Additionally, specific processing methods, e.g., 3D printing, electrospinning, spray coating, or shear-induced alignment, provide better control over the graphene orientation, enabling the fabrication of complex geometries with tailored anisotropic properties. The choice of preparation process dramatically impacts the cost-effectiveness and scalability of composites [75,76]. Several studies have compared the effectiveness of various processing methods by characterizing filler dispersion and composite performance. For example, Kim et al. [34] examined thermoplastic polyurethane (TPU)-based composites containing thermally reduced graphene oxide (TRG) obtained via solvent dispersion, in situ polymerization, and melt mixing (Figure 8a). The results indicate that the structure and performance of composites are strongly influenced by the preparation methods, suggesting the possibility of customization by optimizing manufacturing processes. Furthermore, it was found that solvent-treated composites exhibit higher stiffness and a lower electrical percolation threshold than those obtained via melt mixing, consistent with better dispersion of filler particles. Cao et al. [77] developed 3D graphene networks using Ni foam as a template in the ethanol-CVD process. The 3D graphene networks presented excellent templates for the construction of graphene-metal oxide composites (Figure 8b). The NiO-graphene composite showed a high specific capacitance of ≈816 F g^{-1} and stable cycling efficiency, maintaining this specific capacitance even after 2000 cycles at a scan rate of 5 mV s^{-1}. Pham et al. [78] produced poly(methyl methacrylate)/reduced graphene oxide (PMMA/RGO) composites with an electrical conductivity of 64 S m^{-1} at 2.7 vol% by using self-assembly of positively charged PMMA particles and negatively charged GO sheets by reduction with hydrazine (Figure 8c). Moreover, thermomechanical properties were improved, with a 30% increase in storage modulus and a 15 °C increase in glass transition temperature with only 0.5 wt.% RGO.

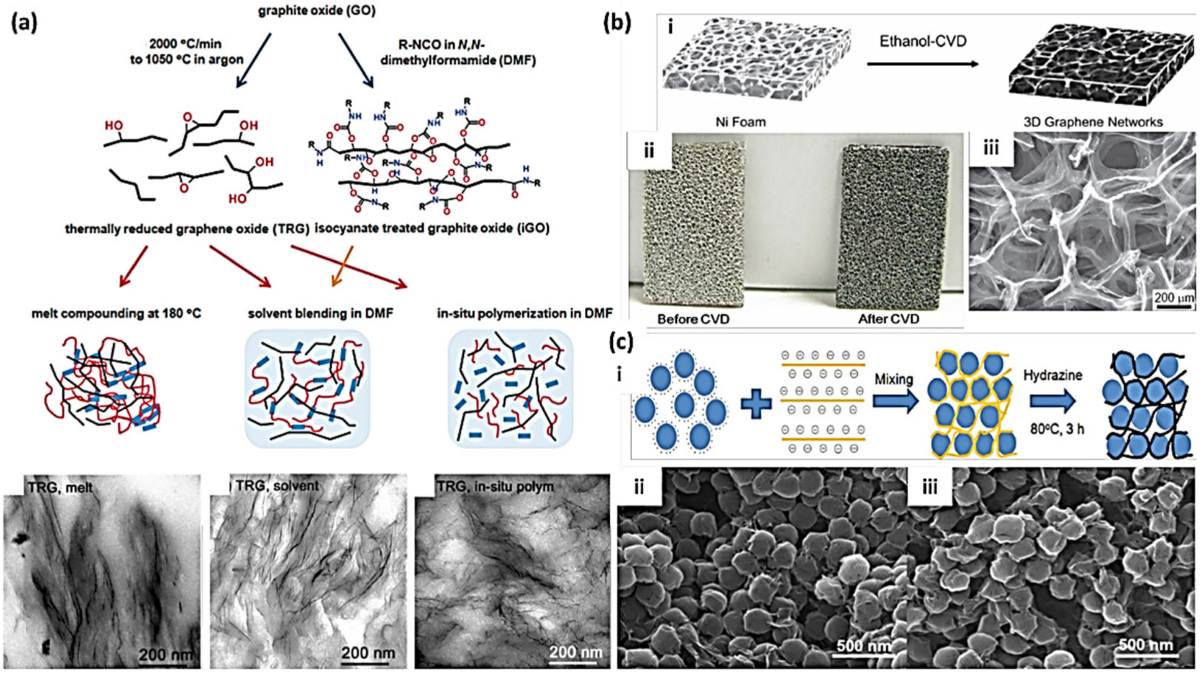


Figure 8. Different fabrication methods for making graphene-polymer composites. (**a**) Representations of TRG/TPU composite formulation routes and TEM images of TRG in TPU and 3 wt.% TRG/PU composites fabricating processes, revealing the improved dispersion of TRG by solution blending and in situ polymerization as compared to melt mixing [34]. (**b**) Porous graphene frameworks manufactured by ethanol-CVD method: (**i**) represents the synthesis diagram of 3D graphene networks on Ni foam by ethanol-CVD, (**ii**) represents Ni foam before and after the growing of graphene, and (**iii**) depicts 3D graphene assemblies after elimination of Ni foam [77]. (**c**) Three-dimensional RGO interaction by a self-assembly method [78]: (**i**) the self-assembly of PMMA latex and graphene oxide, supported by hydrazine reduction of graphene oxide, and SEM images of RGO-PMMA composites packed with various filler loadings; (**ii**) 2 wt.%; and (**iii**) 4 wt.% [14]. Adopted with permission from refs. [14,34,77,78].

2.7. Post-Treatment and Functionalization

Post-treatment processes, such as surface treatments and protective coatings of composites, significantly increase their resistance to humidity and corrosion. In particular, the graphene filler and polymer matrix interaction is highly sensitive to environmental conditions, and surface treatments can significantly improve composite resilience [79–81].

On the other hand, functionalization can be tailored to exploit the unique properties of graphene. By modifying the graphene surface with specific functional groups or incorporating other nanoparticles, functionalization can enhance electrical and thermal conductivity or even provide additional functionalities, such as sensing capabilities. This level of customization is particularly advantageous in applications where precise control over material properties is required, such as in advanced electronics or sensors [82,83]. Functionalization opens new possibilities for graphene-based composites, enabling them to excel in areas where traditional materials fall short. In essence, post-treatment and functionalization strategies in graphene-based hybrid-filler polymer composites constitute sophisticated techniques that unlock the full potential of these materials. They ensure the composite's robustness and longevity and empower it with diverse functionalities [84,85]. As the field of materials science continues to evolve, the synergy between graphene and tailored post-treatment and functionalization processes offers exciting prospects for developing innovative and high-performance materials that can revolutionize various industries [86].

3. Applications

The authors discuss the results and how they can be interpreted from the perspective of previous studies and the working hypotheses. The findings and their implications are discussed in the broadest context possible. Future research directions are also be highlighted.

3.1. Aerospace Evolution: Lightweight Structural Components for Enhanced Performance

Graphene-based hybrid polymer composites are an excellent candidate for aerospace applications due to their exceptional mechanical properties, e.g., strength, stiffness, and lightweight, which enable the creation of robust structural components. The combination of lightweight, higher Young's modulus and tensile strength improves fuel efficiency, payload capacity, and overall aircraft efficiency [87]. Moreover, these composites can resist extreme temperature conditions, mechanical stress, and corrosion, making them suitable for aircraft frames, wings, and engine components. Space shuttles and high-velocity aircraft need a thermal management system to mitigate overheating and structural issues. Graphene composites serve in thermal protection systems, heat shields, and thermal barriers due to their excellent thermal conductivity [88,89]. Figure 9 shows potential applications of graphene-based polymer composites in different aspects of aerospace industries.

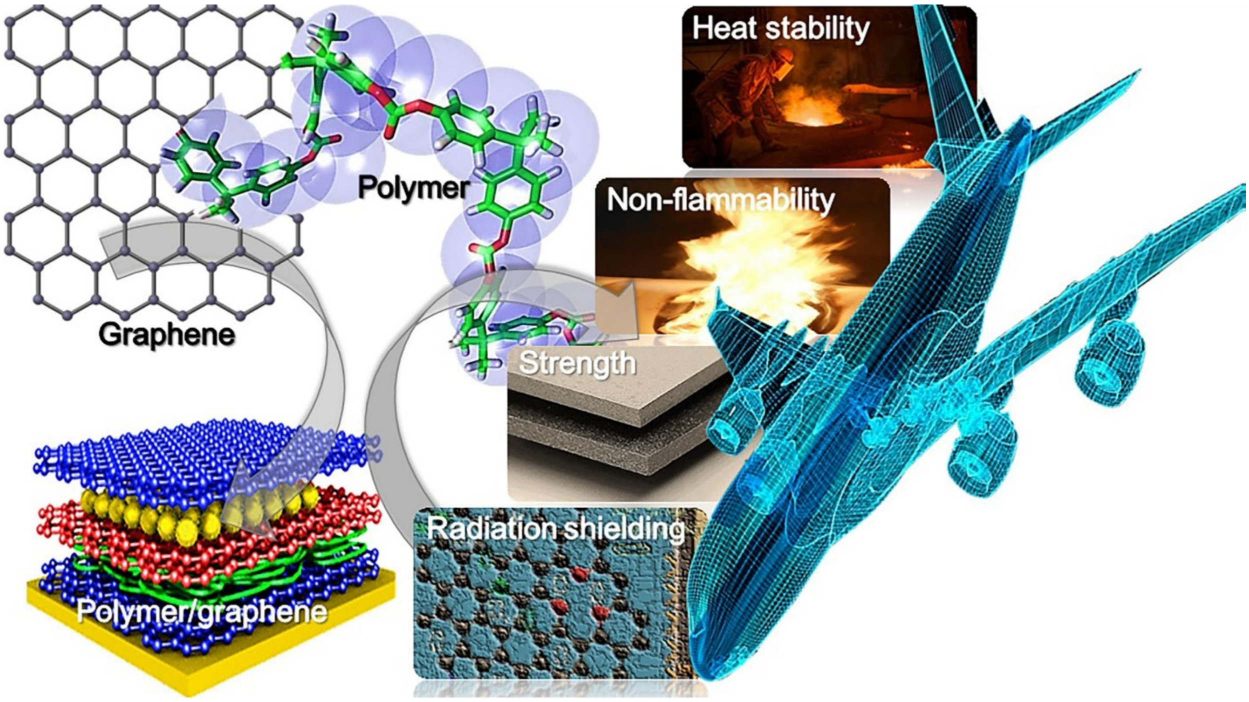


Figure 9. Multiple roles of graphene hybrid polymer composites in aerospace applications. Reprinted with permission from ref. [90].

Furthermore, installing graphene composite-based sensors in aircraft provides real-time surveillance of structural deformation. Lightning strikes during flights cause severe accidents due to the absence of electrically conductive materials in manufactured parts, while graphene-based hybrid composites have excellent thermal and electrical conductivity and are effective in resolving lightning strike problems [91]. Furthermore, space exploration demands robust materials to tolerate extensive radiation and vacuum conditions [92]. Graphene-based hybrid composites offer impressive radiation shielding, mechanical strength, and thermal stability, enabling the manufacture of spacecraft components that are oriented to unforgiving space environments. However, utilizing the full potential of these composites encounters issues such as cost-effectiveness, regulatory compliance, scalability, and standardized practices [93,94]. Öztürkmen et al. [95] prepared GNPs/hexagonal boron nitride (h-BN)/epoxy nanocomposites by integration of GNPs and h-BN into an aerospace-grade epoxy resin via three-roll milling to achieve significantly improved nanocomposite properties. Five mixing cycles with a roller gap of 50 µm led to promising results. It was observed that a hybrid nanocomposite (0.5 wt% GNP and 0.5 wt% h-BN) showed a 69% increase in flexural strength, a 7% increase in thermal conductivity, and a notable improvement of 10^8-fold increased electrical conductivity. Thus, these fillers can be effectively designed for aerospace applications, such as lightweight avionics frames, which require specific thermal and electrical properties. There are many references reporting on graphene hybrid polymer composites with desired properties for the aerospace sector [88,96,97].

3.2. Advanced Electronics: High-Performance Conductive Materials and Flexible Circuits

Given their significant electrical and thermal conductivity [62], lightweight graphene-based hybrid polymer composites [98] present high-performance conductive coatings, optimal fuel efficiency, efficient heat dissipation [99], effective signal transmission, and electromagnetic shielding in electronics. These composites are perfect for flexible circuits and wearable devices due to their flexibility and lightweight nature [100]. Moreover, the synergy of their electrical and thermal properties with mechanical ability has revolutionized smart textiles, sensors [101], and integrated electronic systems [102,103]. In a study [104], highly flexible and electrically conductive multifunctional textile composites were produced using GO films and Ag nanoparticles on a polyethylene terephthalate (PET) fabric. These composites were employed to create flexible organic transistors, where the composite fabric functioned as both a flexible substrate and a conductive gate electrode. Thermal treatment improved the electrical performance of the textile transistor, with a mobility of $7.2\ \mathrm{cm}^2\ \mathrm{V}^{-1}\ \mathrm{s}^{-1}$, an on/off current ratio of 4×10^5, and a threshold voltage of -1.1 V. Furthermore, the textile transistors have demonstrated performance stability even under extensive bending conditions, withstanding a bending radius of up to 3 mm and enduring repeated testing over 1000 cycles. These results indicate that the fabrication methods of graphene/AgNP textile composites for textile transistors could be successfully applied in the development of large-scale flexible electronic clothing.

In their study, Li et al. [105] reported an orientated assembly of giant graphene oxide (GGO) sheets via 3D printing and achieved flexible patterns with high surface area and enhanced electrical conductivity of up to $4.51\ (\pm 0.18) \times 10^4\ \mathrm{S\ m}^{-1}$. Moreover, these orientated patterns were tested for the electrically driven soft actuators, and it offered controlled deformation at a lower voltage. Figure 10 shows the detailed analysis and fabrication process of graphene hybrid composites in flexible and printable electronics by manufacturing a soft actuator with printed RGO electrodes. Figure 10a illustrates the electrical stimulation of the bilayer RGO/polydimethylsiloxane (PDMS) actuator. Figure 10b shows the maximum bending angle as a function of driving voltage. Figure 10c depicts digital images of reversible shape change of the actuator at different bending angles, and Figure 10d depicts a hand-shaped actuator enabling independent control of the fingers by various gestures.

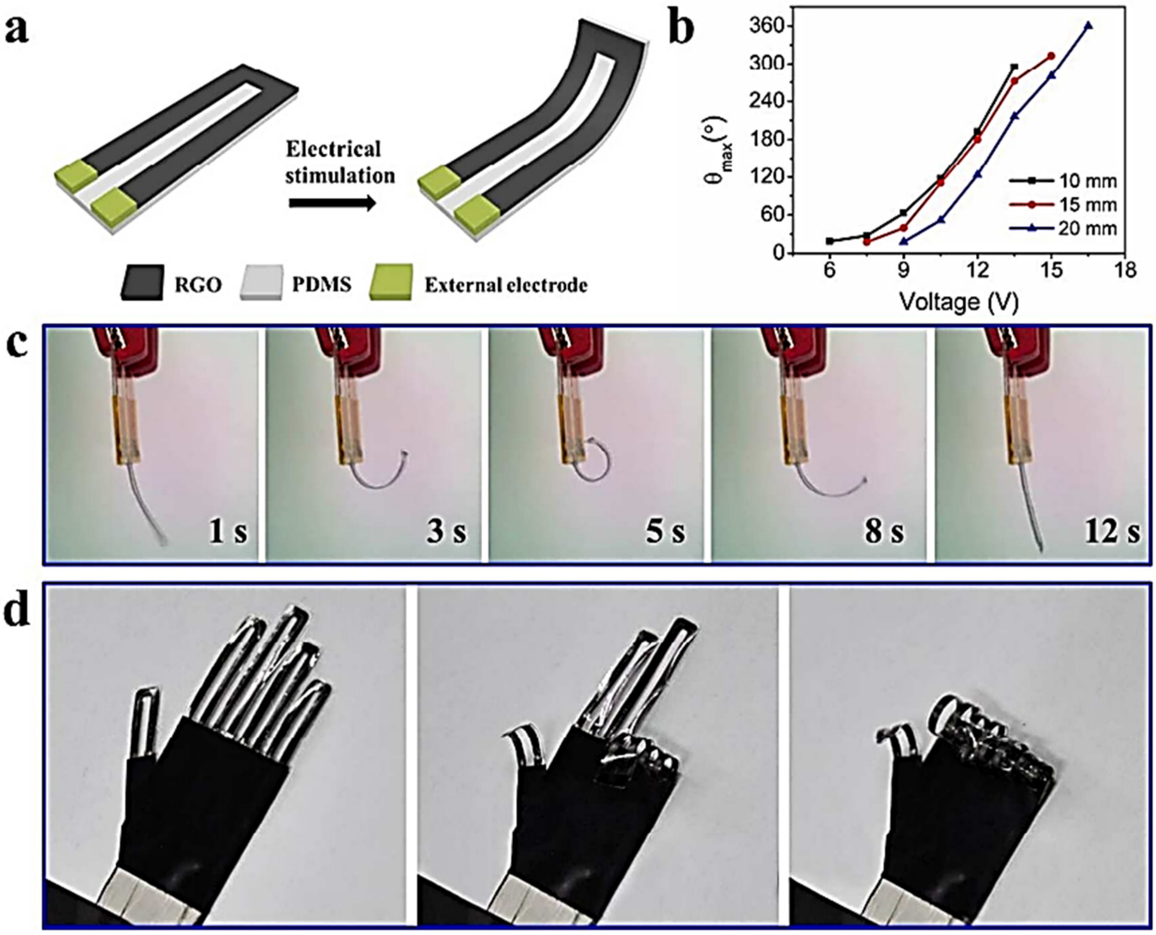


Figure 10. Fabrication of soft actuator with printed RGO electrodes. (**a**) RGO/PDMS bilayer actuator under electrical stimulation. (**b**) The maximum bending angle of the actuators as a function of the driving voltage. (**c**) Digital images of the actuator at various bending angles, with reversible shape change of the actuator. (**d**) The final look of a hand-shaped actuator with independent fingers controlled by various gestures. Reprinted with permission from ref. [105].

3.3. Energy Storage Revolution: Efficient Batteries and Supercapacitor

Graphene-based hybrid polymer composites have revolutionized energy storage technologies [106]. Their remarkable electrical conductivity and large surface area lead to elevated energy storage in batteries and supercapacitors by facilitating faster charging and discharging rates, extending battery lifetime and improving energy density. This progression addresses the energy demands of portable electronics, renewable energy storage systems, and electric vehicles, resulting in green energy solutions [107,108]. Additionally, these composites' electrical and thermal properties offer enhanced solar cells and thermoelectric generators by improving electron mobility, light absorption, and thermal conversion, resulting in efficient waste-heat recovery [100]. Lithium-ion batteries have become the main source of energy for portable electronic devices. However, when used at high charge/discharge rates, the rate of heat generation may exceed the rate of heat dissipation, making the battery inefficient or even posing a fire risk. To keep the battery at an optimal temperature, an effective thermal management system is required. To address this, Khan et al. [109] presented the fabrication and characterization of polyacrylonitrile (PAN) and polymethyl methacrylate (PMMA) nanofiber separators embedded with graphene nanoflakes. The physical properties of nanocomposite separators were significantly improved with increasing graphene concentration. For example, the contact angle with water went from 120° to 145°, the ionic conductivity from 3.31×10^{-4} to 5.52×10^{-4} S m^{-1}, the dielectric constant from 3.5 to 8.5 W m^{-1} K^{-1}, and the thermal conductivity from 1.0 to

5.0 W m^{-1} K^{-1} when the graphene concentration was increased from 0% to 8% in PMMA. Similar trends were also observed in PAN fibers, suggesting that such nanocomposite separators could provide a robust option for producing long-lasting lithium-ion batteries.

He et al. [110] fabricated a very thin (0.5 mm width) graphene-based paper-like electrode for a micro-supercapacitor with a current density ranging from 0.5 to 5 A cm^{-3} and exhibiting a high volumetric capacitance of ~3.6 F·cm^{-3}, with a highly specific capacitance retention of up to 94%, even after 20,000 cycles. Figure 11 shows the detailed preparation process of the composite and its integration into a micro-supercapacitor.

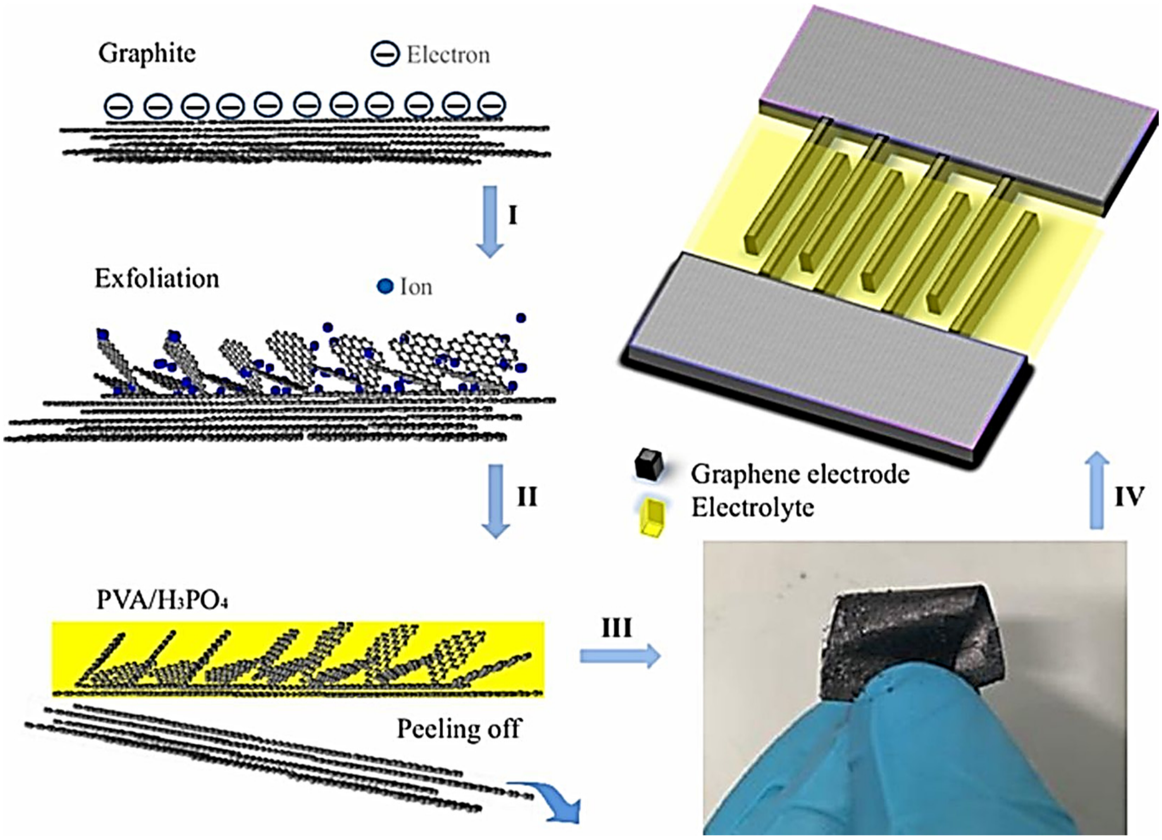


Figure 11. Preparation process of a micro-supercapacitor. (**I**) Electrochemical exfoliation of graphite foil. (**II**) Coating the graphene/graphite foil with gel electrolyte (PVA/H3PO4). (**III**) Peeling off flexible graphene paper-like electrode. (**IV**) Building of interdigitated micro-supercapacitor devices using the paper-like electrode. Reprinted with permission from ref. [110].

3.4. Transportation Innovation: Lightweight Automotive Components for Enhanced Efficiency

Graphene-based hybrid polymer composites are fundamental in developing robust and fuel-efficient vehicles due to their unique mechanical and lightweight properties. The high electrical conductivity of graphene-based composites dramatically improves the charging efficiency of lithium-ion batteries, resulting in optimum battery lifetimes in electric vehicles [111]. High mechanical strength [112] and higher flexibility facilitate tailored, impact-resistant, and safely enhanced vehicle modules [15]. Furthermore, outstanding thermal conductivity influences electric vehicles' energy efficiency and component durability by heat dissipation through batteries, electric motors, and power electronics [113–115]. Kenaf-based composites are known for their high strength-to-weight ratio, but the weakness of the fiber-matrix interface limits their automotive application. To address this problem, a new multiphase composite fabrication technique has been developed, using GNPs, kenaf fibers, and an epoxy matrix. The composites were produced via vacuum infusion molding, with uniform dispersion of the GNPs using a bath sonicator. Composites containing 0.2 wt.% GNP showed the greatest improvements in mechanical properties, with a 30.5% increase in tensile strength, a 61.5% increase in tensile modulus, a 17.6% increase in flexural strength, a 22.7% increase in flexural modulus, a 35.1% increase in interlaminar shear strength, and a 17.1% increase in fracture toughness. Furthermore, the resistance to water absorption was improved by 7%. These improvements were attributed to the uniform distribution of GNPs and better bonding to the fiber surface. The composites developed in this way have the potential to be used in the interior parts of automotive vehicles, such as dashboards, interior panels, and luggage compartments [116]. Figure 12 presents various graphene composites examined for load-bearing and structural automotive applications, such as seats for Daimler Chrysler sports cars, a diagonal support beam for Porsche, and bumper structures for BMW.

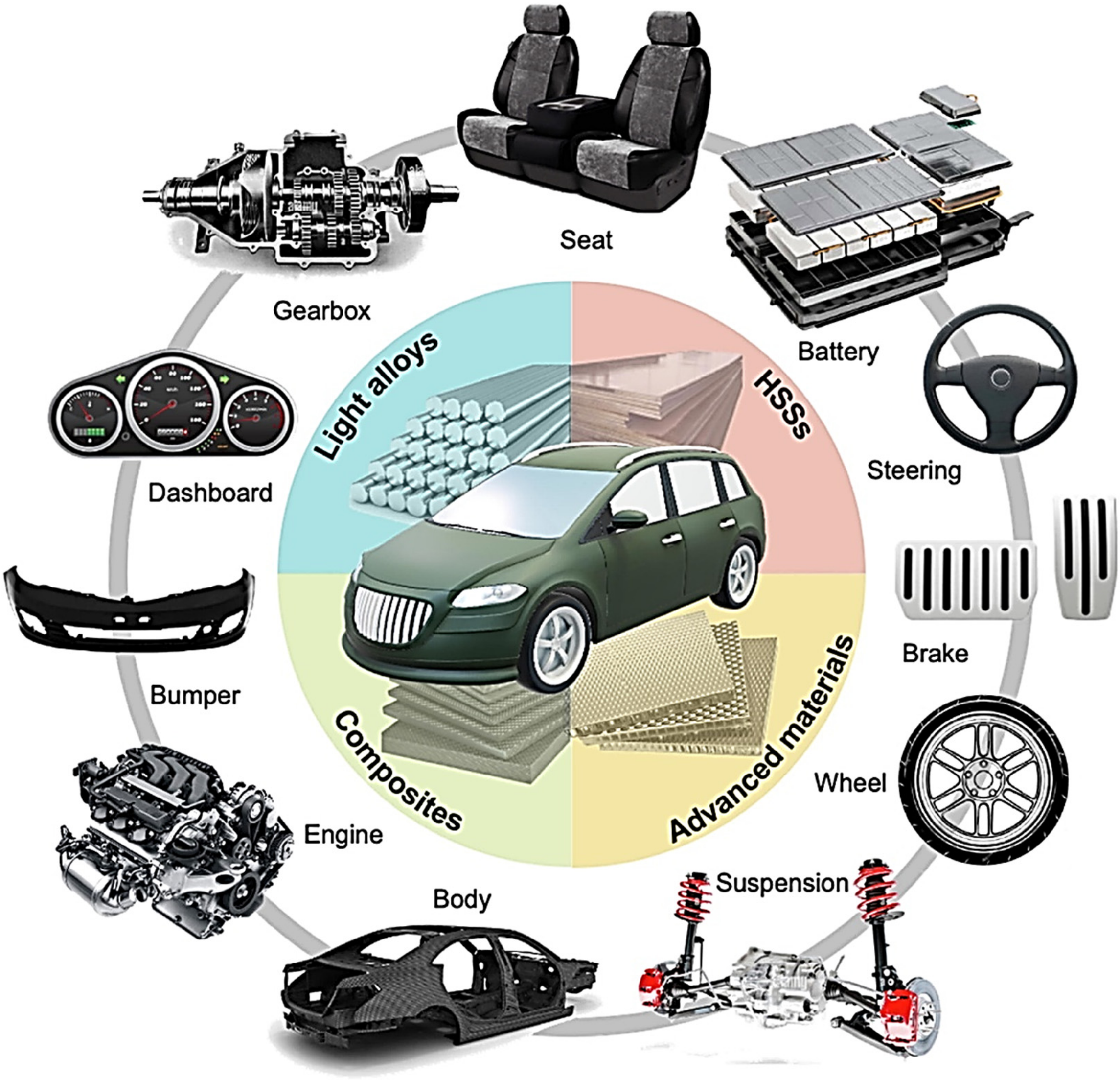


Figure 12. Role of graphene and advanced hybrid polymer composites in load-bearing automobile structure preparation [117]. Reprinted with permission from ref. [118].

4. Conclusions

In conclusion, graphene hybrid polymer composites are at the forefront of material innovation, offering a unique combination of mechanical strength, electrical conductivity, and thermal stability. Our review paper delves into the critical factors influencing the performance and industrial applications of these composites. The choice of graphene type, whether graphene oxide, reduced graphene oxide, or pristine graphene, sets the stage for the composite's properties, with aspect ratio playing a key role in determining strength and flexibility. The compatibility between the filler and the polymer matrix is paramount, ensuring a robust interface that allows for efficient stress transfer and prevents delamination. Achieving a homogeneous dispersion of graphene within the polymer matrix is a technical challenge but crucial for consistent composite properties. Fiber orientation is another significant aspect, affecting the anisotropy of mechanical and electrical properties, enabling tailored solutions for specific applications. Synergistic effects, resulting from the combination of graphene with other fillers, can lead to enhanced performance that exceeds the sum of its parts. The processing techniques used, such as extrusion, molding, or additive manufacturing, affect the scalability and quality of the final product, while post-treatment and functionalization offer opportunities to further tune the composites' properties, enhancing conductivity or chemical resistance. These factors collectively influence the effectiveness of graphene hybrid polymer composites in various industrial applications. In aerospace, their lightweight strength offers fuel-saving benefits; in advanced electronics, their conductivity and flexibility are invaluable; for energy storage, their high surface area and conductive properties improve battery and supercapacitor performance; and in transportation, their resilience and durability contribute to safer and more efficient vehicles. Understanding and optimizing these factors is key to harnessing the full potential of graphene hybrid polymer composites across diverse industries.

5. Future Perspectives

Developing efficient and scalable manufacturing methodologies for high-quality graphene materials is critical for multiple industries, including electronic devices, thermal management, and transparent conductive electrodes. Chemical vapor deposition (CVD) on metal sheets offers a promising route to infinite-length graphene sheets suitable for roll-to-roll processing, but advances in quality and transfer processes are needed. Controlling graphene layer thickness remains challenging, especially for double- and triple-layer graphene. Understanding the underlying mechanisms is essential to improve economic production through chemical methods, such as graphite oxidation and subsequent reduction. Future research should focus on optimizing graphene surface functionalization, investigating new structures, and developing graphene hybrid polymer composites. These efforts aim to combine the unique properties of graphene with the versatility of polymers, facilitating their application across a wide range of applications. However, significant research gaps persist, particularly in optimizing fabrication techniques, understanding interfacial interactions, and tailoring the properties of graphene-polymer composites for specific applications. Addressing these gaps is thus crucial for driving innovation in multifunctional materials.

Author Contributions: Writing—original draft preparation, Z.A.; writing—review and editing, S.Y.; visualization, J.Y.; analysis, and revision, A.D. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data are contained within the article.

Conflicts of Interest: The authors declare no conflicts of interest.

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