

Review Article

A Review on the Contemporary Development of Composite Materials Comprising Graphene/Graphene Derivatives

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As two-dimensional materials with a high specific surface area, strength, and electrical and thermal conductivity, graphene (Gr) and its derivatives have great application prospects in various fields such as structural reinforcement, energy storage, optics, and electrical and thermal conductivity. However, the current preparation method of Gr is complicated and expensive. Its preparation cost can be reduced using Gr composites that include inorganic materials or other polymers. Composite materials comprising Gr and inorganic materials are generally prepared using sinter molding, chemical reduction, or chemical deposition. Gr should be able to mitigate several of the defects resulting from inorganic materials, including their poor toughness, small specific capacity, and low photoelectric conversion rate. The mitigation of these defects should result in the expansion of the applications of Gr composites for use in capacitors, catalysts, and sensors, among other devices. Although solution blending or in situ polymerization produces a relatively good mixing effect, in general, melt blending is used in the large-scale processing of Gr composites with polymer materials. Gr can be used to partially mitigate several weaknesses of polymer materials, including their low strength, poor electrical and thermal conductivities, and poor isolation performance, and expand the applications of Gr composites for use in electrical and thermal conductive materials and functional membrane materials, among other applications. The goal of the present paper is to introduce the structural properties of Gr and a surface modification method for Gr. In addition, the preparation methods and application fields of different Gr composite materials are reviewed. Finally, the development prospects and key issues are discussed.

1. Introduction

In the 1950s, Wallace [1] proposed the concept of graphene (Gr) and theoretically explored the electronic structure of Gr. In 2004, the physicists Novoselov et al. [2] successfully separated Gr from graphite at the University of Manchester. Because of its unique two-dimensional planar structure and other advantages, including its high strength and electrical and thermal conductivities [3–7], Gr has attracted the interest of both researchers and commercial enterprises. However, the current preparation method for Gr is relatively complicated and expensive. Additionally, the difficulty of the molding process alone limits its applications. For use in practical applications, Gr and its derivatives are often composited with inorganic materials and polymers to

prepare composite materials. The prepared composite materials have excellent properties, are of low cost, and have great application prospects for use in many fields, including structural reinforcement, energy storage, optics, conductive heat conduction, and biomaterials [8–12].

In recent years, there have been many research literatures on Gr composites, but the quantity of review literature is limited. For researchers who want to study Gr composites for the first time, it is difficult to have a comprehensive understanding. Some of literature on Gr composites describes the research status based on different application fields, and the content covered limited types of materials. It is difficult for researchers to see the preparation methods and possible application fields of a certain material. In this review, the research progress of Gr composites is

summarized from the perspectives of Gr/inorganic and Gr/polymer composites. The preparation methods and properties of different materials are described based on the matrix types of composite materials. It may have some guiding significance for the researcher to choose the matrix material and preparation method when preparing Gr composite materials with certain properties.

This review, which encompasses the data from 144 papers, discusses the structural properties and applied surface functionalization methods of Gr. The methods used for the preparation of Gr/inorganics and Gr/polymer composite materials and their possible applications are summarized herein. In addition, this paper discusses the developmental prospects of Gr composites and their key technical issues.

2. Graphene

2.1. Introduction to Graphene. Gr is a two-dimensional crystal material composed of layers of carbon atoms [13]. Its unique structure provides its remarkable performance for various applications. Its large specific surface area, excellent electrical performance, very small resistivity, and very high thermal conductivity are properties that stem from its unique structure [14–16]. At present, the primary synthetic methods used for Gr synthesis include chemical gas deposition (CVD), epitaxial growth, mechanical exfoliation, and oxidation-reduction; the oxidation-reduction method has been used in the widest range of applications [17–21].

2.2. Functionalization of Graphene. In the preparation of composite materials, the functionalization of Gr is necessary because the neat structure of Gr has weak interactions with other materials, which results in its poor dispersion.

The backbone of Gr consists of a stable multiring aromatic hydrocarbon chain; its edges and defects have the highest reaction activity [22]. The surfaces of Gr derivatives, such as graphene oxide (GO), contain a large amount of oxygen-containing functional groups, which can react with other functional groups and form chemical bonds [23, 24]. The functionalization of Gr can be divided into the following two categories: non-covalent and covalent functionalization [25, 26] (Figure 1).

Surface functional modification using noncovalent bonds primarily involves the formation of bonds between chemical substances and the Gr surface via intermolecular forces [27, 28], which has the advantage of maintaining the bulk structure and desirable properties of Gr or GO without destroying either its bulk structure or properties. Simultaneously, surface functional modification using non-covalent bonds can be used to improve the dispersibility of Gr, but the resulting graphene composite is unstable and its intermolecular forces are weak. Comparatively, covalent bond functional modification is achieved by introducing a functional group capable of forming a covalent bond with reactive double bonds or another oxygen-containing group on the Gr or GO surface [29, 30]. Compared with non-covalent

bond functionalization, covalent bond functionalization is more stable but affects performance because it destroys the structure of Gr.

3. Graphene Composite Materials

Figure 2 shows the primary preparation methods and applications of Gr composite materials.

Figure 2 shows Gr is a high-strength, two-dimensional material with high electrical and thermal conductivities. After being composited with inorganic materials or polymers, Gr composites are generally classified into the following two categories: Gr/inorganic and Gr/polymer composite materials [31, 32]. Composite materials have been widely used in photoelectric conversion materials, catalysts, structure-reinforcing materials, adhesives, functional film materials, and biomedical materials, among others.

3.1. Graphene/Inorganic Composite Materials

3.1.1. Preparation of Graphene/Inorganic Composite Materials. Gr/organic composite materials primarily include Gr/metal composite materials, Gr/ceramic composite materials, and Gr/other inorganic composite materials. Selection of the preparation method is dependent on the properties of inorganic materials and the application of the composite materials involved. The primary methods used for preparation include sinter molding, hydrothermal, electrochemical deposition, chemical reduction, and sol-gel methods, among others. Specific advantages/disadvantages and the application ranges of these methods are listed in Table 1.

3.1.2. Applications of Graphene/Inorganic Composite Materials

(1) Graphene/Metal Composite Materials

(i) Graphene/metal elements

Table 2 provides a summary of the properties and preparation methods of Gr/metal composite materials.

Because of its highly desirable mechanical properties, Gr has demonstrated significant potential in the reinforcement of metallic materials. Table 2 shows the composite materials comprising Gr and elemental metallic materials have primarily been used for increasing the mechanical strength of materials. Tensile strength increased by over 50% after Gr or RGO was added to metallic elements such as Al and Cu [37–41]. For example, Gr-Cu/Al prepared by Zhao showed a 77.5% increase in tensile strength over that of pure Cu.

Gr is able to enhance the thermal conductivity of metal materials because its thermal conductivity is higher than that of most metallic materials. Rho prepared a graphene/Cu composite material with a

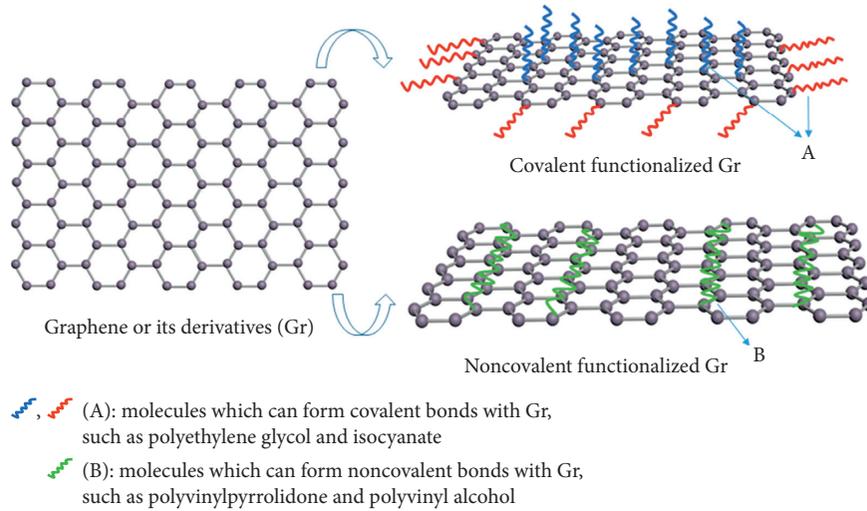


FIGURE 1: Covalent and noncovalent functionalization of graphene and its derivatives.

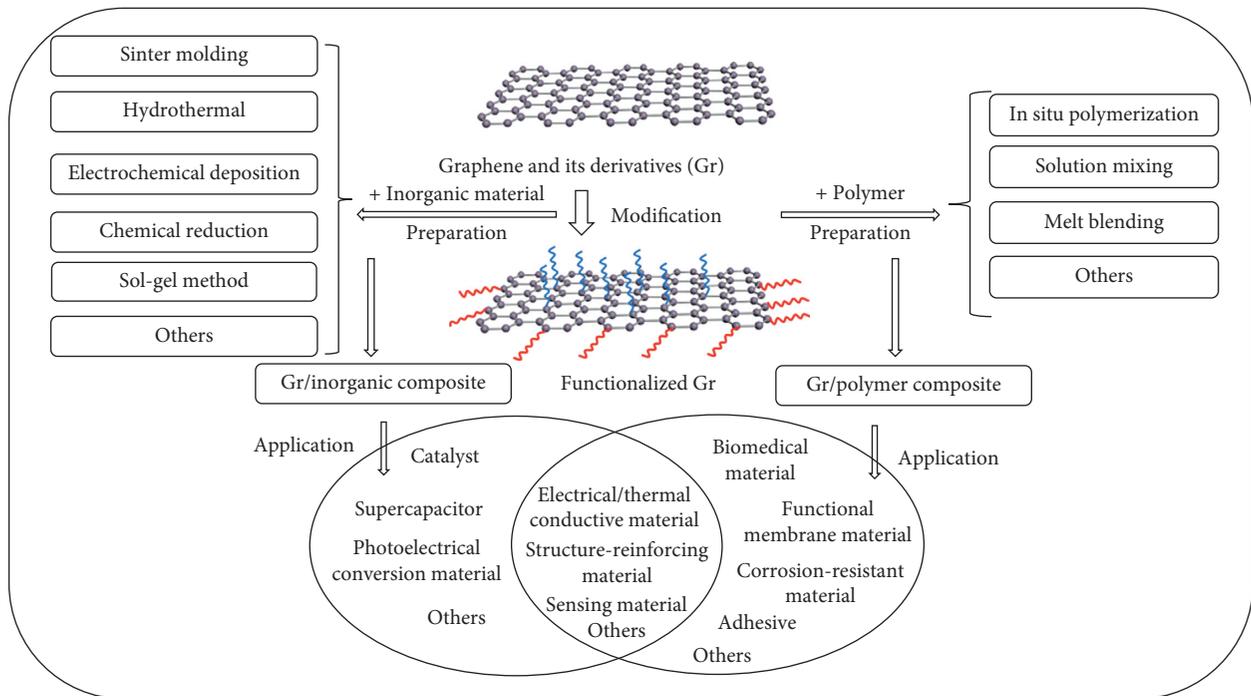


FIGURE 2: Preparation and application of graphene composite materials.

porosity of 35%, which exhibited a thermal conductivity 40% higher than that of pure Cu. Because of its large specific surface area, Gr can be used to increase the sensitivity of composite materials used in sensors. The addition of Gr to catalyst carriers enlarges the contact area between reagents and significantly enhances catalytic performance. Sensors using certain metal elements (e.g., Pt and Ag) as a matrix can be quite sensitive, and the addition of these metal elements can significantly increase catalysis efficiency [44–47]. Ouyang used Gr to coat a flexible Ag membrane sensor, which was used to detect malachite green with high sensitivity.

(ii) Graphene/metal oxide

Table 3 provides a summary of the properties and preparation methods of Gr/metal oxide composite materials.

Table 3 shows supercapacitors with excellent performance can be prepared using Gr/metal oxide composites. The specific capacities of the obtained capacitors were higher than those comprising metal oxides containing no Gr, and the cycle stability was good [49–56]. Of particular interest, one study by Liu et al. described the preparation of RGO/V₂O₅ composite material, which exhibited a capacitance of

TABLE 1: Preparation methods, advantages, disadvantages, and the scope of application of graphene/inorganic composite materials [33–36].

Preparation method	Advantages	Disadvantages	Scope of applications
Sinter molding	Simple fabrication process; shape of fabricated product close to the final shape of product needed; option of pressureless sintering for products with complex shapes; option of using hot-pressed or spark plasma sintering, which provide excellent properties	Limited variation in shape when using a single mold, high energy consumption at high temperatures, low production efficiency	Metallic powders or mixtures of metals and nonmetals primarily used in the fabrication of alloy, ceramic, and composite materials
Hydrothermal	Fabrication of materials with high purity, good dispersion of Gr in matrices, good and controllable grain shape, low production cost	High equipment requirement, relatively difficult technique, presence of various influencing factors during preparation	Metal or metal oxide composite materials usually used in the fabrication of electronic materials, magnetic materials, and sensor matrices
Electrochemical deposition	Uniform distribution of Gr, fabrication of a more complete structure, ability to obtain a composite material with uniform properties	Production of a thin coating of composite material, fabrication of a composite with low mechanical strength	Facile precipitation of metals and alloys from solution, which is primarily used in the production of metallic films
Chemical reduction	Simple and low-cost process resulting in a material with good performance	Possibility that reducing agent may be hazardous and dangerous	Metal-based Gr composite materials
Sol-gel	Easily controllable process, facile reaction, ability of Gr to mix uniformly with matrices at the molecular scale	Low production efficiency; occurrence of micropores in composite materials; easy occurrence of shrinkage, which affects product shape	Structural and film materials with metal oxide/Gr composite materials, use of composite materials as insulating materials

TABLE 2: Properties and preparation methods of graphene/metal composite materials.

Properties	Type	Preparation method	Loading	Result(s) after addition of graphene	Reference
Mechanical	Reduced graphene oxide (RGO)/Cu	Electrochemical deposition	2.5 vol.%	Elastic modulus and yielding strength were 1.3 and 1.8 folds, respectively, higher than those of pure Cu	[37]
	Gr/Cu	Sintering	0.5 wt.%	Yielding strength and tensile strength were 233.3% and 35.7%, respectively, higher than those of pure Cu	[38]
	Gr nanosheets (GNS)/Cu	Sintering	3.0 wt.%	Tensile strength was 69% higher than that of pure Cu	[39]
	Gr-Cu/Al	Sintering	3.0 wt.%	Tensile strength and hardness were 77.5% and 29.1%, respectively, higher than those of pure Al	[40]
	Gr/Al	Sintering	1.0 wt.%	Bending strength was 47% higher than that of 6061 Al matrix	[41]
Thermal	Gr/Cu	Sintering	—	Thermal conductivity was 40% higher than that of pure Cu	[42]
	Gr/Cu	Electrochemical deposition	—	Thermal conductivity was 24% higher than that of pure Cu	[43]
Sensing	Gr/Ag	Electrochemical deposition	—	Concentration of antibiotics in water was determined successfully	[44]
	Gr/Ag	Chemical reduction	—	Precise detection of malachite green was achieved successfully	[45]
Catalytic	RGO/Pt	Chemical reduction	—	Catalytic efficiency of fuel batteries was enhanced successfully	[46]
	Graphene nanospheres (GNs)/Pt	Chemical reduction	—	Catalytic efficiency of hydrogen evolution reaction was enhanced successfully	[47]
Antibacterial	RGO/Ag	Chemical reduction	—	Use as an antibacterial and photothermal ablation agent was demonstrated	[48]

TABLE 3: Properties and preparation methods of graphene/metal oxide composite materials.

Properties	Type	Preparation method	Result(s) after addition of Gr	Reference
Electrical	GO/CuO	Electrochemical deposition	Specific capacity was enhanced by 72%	[49]
	Gr/SnO ₂	Chemical reduction	Excellent cycling stability was achieved	[50]
	RGO/V ₂ O ₅	Hydrothermal method	Specific capacitance was 384 F/g at 0.1 A/g	[51]
	RGO/V ₂ O ₅	Hydrothermal method	Capacitance retention was nearly 81.5% after 200 cycles	[52]
	RGO/V ₂ O ₅	Hydrothermal method	Specific capacitance was 450.5 F/g at 0.5 A/g; capacitance retention was nearly 73.3% at 10 A/g	[53]
	Microwave-exfoliated graphene oxide (MEGO)/MnO ₂	Chemical reduction	Mass specific capacitance and volumetric specific capacitance achieved values of 256 F/g and 640 F/cm ³	[54]
	Gr/MnO-NiO GrF/Co ₃ O ₄	Interface method In situ growth	Specific capacity was 813 F/g at 0.5 A/g Specific capacity was 277.5 F/g at 25 A/g	[55] [56]
Catalytic	Gr/SnO ₂	Hydrothermal method	Higher photocatalytic activity was achieved, resulting in the elimination of 93% of rhodamine B	[57]
	Gr/MnO ₂	Hydrothermal method	Methanal conversion was 80% higher than that of MnO ₂	[58]
	Gr/TiO ₂	Hydrothermal method	Degradation of methylene blue was more efficient than that of pure TiO ₂	[59]
	RGO/V ₂ O ₅	Hydrothermal method	Maximum photocatalytic degradation was achieved, and discolor efficiency for methylene blue was 85%	[60]
	Gr/TiO ₂	Hydrothermal method	Excellent adsorption performance and catalytic capability under visible light were achieved; the removal rate of methylene blue was over 90%	[61]
Sensing	RGO/CuO	Electrochemical deposition	Detection of low concentrations of NO ₂ (1–75 ppm) was achieved	[62]
	RGO/SnO ₂	Drop casting	Simultaneous detection of H ₂ S and acetone was achieved; this method can be used to diagnose human diabetes using a noncontact technique	[63]
	SnO ₂ @RGO-poly aniline (PANI)	In situ polymerization	Room-temperature response to 100–10000 ppm methane was intensified	[64]

450.5 F/g at 0.5 A/g and a capacitance retention of 81.5% at 10 A/g.

The photothermal effect of Gr can enhance the catalytic efficiency of traditional thermal catalysts. Table 3 shows Gr and metal oxides are excellent materials for use in the photocatalytic degradation of dye [57–61]. For example, the photocatalytic efficiency of Gr/SnO₂ composites prepared by Othmen et al. for the degradation of rhodamine B was 93%.

The addition of Gr can increase the specific surface area of composite materials, intensify the carrier transfer between gas and composite materials, and enhance the sensitivity of gas detection. Several research groups have prepared various Gr/metal oxide composite materials able to detect different gases [62–64]. Of particular interest is a composite sensor prepared by Choi et al. that can detect H₂S and acetone gas; this sensor has been used to detect diabetes.

(2) *Graphene/Ceramic Composite Materials.* Table 4 presents a brief summary of the properties and preparation methods of Gr/ceramic composite materials.

Currently, the number of studies on Gr/ceramic composite materials is smaller than that of studies on Gr/metal

composite materials. Table 4 shows these studies are primarily focused within the fields of mechanics and on the electrical and thermal properties of the composite materials. After the addition of Gr to ceramic materials, the resistance against force conduction can prevent the diffusion of cracks, resulting in a toughening effect. The toughening mechanism is summarized by Ahmad et al. [74]. Securely anchored Gr around the matrix can form large area of interfaces with the matrix, increasing the interfacial friction; therefore the energy required for pulling out Gr from the matrix will be greater. The fracture toughness of the Gr/Al₂O₃ ceramic composite material prepared by Ahmad et al. [66] was 72% higher than that of the pure Al₂O₃ ceramic.

At present, the use of Gr to enhance the electrical properties of ceramics remains in the research stage. The electrical and thermal conductivities of composite materials are obviously affected by the paths constructed by Gr in the matrix. Lin et al. [72] and Xia et al. [73] determined the addition of Gr reduces the thermal conductivity of the composite. In addition, the electrical conductivity of Gr/B₄C prepared by Tan et al. [70] was 5000 S/m.

There is an intensely research interest in hybrid ceramic composites. The addition of nanoparticles greatly enhances the mechanical properties of hybrid composites. Better mechanical properties are correlated with distinctive

TABLE 4: Properties and preparation methods of graphene/ceramic composite materials.

Properties	Composition	Preparation method	Loading of Gr	Result(s) after addition of Gr	Reference
Mechanical	Gr/Al ₂ O ₃	Sintering	1.0 wt.%	Bending strength and fracture toughness were 26.4% and 67.6% higher than those of pure Al ₂ O ₃ ceramic	[65]
	Gr/Al ₂ O ₃	Sintering	0.5 wt.%	Fracture toughness was 72% higher than that of pure Al ₂ O ₃ ceramic	[66]
	Gr/Si ₃ N ₄	Sintering	1.5 vol.%	Fracture toughness was 57.5% higher than that of composite material	[67]
	Gr/AlN	Sintering	1.49 vol.%	Bending strength and fracture toughness were 17.28% and 30.17%, respectively, higher than those of pure AlN ceramic	[68]
Electrical	Gr/Al ₂ O ₃	Sintering	15 vol.%	Electrical conductivity parallel to voltage direction reached 5709 S/m	[69]
	Gr/B ₄ C	Sintering	4 vol.%	Electrical conductivities perpendicular and parallel to voltage direction were 5000 ± 26 S/m and 3250 ± 22 S/m, respectively	[70]
	Gr/Si ₃ N ₄	Sintering	25 vol.%	Maximum electrical conductivity perpendicular to the pressed direction was 4000 S/m	[71]
Thermal	Gr/Al ₂ O ₃	Sintering	8.5 vol.%	Thermal conductivity decreased from 32 W/mK (Al ₂ O ₃ ceramic) to 20 W/mK	[72]
	Gr/AlN	Sintering	2 wt.%	Thermal conductivity decreased from 92.5 W/mK (AlN ceramic) to 37.4 W/mK	[73]

toughening mechanisms of crack-deflection as well as crack-bridging induced by reinforcing phases upon establishing sturdy interfaces with the alumina matrix. The pull-out effect of nanoparticle is also taken into consideration. Ahmad et al. [75] prepared alumina (Al₂O₃) ceramic matrix with silicon carbide nanoparticles (SiC_{NP}) and graphene nanoplatelets (Gr). The fracture toughness and microhardness improved by 160% and 27%, respectively. In addition, Ahmad and Islam [76] prepared inductively sintered alumina ceramic hybrid nanocomposites with 4 vol.% multiwalled carbon nanotubes (mWCNT) and 6 vol.% SiC_{NP}. Homogenously dispersed reinforcing inclusions led hybrid nanocomposites to respective 81% and 25% higher fracture toughness and microhardness over the monolithic alumina. Some researchers have studied the application of hybrid ceramic composites in the field of corrosion resistance and wear resistance. Graphene nanoplatelet-coated B₄C reinforced Al-Si matrix semiceramic hybrid composites were prepared by Polat et al. [77]. When the boron carbide surface was coated with 0.5 vol.% Gr, it was determined that the specific wear resistance increased by 55% and the corrosion resistance decreased by 12%.

(3) *Graphene/Other Inorganic Composite Materials.* The application of Gr in the field of electrochemistry, optics, and catalysis can also be expanded after compositing with other inorganic materials. Geng et al. [78] prepared GO/CdSe quantum-dot composites and a flexible light-transmitting conductive film device, which had a light sensitivity and photoelectric conversion efficiency four- and tenfold, respectively, higher than those of pure CdSe quantum dots. Gao et al. [79] prepared GO/CdS composites via a self-assembly process. The GO/CdS composites exhibited high-efficiency photocatalytic degradation and antibacterial effects for several types of sewage under visible light irradiation, which was different from the results obtained for pure CdS nanoparticles. Yu et al. [80] prepared CdTe quantum-dot/Gr

nanocomposites, and the prepared electrodes exhibited excellent electrochemical catalytic activity, good biocompatibility, and high sensitivity for detecting the oxidation of uric acid (UA) and dopamine (DA).

3.2. Graphene/Polymer Composite Materials

3.2.1. Preparation of Graphene/Polymer Composite Materials.

At present, the preparation of Gr/polymer composite materials can be divided into the following dispersion methods: in situ polymerization, melt blending, and solution blending. Table 5 presents a summary of these methods. Among the three methods, in situ polymerization is the most complicated because its reaction process is difficult to control and it has fewer applications than melt blending and solution blending. Melt blending is easily industrialized, and solution blending provides a uniform dispersion.

3.2.2. Applications of Graphene/Polymer Composite Materials

(1) Graphene/Conventional Thermoplastic Composite Materials

(i) Conventional plastics

Table 6 provides a summary of the properties and preparation methods of Gr/conventional thermoplastic composite materials.

Because of its high theoretical strength, Gr has been used as a reinforcing filler for most conventional plastics. Table 6 shows how a small amount of Gr effectively enhanced the strength and modulus of polyolefin materials [84–87]. For example, the mechanical strength of functionalized-RGO/poly(vinylidene fluoride) (F-RGO/PVDF) films prepared by Luo et al. was 42% higher than that of conventional RGO/PVDF films.

TABLE 5: Preparation methods, advantages, disadvantages, and the scope of applications of graphene/polymer composite materials [81–83].

Preparation method	Advantages	Disadvantages	Scope of applications
In situ polymerization	Uniform dispersion; achievement of product control via changes in reaction conditions	Control issues from the cooperative effect caused by reaction temperature, among other factors; complicated reaction, making industrialization relatively difficult	Various polymer-based composite materials in extensive fields
Solution mixing	Uniform dispersion; beneficial method for achieving obvious enhancement of desirable properties	Residual solvent, difficult recycling process, and environmental pollution; no suitability for scaled manufacturing	Wide applications, various polymer-based Gr composite materials, high-insulation materials and coatings
Melt blending	Compliance with nonhazardous solvent use and polar and nonpolar polymers; simple operation and easy achievement of industrialization	Poor dispersion properties, facile dust formation	Composite materials with thermoplastic matrices; applications in the field of electrical and thermal conduction (but still in the stage of marketing promotion)

TABLE 6: Properties and preparation methods of composite materials comprising graphene and conventional polyolefin or its derivatives.

Properties	Types	Preparation method	Loading of Gr	Result(s) after addition of Gr	Reference
Mechanical	Partially RGO/(polypropylene/ethylene propylene rubber) ((pRGO)/(PP/EPR))	Melt blending	2.0 wt.%	Tensile strength and Young's modulus were enhanced	[84]
	Alkyl-graphene nanoplatelets (GNPs)/PP	Melt blending	0.5 phr	Bending modulus, which affects strength and energy-storage modulus, was enhanced	[85]
	Carbon nanotubes (CNTs)/Gr/polystyrene (PS)	Solution blending	—	Glass transition temperature (T _g), modulus, and hardness were enhanced	[86]
	Functionalized-RGO/poly(vinylidene fluoride) (F-RGO/PVDF)	Solution blending	—	Tensile strength was 42% higher than that of conventional RGO/PVDF composite films	[87]
Thermal	Gr/polyethylene (PE)	Melt blending	10.0 wt.%	Thermal conductivity reached 1.84 W/mK	[88]
	Gr/polyvinyl alcohol (PVA)	Solution blending	20–65 wt.%	Thermal conductivity increased from 10.15 to 36.81 W/mK	[89]
	GO/PVDF	Melt blending	1.0 wt.%	Thermal conductivity was enhanced by 132.5%	[90]
Electrical	GO/PVA/PVP (polyvinylpyrrolidone)	Solution blending	—	Electrical conductivity was 6.13×10^{-4} S/cm	[91]
	Large-area RGO/poly(vinylidene fluoride-co-hexafluoropropylene) (rLGO/PVDF-HFP)	Solution blending	27.2 wt.%	Electrical conductivity was 653000 S/m	[92]
	Poly(ethylene glycol)- (PEG-) Gr/PVDF	Solution blending	15 wt.%	Electrically active crystal content was increased by 65.9%	[93]
	Few layer graphene (FLG)/PVDF	Solution blending	1–90 wt.%	Electrical conductivity was 81.9 S/cm	[94]
Insulating	Hexadecylamine-functionalized Gr sheet (HDA-GS)/PVA	Solution blending	5.0 wt.%	Oxygen permeation coefficient was reduced by 81%	[95]
	Thermally reduced GO (TRGO)/PP	Melt blending	1.0 wt.%	Oxygen permeation coefficient was reduced by 35%	[96]
Medical	GO/ultra-high-molecular-weight polyethylene (UHMWPE)	Solution blending	2.0 wt.%	Wear reduced significantly; good biocompatibility achieved at all concentrations studied	[97]
	GO/high-density PE/UHMWPE	Melt blending	1.0 wt.%	Tensile strength and modulus and biocompatibility were enhanced	[98]
	Mechanically exfoliated graphene (MEG)/PVA	Gel spinning	0.3 wt.%	High strength, good antibacterial performance, and low cytotoxicity were achieved	[99]
Other	GNPs/PP	Melt blending	0.5 wt.%	Aging of PP matrix was slowed	[100]
	GO/PP-g-poly[2-(dimethylamino)ethyl methacrylate (DMAEMA)]	Self-assembly	—	Organic material in wastewater was effectively removed	[101]
	Functionalized GO (FGO)/PVA	Self-assembly	—	Excellent fire retardancy of coated PVA films was achieved	[102]

Gr has also been used to enhance the electrical and thermal conductivities of polyolefin materials. In general, the required Gr content is higher than 10 wt.% because only a large amount of Gr forms electrically conductive and thermally conductive paths in polymer matrices [88–94]. For example, the thermal conductivity of the Gr/polyvinyl alcohol (PVA) prepared by Liu et al. was approximately 260% that of PVA. In addition, the electroactive crystal content of poly(ethylene glycol)- (PEG-) Gr/PVDF prepared by Chen et al. was 65.9% higher than that of the matrix alone.

Because of its sheet structure, Gr has been used to enhance the insulating properties of polyolefin materials. Chang [95] prepared a hexadecylamine-functionalized Gr sheet/polyvinyl alcohol (HDA-GS)/PVA composite and found that an HDA-GS loading of 5 wt.% resulted in a 81% reduction in the oxygen transmission coefficient compared with that of PVA alone.

Because the addition of Gr enhances the strength of composite materials, several researchers combined it with several biocompatible polyolefin materials to fabricate biomedical materials with high biocompatibility and good antibacterial properties [97–99]. For example, Gowland et al. prepared GO/ ultra-high-molecular-weight polyethylene (UHMWPE) composite materials for use in the replacement of bone joints.

In addition, Gr/polyolefin composite materials have potential applications in various fields, including uses in antiaging and flame-retardant materials [100–102].

(ii) Conventional engineering plastics

(a) Gr/polyamide composite materials

The blending of Gr and polyamide (PA) was performed to enhance the electrical conductivity of PA. Ma et al. [103] and Xiang et al. [104] prepared polycaprolactam/thermally reduced GO composite materials (PA6/TRGO) and found these materials possessed an ultralow threshold for percolation flow. In addition, both groups showed significant increases in electrical conductivity compared with that of PA alone. In addition, Gr has been used to improve the permeability of PA-based reverse osmosis (RO) films. Shi et al. [105] prepared a GO/PA ultrathin film with excellent properties useful for the desalting process of RO films as well as excellent stability and remarkable permeability. Lai et al. [106] embedded GO nanosheets (GONS) into ultrathin crosslinked PA layers. After the addition of 0.02 g/m² GO, the removal rates of the obtained thin-film nanocomposite (TFN) membranes were 95.8% and 97.7% for Na₂SO₄ and MgSO₄, respectively.

Because of the excellent flexibility of PA, Yin et al. [107] used PA to prepare flexible skin-like pressure sensors using Gr/PA interlocked fabrics that could be used in the preparation of artificial electronic skin and wearable electronic devices.

(b) Graphene/polyester composite materials

Gr has been used to increase the sensing efficiency of Gr/polyester composites. Wang et al. [108] prepared a multifunctional RGO/poly(ethylene terephthalate) (PET) fabric. When it was used in a strain sensor, the RGO/PET fabric exhibited high sensitivity, demonstrating its great potential for use in the development of wearable smart devices. Seo et al. [109] coated PET with a graphene nanoplatelet- (GNP-) polyaniline composite material for the fabrication of a flexible sensor for detecting hazardous chemicals.

In addition, Gr has been used to enhance the heat resistance of polyester materials and fire retardancy when composited with specific materials. Xing et al. [110] used low-molecular-weight poly(ethylene terephthalate) (PET-) grafted GONS to improve the compatibility between GO and PET, which simultaneously improved the thermal stability and crystallization properties of PET. Qi et al. [111] prepared an aluminum hypophosphite/RGO (AHP/RGO) hybrid flame-retardant material with high thermal stability. The AHP/RGO exhibited better flame retardancy than neat AHP for use with poly(butylene terephthalate) (PBT).

(c) Graphene/polycarbonate composite materials

The addition of Gr to polycarbonate (PC) materials has been used for electromagnetic shielding. Bagotia et al. [112] added in situ RGO (IRGO) to a PC/ethylene-methyl acrylate (EMA) matrix, and the resultant mechanical hardness of the composite material was significantly higher than that of PC/EMA. When the IRGO content was 15 phr, the best effects from electromagnetic shielding were achieved in the X-band frequency range (8.2–12.4 GHz). In addition, Gr has been used to enhance the solvent resistance of PC. Wang et al. [113] prepared superhydrophobic and superhydrophilic porous RGO/PC single-sheet composite materials that were able to adsorb a variety of oils and organic solvents from water.

Furthermore, Gr/PC composites have demonstrated their use for potential applications in smart materials. Leeladhar and Singh [114] prepared a GO/PC composite material with a double-layer structure that exhibited a smart response to infrared light. Therefore, the composite material was able to act as a light actuator,

which could be used in applications such as sunlight-driven curtains, infrared light-driven curtains, and boxes that automatically fold upon exposure to light.

(d) Graphene/polyphenylene ether

Gr has been used to enhance the dielectric properties of polyphenylene ether. Wang et al. [115] prepared TRGO-coated calcium titanate (CCTO) as a filler material for polyphenylene ether. After the addition of 30 wt.% of Gr-coated CCTO (g-CCTO), the dielectric constant of the resulting composite material was 8.60. Notably, the dielectric constant of the untreated CCTO/polyphenylene oxide (PPO) composite was 7.08. Gr/polyphenylene ether has potential applications in the gas separation and capture of carbon dioxide. Rea et al. [116] prepared Gr/PPO composites. The composites containing a small amount of Gr possessed higher permeability and higher He/CO₂ selectivity than pure PPO.

(e) Graphene/polyurethane

The addition of Gr has been used to enhance the performance of polyurethane (PU) coatings and adhesives. Mohammadi et al. [117] prepared an environmentally friendly GO/hydrophilic PU nanocomposite (WPU/GO) with a 99.8% antiseptic inhibition efficiency. In addition, Wan and Chen [118] fabricated self-repairing GO/hydrophilic PU nanocomposites via solution blending. The resulting nanocomposite possessed good thermal stability and mechanical properties and could be used in coatings and adhesives.

Several research groups showed Gr/PU composite materials have potential applications in controlling water pollution. Zhou et al. [119] prepared anthraquinone-2-sulfonate-RGO-PU foam (AQS-RGO-PUF) and showed AQS-RGO-PUF possessed a superior catalytic performance for the biodecoloration of acid red 18 (AR18) compared with the performance of PUF alone. In addition, the combination of the *Shewanella* strain with AQS-RGO-PUF enhanced the treatment efficiency for azo dye-containing wastewater. Xia et al. [120] prepared superhydrophobic RGO-coated PU (RGO@PU) sponges with a high oil-absorption rate (oil removal occurred within 5 seconds) and a high oil-water separation efficiency (approximately 99%).

(iii) Graphene/biodegradable polymer composite materials

Table 7 provides a summary of the properties and preparation methods of composite materials comprising Gr and biodegradable polymers.

The majority of biodegradable polymers exhibit excellent biocompatibility, and their potential applications in the biomedical field can be expanded upon reinforcement with Gr. Table 7 shows the

majority of Gr/biodegradable materials possess good biocompatibility and high bacterial resistance and promote the regeneration of tissue and bone. The PCL/GNS bracket prepared by Caetane et al. [134] significantly promoted cell proliferation and bone regeneration.

The two-dimensional structure of Gr has been used to enhance the isolation performance of biodegradable composite materials. Previously, our research group prepared a PLA/GO composite material, and the experimental results indicated the introduction of only a small amount of GO enhanced the isolation performance of PLA. You et al. [121] prepared a composite nanofilm containing poly(3-hydroxybutyric acid-co-3-hydroxyvaleric acid)/GO (PHBV/GO) and found the oxygen permeability coefficient was 48 cm³/(m²·day) when the GO loading was 2.0 wt.%, which was lower than that of PHBV alone.

In addition to the above findings, the addition of Gr to biodegradable polymers has been applied in electrical materials, fire retardant materials, sensors and catalysts, etc. [123, 124, 126, 127]. However, the number of studies in these fields is relatively few, and further study on the enhancement of relevant properties is needed.

(2) Graphene/Conventional Thermoset Plastics

(i) Graphene/unsaturated polyester

When Gr exhibits good dispersibility within a polymer matrix, Gr acts as a reinforcing filler with excellent mechanical properties. He et al. [136] synthesized GO and GO derivatives containing vinyl and alkyl functional groups (mGO) and dispersed these substrates in unsaturated polyester resin (UPR) to fabricate composite nanomaterials. The fracture energy (G_{IC}) was 55% higher than that of neat GO when the loading of mGO was 0.04 wt.%.

In addition, Gr has been used to enhance the fire retardancy of thermoset materials such as UPR. Jiang et al. [137] first synthesized a novel organophosphorus oligomer (DHP) and then prepared the G-DHP composite material. The introduction of 2.0 wt.% G-DHP significantly enhanced the fire retardancy of the UPR matrix, which was demonstrated by a 41.79% decrease in the peak exothermic rate (PHRR).

(ii) Graphene/phenolic resin

Traditional phenolic resin (PF) cannot meet the necessary performance requirements, especially those needed for the ablation resistance of advanced aerospace vehicles because PF is easily oxidized. Lv et al. [138] prepared SiO₂/RGO/PF binary hybrid nanomaterials to help oxidize PF. When the content of the dispersed SiO₂ nanoparticles on graphene (G-S) was 3 wt.%, the 3 wt.% of G-S (P-G-S-3) exhibited

TABLE 7: Properties and preparation methods of composite materials comprising graphene and biodegradable polymers.

Composition	Preparation method	Loading of Gr	Result(s) after addition of Gr	Reference
GO/poly(3-hydroxybutric acid-co-3-hydroxyvaleric acid (PHBV))	Solution blending	2.0 wt.%	Oxygen permeation rate decreased from $63 \text{ cm}^3/(\text{m}^2\cdot\text{day})$ to $48 \text{ cm}^3/(\text{m}^2\cdot\text{day})$	[121]
GO/collagen/PHBV	Solution blending	0.3 wt.%	High mechanical strength and good antibacterial performance, necessary for covering wounds, were achieved	[122]
Functionalized graphene sheets (FGS)/PHBV	Ball milling	0.1–3.0 wt.%	High gas resistance and low percolation threshold, significantly enhanced electric conductivity	[123]
FGO-hydroxyquinoline/poly(lactic acid) (FGO-HQ/PLA)	Melt blending	6.0 wt.%	Excellent fire retardancy was achieved	[124]
GO-g-poly-L-lactide (PLLA)/PLLA	Solution blending	0.1–3.0 wt.%	High antibacterial activity was achieved; potential for use in biomedical fields was demonstrated	[125]
Zeolitic imidazolate framework-8 (ZIF-8)@GO/PLA	Solution blending	3.0 wt.%	Tensile strength higher than that of pure PLA was achieved; strong removal capability for methylene blue was demonstrated	[126]
Graphene nanorods (GNRs)/PLA	Melt blending	—	Use in sensor that monitors common aquatic and land environments was demonstrated	[127]
Gr/PLA	Melt blending	1.0 wt.%	Oxygen permeation rate was reduced by 36%	[128]
GO with octadecylamine/polybutylene succinate (GOODA/PBS)	Solution blending	0.1 wt.%	Tensile strength and Young's modulus were enhanced by 50% and 58.9%, respectively	[129]
GONS/poly(butylene adipate-co-terephthalate) (PBAT)	Solution coagulation	0.35 vol.%	Coefficients of permeability of O_2 and water vapor were reduced by 70% and 36%, respectively	[130]
GNPs/PBAT	Melt blending	0–15.0 vol.%	Dielectric constant of polymer was enhanced significantly	[131]
RGO/polycaprolactone (PCL)	Solution blending	0-0.1 wt.%	Regeneration of dental pulp stem cell (DPSC) nerve was promoted	[132]
Poly-D-glucosamine (PDG)/PCL	Melt blending	—	Tensile and yield strengths were 2.4- and 2.6-fold, respectively, those of PCL, indicative of self-healing behavior	[133]
GO/P1-latex protein/PCL	Melt blending	0.13–0.78 wt.%	Gr/P1-latex/PCL protein bracket was used in bone regeneration	[134]
Gr/PCL	Melt blending	0.25–0.75 wt.%	Gr/PCL was demonstrated to be beneficial for tissue formation and bone regeneration	[135]

a lower thermal oxidative decomposition rate and a higher termination temperature for thermal oxidative decomposition than neat PF.

Gr/phenolic resin composites have also been used as dielectric materials. Wu et al. [139] prepared three-dimensional Gr network/phenolic resin composite materials with adjustable and low negative dielectric constants. The results of this study showed negative dielectric constants could be controlled by controlling the Gr content.

(iii) Graphene/epoxy resin composite materials

The addition of Gr was used to enhance the corrosion resistance of epoxy resin (ER). Zheng et al. [140], Mousavi et al. [141], and Yang et al. [142] prepared modified GO/ER composite materials able to obviously enhance the corrosion resistance of epoxy coatings on matrices.

Furthermore, the addition of Gr was used to enhance the fire retardancy of ER. Zhang et al. [143] studied the effect of graphene nanoplatelets (GNP)

on the combustion properties of ER. The PHRR of GNP/ER composites was much lower than that of pure ER.

In addition, Gr was used to enhance the thermal conductivity of ER. Olowojoba et al. [144] prepared RGO/ER composite materials. When the RGO content was 0.06 wt.%, the thermal conductivity of the composite material was 0.25 W/mK, which was 40% higher than that of unmodified ER.

4. Prospects and Developmental Trends

Because of the in-depth studies on Gr, the fundamental research and application of Gr composite materials have progressed and demonstrated Gr has significant application potential in the fields of aerospace, transportation, medical equipment, electrical and thermal conductive materials, and construction materials. In Gr/inorganic composites, Gr provides good mechanical strength and can be used as a structural material useful for compositing with elemental metals; sinter molding is generally adopted for these types of

composites. For the compositing process between metal oxides and Gr, hydrothermal reduction or chemical reduction methods are usually adopted, and the resulting product is generally used in supercapacitors. Because of the unique two-dimensional structure of Gr, it can also be used in sensors and catalysts. Ceramic/Gr composites are less studied than metal/Gr composites. Sintering is commonly adopted for the fabrication of ceramic/Gr composites, and these types of composites are generally used for structural reinforcement. For Gr/polymer composite materials, melt blending is advantageous for industrial production while solution blending or in situ polymerization may be more advantageous for laboratory preparations. Gr is able to combine with different polymers and provide significant advantages for the preparation of sensing materials, functional films, biomedical materials, and anticorrosion coatings, among other applications. However, several problems that must be solved remain. (1) The Gr preparation method of Gr is complicated and expensive; thus, inexpensive batch preparation is difficult. (2) The processing temperatures for inorganic materials such as metals and ceramics are high and could destroy the structure of Gr. Therefore, processing difficulty and cost are high when Gr and inorganic materials are composited. (3) When Gr is composited with organic polymers, Gr usually exists in the form of agglomerates because its specific surface area and surface free energy are large and strong electrostatic and Van der Waals forces exist between the Gr sheets. Agglomerates easily cause defects in the polymer composite, which is not conducive to enhancing its material properties. Therefore, the search for effective modification methods and the achievement of good dispersion and the stripping effect of Gr in polymer matrices are important prerequisites in the manufacturing of ideal nanocomposites. In the future, the preparation methods and structure-property relationships of Gr composite materials should be optimized so Gr composite materials can ultimately promote additional development in materials engineering.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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