Green Graphene-Based Polymer Composites: A Pathway Toward Sustainable Polymers

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Editor's note: Green composites are becoming increasingly recognized as a sustainable alternative to the environmental problems caused by the extensive use of petroleum-based plastics. In this review paper, Yazdani provides an overview of recent advancements in graphene-based sustainable and green composites, focusing specifically on those developed using biodegradable polymers as the main matrix.

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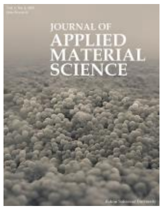

Review

Green Graphene-Based Polymer Composites: A Pathway Toward Sustainable Polymers

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Abstract

Green composites, as environmentally friendly materials, are rapidly gaining recognition as a sustainable solution to the environmental issues caused by the widespread use of petroleum-based plastics. These composites typically incorporate natural fibers and biodegradable polymers specifically designed to reduce environmental impact. This review paper aims to provide a comprehensive overview of green composites, with a particular focus on those developed from natural fibers such as cellulose, starch, and other bio-based materials. Compared to traditional plastics, these materials exhibit enhanced physical and mechanical properties, along with higher biodegradability. However, challenges such as surface compatibility and the retention of mechanical properties after moisture absorption persist, requiring further research. The paper also explores recent advancements in improving the properties of these composites through the incorporation of nanomaterials like nanocellulose and graphene. Furthermore, the paper discusses the diverse applications of green composites across industries such as packaging, biomedical, and automotive sectors.

Keywords: Green composites, Graphene oxide, Sustainable materials, Natural fiber reinforcement.

1. Introduction

In recent decades, growing environmental awareness and the increasing demand for sustainable materials have stimulated extensive interest in the use of natural and renewable resources for the development of advanced composite systems [1-3]. These materials, known as "green composites" or "biocomposites," have emerged as promising alternatives to conventional composites based on fossil resources due to their unique features, including light weight, high strength, recyclability, biodegradability, reduction of greenhouse gas emissions, decreased reliance on non-renewable resources, and diminished environmental impact [4-6]. Green composites have established a distinct position in the field of composite materials, as they not only serve as effective solutions to environmental challenges but also promote the efficient utilization of available renewable raw materials. These materials typically consist of biodegradable or bio-based polymer matrices reinforced with natural fibers or environmentally benign nanomaterials. Their appealing features include low density, high specific strength, biodegradability, recyclability, reduced carbon footprint, and decreased dependence on non-renewable resources [7, 8].

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Initial research in this field concentrated on incorporating natural fibers such as hemp, flax, and jute into biodegradable polymer matrices (Table 1). These early green composites were primarily investigated as substitutes for conventional, non-biodegradable materials in various applications. The mid 2000s to 2010s witnessed a surge in the development of biodegradable polymers, particularly polylactic acid (PLA), polyhydroxybutyrate (PHB), and thermoplastic starch (TPS). These matrices attracted significant attention due to their inherent biodegradability, mechanical performance, and low toxicity. Reinforcement with natural fibers or nanoparticles, such as cellulose nanofibers (CNFs), led to improvements in thermal stability, water resistance, and mechanical integrity. Over the past decade, research has increasingly focused on enhancing the functional performance of green composites to make them competitive with conventional materials in high-performance sectors. Strategies such as fiber surface modification, hybrid reinforcement, and nano enhancement have enabled the extension of green composites into demanding applications including aerospace, automotive, biomedical devices, cosmetics, water treatment, agriculture, and electronics [7, 9-12].

In developing green supercapacitors, materials such as graphite foil, sodium acetate, and ester-based porous membranes, which also exhibit superior performance compared to conventional materials, have been identified as more environmentally friendly alternatives. Similarly, in brake lining materials, hybrid composites reinforced with basalt, shell, and alumina, and phenolic resin polymers have demonstrated lower wear rates and water absorption compared to traditional asbestos-based brake materials. Additionally, the use of environmentally compatible materials in additive manufacturing, such as blending polylactic acid with MoS_2 as solid lubricants, has shown potential for reducing environmental impact, despite certain challenges related to stability [13, 14].

One major area of innovation involves the integration of graphene-based materials into green composites. Graphene, graphene oxide (GO), and reduced graphene oxide (rGO) exhibit outstanding mechanical, thermal, and electrical properties and serve as highly effective multifunctional reinforcements. Their incorporation improves load transfer, reduces moisture uptake, and enhances barrier and conductive properties. Importantly, green synthesis approaches for graphene derivatives—using plant extracts, biomass, or other non-toxic reducing agents—have emerged as sustainable and cost-effective alternatives to traditional methods. Additionally, in green composites, graphene serves as a reinforcing agent that improves the adhesion between the matrix and natural fibers, leading to enhanced mechanical strength and reduced cracking [20-22].

Green graphene-based polymer composites represent a class of advanced sustainable materials that combine

Table 1. Overview of green composites: components and applications

Type of Green Composite	Bio-based Polymer Matrix	Natural Reinforcement	Key Properties	Typical Applications	Ref.
PHB + Bamboo	Polyhydroxybutyrate (PHB)	Bamboo fibers	Good thermal stability, biodegradable	Construction, furniture	[1]
Soy Resin + Hemp	Soy-based resin	Hemp fibers	Moderate mechanical performance	Coatings, building materials	[2]
Green PE + Sisal	Bio-based Polyethylene	Sisal fibers	Lightweight, moisture-resistant	Automotive, household goods	[4]
PCL + Flax	Polycaprolactone (PCL)	Flax fibers	Flexible, biocompatible	Tissue engineering, scaffolds	[5]
PLA + Hemp	Polylactic Acid (PLA)	Hemp fibers	Biodegradable, moderate strength	Packaging, automotive parts	[7]
PLA + Cellulose Nanofibers	Polylactic Acid (PLA)	Cellulose nanofibers (CNF)	High strength, improved barrier properties	Food packaging, biomedical devices	[15]
TPS + GO	Thermoplastic Starch (TPS)	Graphene Oxide (GO)	Improved mechanical and thermal stability	Biodegradable films, electronics	[15]
PLA + Vermiculite + Waste Cellulose	PLA	Waste cellulose + Vermiculite	Enhanced mechanical strength, eco-friendly	Packaging, agriculture	[16]
Chitosan + Graphene Oxide	Chitosan	Graphene Oxide (GO)	High adsorption, antimicrobial, and mechanical stability	Water treatment, biomedical	[17, 18]
PLA + PCL	PLA + PCL blend	–	Shape memory effect, tunable flexibility	Smart textiles, medical implants	[19]

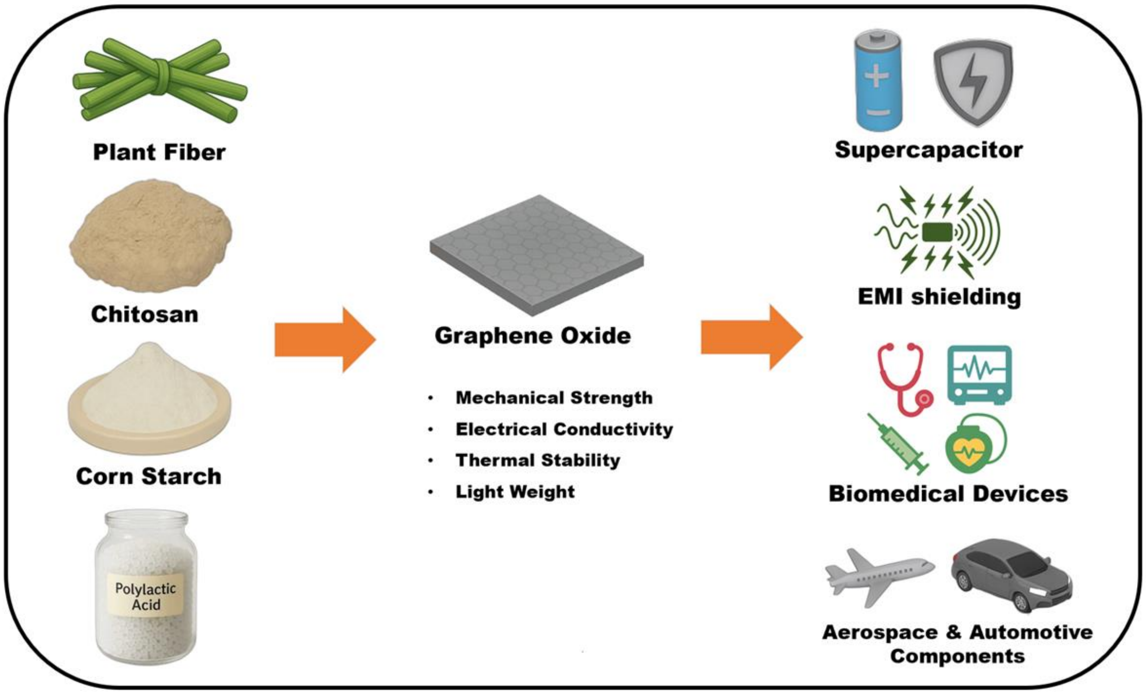

Figure 1. Schematic representation of graphene oxide bio-based polymer composites: sources, enhanced properties, and major application areas.

the desirable attributes of eco-friendly polymer matrices with the multifunctionality of graphene. These composites leverage the exceptional characteristics of graphene, such as high conductivity, strength, and thermal stability, to develop multifunctional materials suitable for diverse applications. Recent studies have highlighted their use in energy storage, electromagnetic interference shielding, aerospace, and biomedical applications. For instance, green-synthesized graphene composites show great promise in supercapacitors due to their improved charge storage capacity and durability, aligning with the demands of eco-friendly technologies [23]. Similarly, conductive polymer–graphene composites have demonstrated effective electromagnetic shielding capabilities, addressing radiation pollution with lightweight and sustainable materials [24]. Moreover, the advent of precise fabrication methods, particularly 3D printing and layer-by-layer assembly, has enabled customizable architectures suitable for next-generation electronics, optoelectronics, and sustainable packaging [25, 26]. A comprehensive schematic of the material sources, functional improvements, and industrial uses of green graphene-based polymer composites is presented in Figure 1, providing a visual summary of the concepts discussed in this review.

Despite these advances, critical challenges remain, and Further research is required to optimize their properties and scalability for commercial applications, ensuring they meet both environmental and technological demands. This comprehensive review summarizes recent progress in green polymer matrices and natural reinforcement systems, highlights the pivotal role of graphene and its derivatives in enhancing green composite performance, and explores their diverse industrial and environmental applications. Finally, it discusses the current limitations and future research opportunities towards scalable, high-performance, and environmentally friendly composite technologies.

2. Popular categories of green graphene-based nanocomposites

2.1. Green graphene-based nanocomposites on chitosan basis

Chitosan (CS), a natural biopolymer primarily derived from crustacean shells, is recognized for its exceptional biocompatibility, biodegradability, and chemical versatility [27]. These properties, combined with its abundance and low cost, have made chitosan a valuable component in the development of green nanocomposites. When reinforced with materials such as keratin and graphene derivatives, chitosan-based composites exhibit enhanced mechanical and functional properties, making them attractive for sustainable environmental and biomedical applications [12, 28].

Due to the presence of active amino and hydroxyl functional groups, chitosan is particularly effective in adsorbing a wide range of chemical species, including heavy metals. This makes it a promising candidate for environmental remediation. To improve its adsorption capacity, solubility, and mechanical strength, chitosan has been modified with nanomaterials such as graphene oxide (GO) and metal oxide nanoparticles [17]. GO, with its high surface area and oxygen-containing groups, enhances metal ion binding, while metal oxides like MnO2, Al_2O_3, and SiO_2 offer additional active sites for adsorption [17].

For example, Naicker et al. reported that incorporating metal oxides into magnetic chitosan chloride–graphene oxide (MCSCl-GO) composites significantly enhanced the removal of Cr(VI) ions from aqueous solutions through electrostatic and ionic interactions. Among these, MCSCl–GO–MnO_2 showed the highest efficiency due to its strong affinity and spontaneous interaction with Cr(VI) [17]. Similarly, Peryasamy et al. developed a hydrotalcite-modified GO-Chitosan composite (n-GO@HTCS) capable of removing chromium via a combination of electrostatic adsorption, surface complexation, and ion exchange. Notably, this composite could be regenerated and reused for up to five cycles [29]. Sherlala et al. designed chitosan–magnetic graphene oxide (CMGO) nanocomposites for arsenic adsorption, achieving a high specific surface area (152.38 m²/g) and superparamagnetic behavior (49.30 emu/g), facilitating magnetic separation and efficient chemisorption of arsenic [30].

Beyond metal ion remediation, chitosan-GO composites have been employed in wastewater treatment. Chang et al. synthesized a chitosan/polyacrylate/GO hydrogel via a semi-dissolution/acidification/sol-gel transition (SD-A-SGT) method, which effectively adsorbed both anionic and cationic dyes [31]. Zhang et al. developed a core–shell composite using modified chitosan and alginate as the shell and tungsten oxide as the core for uranium removal, enhancing chelation capacity via radical-mediated complexation [32]. In another study, Jeyaseelan et al. introduced hybrid composites containing pectin, chitosan, and lanthanide or alkaline earth elements for fluoride removal across a wide pH range, demonstrating reusability up to six cycles [33].

Despite these advancements, chitosan's instability in aqueous environments and relatively weak mechanical properties pose challenges for industrial applications. Strategies to address these limitations include functional group modification, crosslinking, and the incorporation of reinforcing fillers. Rozova et al. fabricated elastic chitosan/GO composites with improved strength and modulus due to homogeneous dispersion and strong interfacial bonding. Additionally, GO incorporation increased the free volume of the matrix, enhancing both mechanical integrity and adsorption performance in aqueous systems, where pure chitosan films failed [34].

Hu et al. created lightweight, compressible, and hydrophobic aerogels using a chitosan matrix reinforced with reduced GO nanosheets and modified with hydrophobic silicon/polydimethylsiloxane particles. These aerogels demonstrated exceptional oil absorption (18–45 g/g), chemical and thermal stability, and reusability under extreme environmental conditions, positioning them as promising candidates for oil spill cleanup [27].

Chitosan's biocompatibility and antimicrobial activity also make it a compelling material for medical uses such as wound dressings, anticoagulants, membranes, and scaffolds for tissue engineering. However, extensive hydrogen bonding among its functional groups limits solubility, stability, and mechanical strength, particularly in physiological conditions. To overcome these issues, reinforcements such as cellulose nanocrystals, carbon nanotubes, calcium phosphates, and GO have been employed. For instance, Yang et al. showed that adding 1 wt% GO to chitosan increased tensile strength by 122% and elastic modulus by 64%, while Gea et al. reported a 200% increase in modulus with 10 wt% GO. Zhou et al. demonstrated that 10 wt% GO not only improved the thermal stability and mechanical properties of chitosan but also enhanced cell adhesion, proliferation, and viability of murine mesenchymal stem cells [18, 35].

Other studies have extended these findings. Ruiz et al. examined PVA/CS/GO films, reporting enhanced mechanical strength and antibacterial activity due to GO-bacteria interactions [35]. Tavakkoli et al. optimized Cs/GO composites for orthopaedic applications,

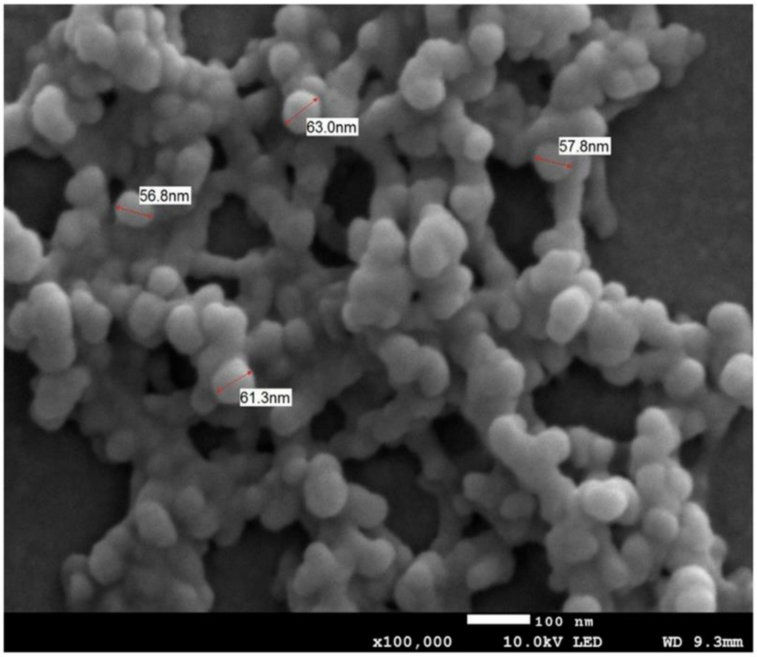

Figure 2. *B. balsamifera*-CNPs view from SEM. Reprinted with permission from [38]. 2025, MDPI.

Table 2. Summary of green chitosan–graphene nanocomposites and their key applications

Study	Composite Composition	Target/Application	Key Outcomes
Naicker et al. [17]	MCSCl–GO with MnO_2, Al_2O_3, SiO_2	Cr(VI) adsorption	MnO_2 composite showed the highest affinity and spontaneous uptake
Peryasamy et al. [29]	n-GO@HTCS (GO + hydrotalcite + CS)	Chromium removal	Spontaneous, endothermic adsorption; reusable up to 5 cycles
Sherlala et al. [30]	CMGO (Chitosan–Magnetic Graphene Oxide)	Arsenic adsorption	High surface area and magnetic separation; chemisorption confirmed
Chang et al. [31]	CS/Polyacrylate/GO hydrogel	Dye wastewater treatment	Adsorbs both anionic and cationic dyes
Zhang et al. [32]	Core–shell (CS/Alginate shell, WO_3 core)	U(VI) removal	Enhanced complexation and mechanical flexibility
Jeyaseelan et al. [33]	PCML/PCMC (Pectin + CS + Mg + La/Ce)	Fluoride removal	Effective in a wide pH range; reusable up to 6 cycles
Rozova et al. [34]	CS/GO composite films	Mechanical enhancement in aqueous systems	Stronger films, improved adsorption, and water stability
Hu et al. [27]	CS + rGO + hydrophobic Si/PDMS	Oil spill remediation	WCA = 148°, high oil uptake, robust under harsh conditions
Yang et al., Gea et al. [18]	CS + 1–10 wt% GO	Mechanical reinforcement	Up to 200% modulus and 122% tensile strength increase
Zhou et al. [18, 35]	CS/GO scaffold	Bone tissue engineering	Enhanced thermal stability, cell adhesion, and proliferation
Tavakkoli et al. [18, 36]	CS/GO nanocomposites in PMMA bone cement	Orthopedic application	Optimal at 0.3% GO, improved bioactivity, and mechanical properties
Sivashankari et al. [37]	ACGO (Agarose/Chitosan/GO) scaffolds	Biomedical scaffold	Biocompatible, hemocompatible, enhanced water retention, and cell attachment
Villarta et al. [38]	CNPs synthesized with B. balsamifera extract	Antibacterial application	MIC = 25 µg/mL against E. coli, eco-friendly nanoparticle synthesis

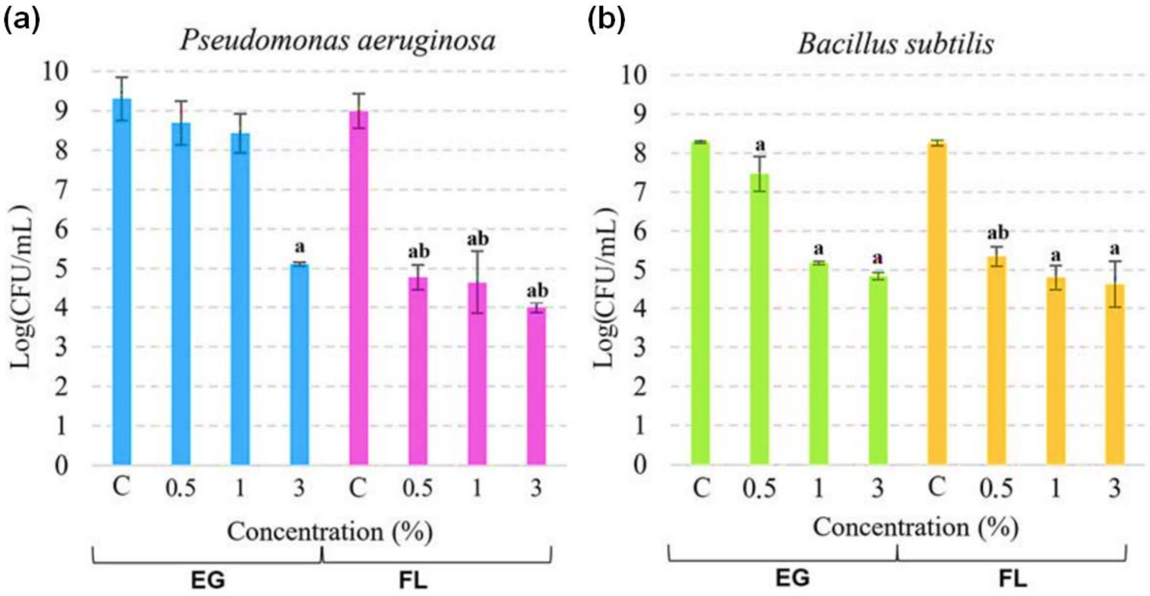

Figure 3. The viability of P. aeruginosa (a) and B. subtilis (b) under the action of EG or Flovan (FL). In the control experiment (depicted as C). Significant deviations from the control are denoted by "a" ($p < 0.05$), while values marked by "b" are significantly different from the samples of the equivalent concentration, but different additives ($p < 0.05$). Reprinted with permission from [41]. 2025, MDPI.

identifying a 2 wt% CS and 0.3 wt% GO formulation as optimal. These films exhibited notable bioactivity, strength, and degradability. When incorporated into PMMA bone cement, 25 wt% Cs/GO enhanced injectability, mechanical properties, and cellular response [18, 36]. Similarly, Sivashankari et al. developed agarose/chitosan/GO scaffolds with tunable porosity and demonstrated that GO content enhanced swelling, biodegradability, and cell proliferation while maintaining hemocompatibility [37].

Finally, Villarta et al. introduced a green synthesis route for chitosan nanoparticles using *Blumea balsamifera* leaf extract. The phenolic-rich extract acted both as a reducing and stabilizing agent, resulting in the formation of spherical nanoparticles with sizes ranging from 56.8 to 63.0 nm and effective antibacterial activity against *E. coli* at a concentration of 25 µg/mL [38]. As illustrated in Figure 2, DLS and SEM analyses confirmed the particle morphology and size distribution, supporting the efficacy of this eco-friendly synthesis method. This sustainable approach underscores the potential of chitosan-based nanocomposites for future pharmaceutical and therapeutic applications [38]. Table 2 summarizes the chitosan-graphene green nanocomposites and their key applications investigated in this study.

2.2. Green graphene-based nanocomposites derived from starch

Starch, as a renewable, abundant, and inexpensive biopolymer, has drawn increasing interest for use in sustainable packaging. However, its native form exhibits significant limitations such as brittleness, poor water resistance, and low processability. To mitigate these shortcomings, plasticizers such as glycerol, sorbitol, ethylene glycol, and urea are commonly employed to disrupt the crystalline structure of starch through gelatinization, resulting in thermoplastic starch (TPS) with improved processability [15]. For example, the glass transition temperature of starch decreases from 87 °C to 25 °C and 35 °C upon incorporation of 30% glycerol and sorbitol, respectively. Similar to native starch, thermoplastic starch (TPS) also suffers from several drawbacks, including water sensitivity, limited thermal stability, brittleness over time due to plasticizer loss, recrystallization, and low mechanical strength. Therefore, extensive efforts have been made to improve its properties by blending with other polymers and incorporating organic nanoparticles—approaches that do not compromise its biodegradability [15].

To enhance its properties while preserving biodegradability, researchers have incorporated

Table 3. Summary of starch-based green nanocomposites and their key applications

Study	Composite Composition	Target/Application	Key Outcomes
Ramazani et al. [15]	TPS + CNF + GO	Mechanical enhancement and barrier properties	66% ↑ tensile strength, 1435% ↑ modulus, 40% ↓ light transmittance, UV blocking
Solati et al. [39]	TPS/PLA + 1–3 wt% Graphene	Compatibility, crystallinity, and biodegradability	1% graphene improved miscibility; >1% caused phase separation and TPS droplets
Rabiei et al. [40]	Starch + MIL-100 + $CoFe_2O_4$	Photocatalytic degradation of tetracycline and dyes	Effective degradation at room temperature
Vasiliauskienė et al. [41]	Corn starch + EG/Flovan flame retardants	Antibacterial biocomposites, microbial modulation	>99.7% Proteobacteria with retardants; up to 5-log ↓ in bacterial viability

nanostructured fillers, particularly cellulose nanofibers (CNF) and graphene-based materials. Ramazani et al. examined the synergistic reinforcement of TPS with CNF and graphene oxide (GO). Their results indicated that the GO nanosheets altered the matrix morphology into a layered structure, limiting molecular mobility and reducing crystallinity through suppressed retrogradation. Mechanical testing revealed a 66% increase in tensile strength and a 527% increase in elastic modulus for TPS/CNF composites compared to neat films. With the addition of 3 wt% GO, these values rose dramatically—fracture strength increased by 440% and elastic modulus by 1435%, though elongation at break decreased by 110%. Furthermore, optical and barrier analyses showed a 40% reduction in visible light transmittance and a 30% reduction in water vapor transmission, with complete UV light blockage [15].

In another study, Solati et al. evaluated the influence of graphene nanosheets on TPS/polylactic acid (PLA) blends. The inclusion of 1 wt% graphene improved interfacial compatibility, as evidenced by closer glass transition temperatures and a more uniform surface. However, at higher graphene concentrations (2–3 wt%), phase separation and TPS droplet formation occurred. Graphene addition also modified crystallization behavior, eliminating cold crystallization and raising the crystallization temperature due to graphene-induced nucleation. All graphene-containing blends exhibited greater crystallinity than their pristine counterparts [39].

Expanding beyond mechanical and thermal enhancements, Rabiei et al. synthesized a starch-based magnetic nanocomposite incorporating MIL-100 (a metal–organic framework) and cobalt ferrite. The resulting hybrid demonstrated effective photocatalytic activity for the degradation of tetracycline and dyes under ambient conditions, positioning starch as a viable platform for environmental remediation [40].

In a microbiological context, Asiliauskienė et al. explored the effects of flame retardants on the bacterial ecology of starch-based biocomposites. Their composites, composed of hemp shives, corn starch, and either expandable graphite (EG) or a Flovan compound, were analyzed after 12 months of incubation. Next Generation Sequencing (NGS) revealed that flame retardants drastically shifted microbial populations: in treated samples, Proteobacteria dominated (>99.7%), whereas untreated composites displayed higher diversity, including Bacteroidetes, Actinobacteria, and Saccharibacteria. Viability assays on *Bacillus subtilis* and *Pseudomonas aeruginosa* showed a significant antibacterial effect from both additives, with bacterial counts dropping up to five logarithmic units. As illustrated in Figure 3, statistical differences between the flame retardants further underscore their distinct microbial impacts [41].

Collectively, these studies demonstrate the multifunctionality and adaptability of starch-based nanocomposites when reinforced with graphene and related materials. These enhancements broaden starch's applicability not only in packaging and structural materials but also in biomedical, environmental, and antimicrobial domains. Table 3 summarizes the starch-based green nanocomposites and their key applications investigated in this study.

2.3. *Green graphene-based nanocomposites derived from polylactic acid*

Polylactic acid (PLA) is a biodegradable aliphatic polyester synthesized from renewable agricultural sources such as corn and sugarcane. As a thermoplastic polymer with high strength and modulus, PLA exhibits relatively low permeability to water, oxygen, and carbon dioxide. Its biodegradability and recyclability into organic compost, combined with its production from carbon dioxide-consuming feedstocks, make PLA an environmentally attractive material. These characteristics have facilitated its extensive applications in medicine, pharmaceuticals, packaging, and agriculture [39].

However, PLA's use is limited by several drawbacks, including low thermal resistance, poor flame retardancy, low impact strength, limited processability, high cost, and low crystallinity, which collectively diminish its mechanical properties such as stability, modulus, and strength. To address these limitations, additives such as plasticizers, lubricants, and fillers, or blending with other polymers, have been employed. For example, the glass transition temperature (T_g) and melting temperature (T_m) of PLA typically range from 50 to 70 °C and 130 to 180 °C, respectively, which are lower than those of common petroleum-based plastics like polyethylene terephthalate (PET) and polystyrene (PS), resulting in reduced energy consumption during processing [39]. Despite this advantage, the relatively slow degradation rate of PLA remains a concern, as it may require several hours or longer to decompose depending on molecular weight and morphology [39].

Blending PLA with thermoplastic starch (TPS) has been extensively studied to lower costs, improve degradability, and reduce TPS's water sensitivity. Martin et al. investigated PLA/TPS composites with varying toughness and plasticizer content, reporting a 112% increase in elongation at break for TPS with the highest plasticizer content; however, incorporation of TPS significantly reduced mechanical strength in the blends [39].

In recent years, the incorporation of graphene-based nanomaterials has gained considerable attention for enhancing PLA properties. Norazlina et al. demonstrated improved mechanical performance of PLA/graphene nanocomposites compared to neat PLA and PLA composites with conventional graphite [39].

Functionalized graphene oxide (f-GO), modified with maleic anhydride and dodecylamine, was shown by Wang et al to improve morphology, thermal stability, crystallization behavior, weathering resistance, and protective properties of PLA-starch matrices. The addition of f-GO enhanced UV protection, surface hydrophobicity, erosion resistance, and induced heterogeneous nucleation, leading to spherical crystal growth during isothermal crystallization. Moreover, it increased matrix thermal stability and storage modulus, making these nanocomposites promising for food and pharmaceutical packaging applications [42].

Scaffaro et al. explored the effect of graphene nanoplatelet (GNP) and pineapple fiber (PF) ratios on mechanical properties and hydrolytic degradation kinetics of PLA-based composites under acidic, neutral, and alkaline conditions. They found that hybrid fillers were well dispersed within the PLA matrix, and mechanical tests revealed increased stiffness proportional to GNP content. Degradation behavior depended on GNP loading: low GNP content accelerated degradation due to interface discontinuities, while higher GNP content created a barrier effect from hydrophobic nanocarbons, partially offsetting the hydrophilicity of lignocellulosic fillers and slowing degradation kinetics, though still faster than neat PLA [43].

Wang et al. also studied the synergistic effect of titanium dioxide (TiO_2) nanoparticles and GO nanosheets in PLA-starch composites. The combined addition significantly accelerated heterogeneous nucleation and crystal growth during isothermal crystallization more effectively than individual fillers. Strong interfacial interactions and synergism between fillers enhanced morphology, storage modulus, thermal stability, surface hydrophobicity, UV shielding, and resistance to ageing-related property changes in PLA-MST-TiO_2-f-GO nanocomposites compared to those with single fillers [44].

In another approach, Ariturk et al. developed green PLA composites by incorporating waste cellulose fibers and vermiculite as natural reinforcements. Their results indicated that this combination improves mechanical properties and environmental compatibility, offering promising bio-based alternatives to petroleum-derived polymers [16].

Batakliev et al. investigated biodegradable blends of PLA and polycaprolactone (PCL) at various weight ratios to achieve materials with desirable mechanical and thermal shape memory properties. Blends with higher PCL content showed increased flexibility, toughness, and elongation at break (up to ~550% for 30PLA/70PCL), while tensile strength and modulus decreased. Shape memory tests revealed excellent shape fixity (~98%) and recovery ratios improving with PLA content, with faster recovery at higher temperatures (70 ˚C). The co-continuous morphology and uniform

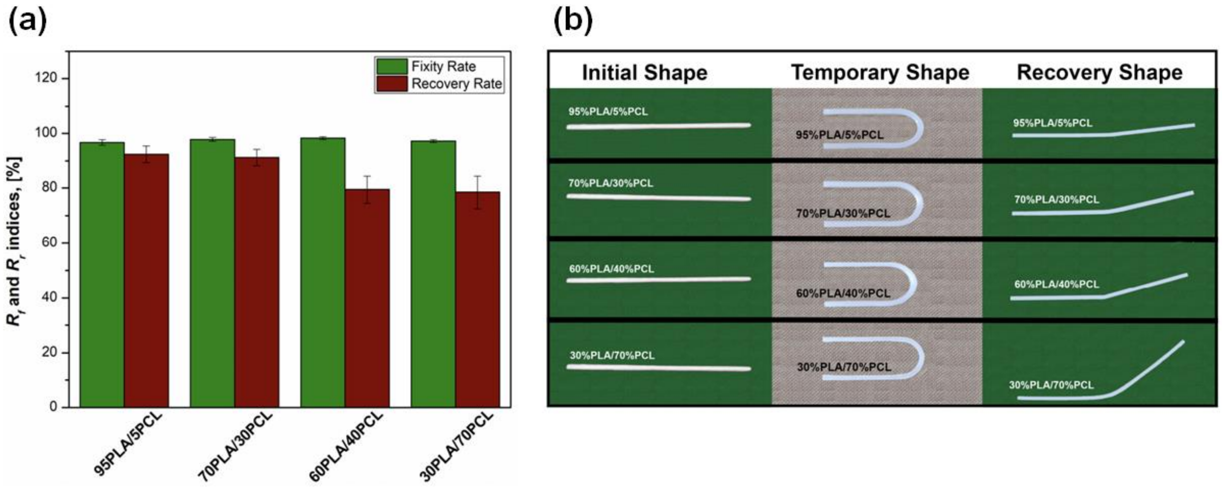

Figure 4. Optical images and comparative diagrams of shape fixation and shape recovery rate evaluation of PLA/PCL composite blend. Reprinted with permission from [19]. 2025, MDPI.

dispersion of the dispersed phase (~500 nm) without compatibilizers contributed to balanced mechanical behavior. Sample images during the shape memory cycle and comparative charts are presented in Figure 4.

Finally, Barbosa et al. examined surface treatments of 3D-printed electrodes fabricated from a PLA/carbon black filament. Among electrochemical activation, acid (HNO_3), alkaline (NaOH), and solvent (DMF) treatments, alkaline treatment notably enhanced electrochemical performance by increasing electroactive surface area and reducing charge transfer resistance through partial removal of insulating PLA. The optimized electrode (3D-SBasic) exhibited superior sensitivity and resolution for acetaminophen detection compared to commercial and conventional electrodes, as demonstrated in Figure 5 [45]. Table 4 summarizes the Green Graphene-Based PLA and their key applications investigated in this study.

2.4. Green graphene nanocomposites based on cellulose

Cellulose is one of the most abundant natural biomolecules and serves as a primary structural component of the cell walls in most plants. It can also be sourced from marine organisms, algae, fungi,

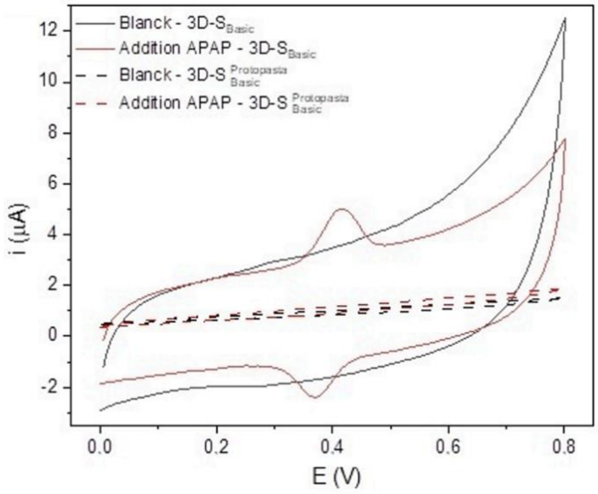

Figure 5. Cyclic voltammograms for 3D-Sbasic (solid line) and 3D-SbasicProtopasta (dashed line) in the presence and absence of APAP. Reprinted with permission from [45]. 2025, MDPI.

Table 4. Summary of studies on green graphene-based PLA nanocomposites and their key applications

Study	Composite Composition	Target/Application	Key Outcomes
Martin et al. [39]	PLA + Thermoplastic starch (TPS, with varying plasticizer content)	Improve degradability, reduce cost, and water sensitivity	↑ Elongation at break (112%); ↓ Mechanical properties when TPS added
Norazlina et al. [39]	PLA + Modified graphene	Mechanical reinforcement	↑ Mechanical properties vs. neat PLA or PLA/graphite composites
Wang et al. [42]	PLA + Functionalized GO (maleic anhydride + dodecylamine)	Packaging (UV protection, stability, crystallinity)	↑ UV shielding, hydrophobicity, modulus, and crystallinity
Scaffaro et al. [43]	PLA + PF + GNP (various ratios)	Mechanical enhancement and hydrolytic degradation	↑ Stiffness; degradation rate dependent on GNP loading
Wang et al. [44]	PLA + Starch + TiO_2 + GO	Synergistic enhancement of properties	↑ Crystallinity, storage modulus, thermal stability, and aging resistance
Ariturk et al. [16]	PLA + Waste cellulose + Vermiculite	Bio-based mechanical reinforcement	↑ Mechanical properties, eco-friendliness
Batakliev et al. [19]	PLA/PCL blends (various ratios)	Mechanical tuning + Shape memory	↑ Elongation (up to ~550%), ↑ Fixity (~98%), ↑ Recovery (at 70 °C), balanced morphology (Figure 2)
Barbosa et al. [45]	PLA + Carbon black (3D-printed electrode, various surface treatments)	Electrochemical sensing (APAP detection)	NaOH treatment → ↑ Surface area, ↓ Resistance, ↑ Sensitivity (Figure 6)

invertebrates, and bacteria, as well as from wood, hemp, cotton, and bast fibers. Figure 6 illustrates the various sources and the chemical structure of cellulose. Morphologically, cellulose features a one-dimensional fibrous architecture with a high aspect ratio; however, its specific structural characteristics vary slightly depending on its origin and the applied processing techniques. Since its discovery, considerable research has focused on the chemical composition and physical structure of cellulose. It is a polydisperse polymer consisting of D-glucopyranose units connected via β-1,4-glycosidic bonds, forming long chains of thousands of monomers. Intrachain hydrogen bonds between hydroxyl groups and ring oxygen atoms contribute to the linear configuration of cellulose chains [46].

Cellulose-based green composites have garnered increasing attention due to their environmental compatibility and stable physicochemical properties. As the most abundant natural polymer, cellulose provides a renewable matrix with desirable mechanical and functional characteristics for composite development

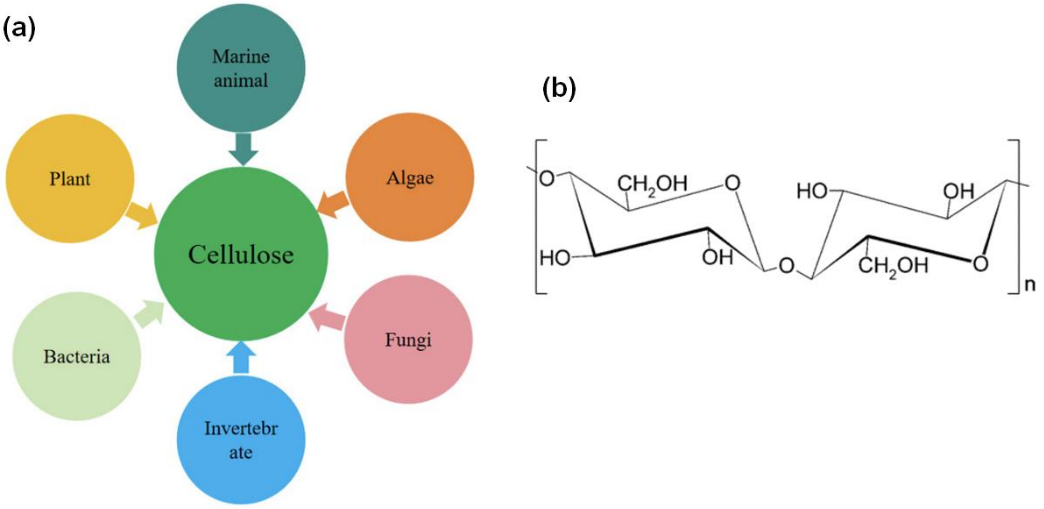

Figure 6. Major sources of cellulose & Chemical structure of cellulose. Reprinted with permission from [41]. 2025, MDPI.

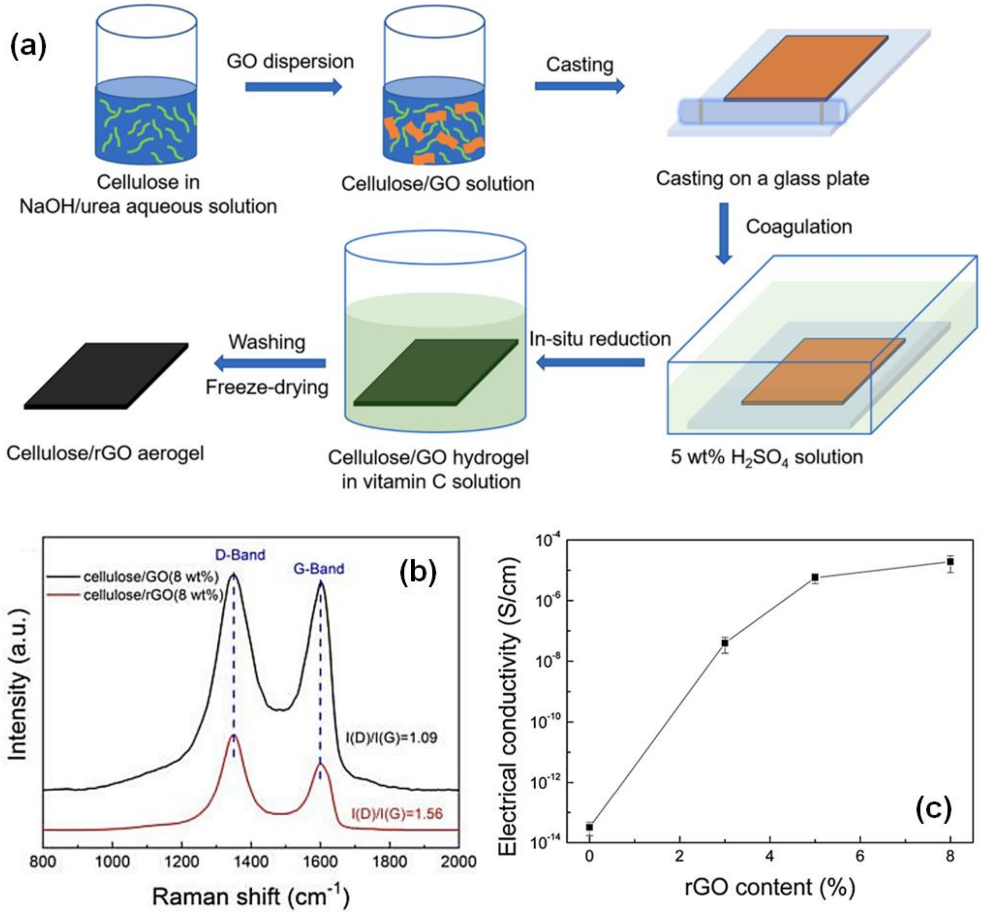

Figure 7. (a) Schematic representation of the preparation of cellulose/rGO composite aerogels. (b) Raman spectra and the D/G band intensity ratio of cellulose/GO (8 wt%) (top) and cellulose/rGO (8 wt%) composite aerogels (bottom); and (c) electrical conductivity of cellulose/rGO aerogels as a function of rGO content. Reprinted with permission from [52]. 2025, MDPI.

[47]. Its incorporation into green composites has been shown to improve mechanical strength and thermal stability [48], as well as enhance water resistance and biodegradability, offering significant advantages for sustainable material design [1]. These composites present a viable solution to mitigate plastic waste derived from petroleum sources and support environmental sustainability initiatives [49]. Moreover, the inclusion of cellulose can positively influence the structural and functional properties of polymer matrices, expanding their application potential [50].

Despite its advantages, the high hydrophilicity of cellulose fibers poses challenges for compatibility with hydrophobic polymer matrices and can lead to reduced mechanical performance due to moisture absorption. To address this issue, various surface modification strategies, such as physical treatments and chemical grafting, have been developed to reduce fiber hydrophilicity and enhance interfacial bonding with the matrix [5].

In a notable application, Snari et al. introduced a fluorometric sensor based on cellulosic nanomaterials functionalized with Morin [2-(2,4-dihydroxyphenyl)-3,5,7-trihydroxychromen-4-one], a natural ligand capable of forming strong luminescent complexes with Al^{3+} ions. This sensor shows promise for quality control in detecting aluminium ion contamination in herbal tea production, as well as for qualitative detection of Al^{3+} in blood serum samples [51].

Cellulose also plays a role in the preparation of graphene oxide (GO)-based suspensions, where it may be added before or after GO reduction to enhance dispersion stability. When introduced prior to reduction, cellulose facilitates GO adsorption onto its surface, effectively preventing nanosheet aggregation and promoting a homogeneous distribution. This stabilization is typically attributed to a confinement effect, in which the cellulose framework limits GO sheet mobility. However, the stabilization efficiency is highly dependent on the cellulose-to-GO ratio. Insufficient cellulose may fail to prevent agglomeration, while excessive cellulose content can diminish the functional benefits of GO, including its electrical and mechanical performance [52].

In a study by Chen et al., cellulose/reduced graphene oxide (rGO) aerogels were fabricated as chemical vapor sensors. As shown in Figure 7a, the aerogels were prepared by dissolving cellulose and dispersing GO in a NaOH/urea aqueous solution, followed by in-situ chemical reduction and freeze-drying. Figure 7b presents the Raman spectra of the composites containing 8 wt% filler, revealing an I(D)/I(G) ratio of 1.56 for rGO and 1.09 for GO. Although this increase in I(D)/I(G) appears counterintuitive, since reduction typically decreases sp^3-type defects, such behavior is often attributed to the formation of smaller sp^2 domains and a higher density of edge defects, enhancing the D band. Ascorbic acid, the reducing agent used in this study, enabled sufficient GO reduction to restore electrical conductivity in rGO, making the composite suitable for piezoresistive sensing applications. As illustrated in Figure 7c, conductivity increased markedly with rGO

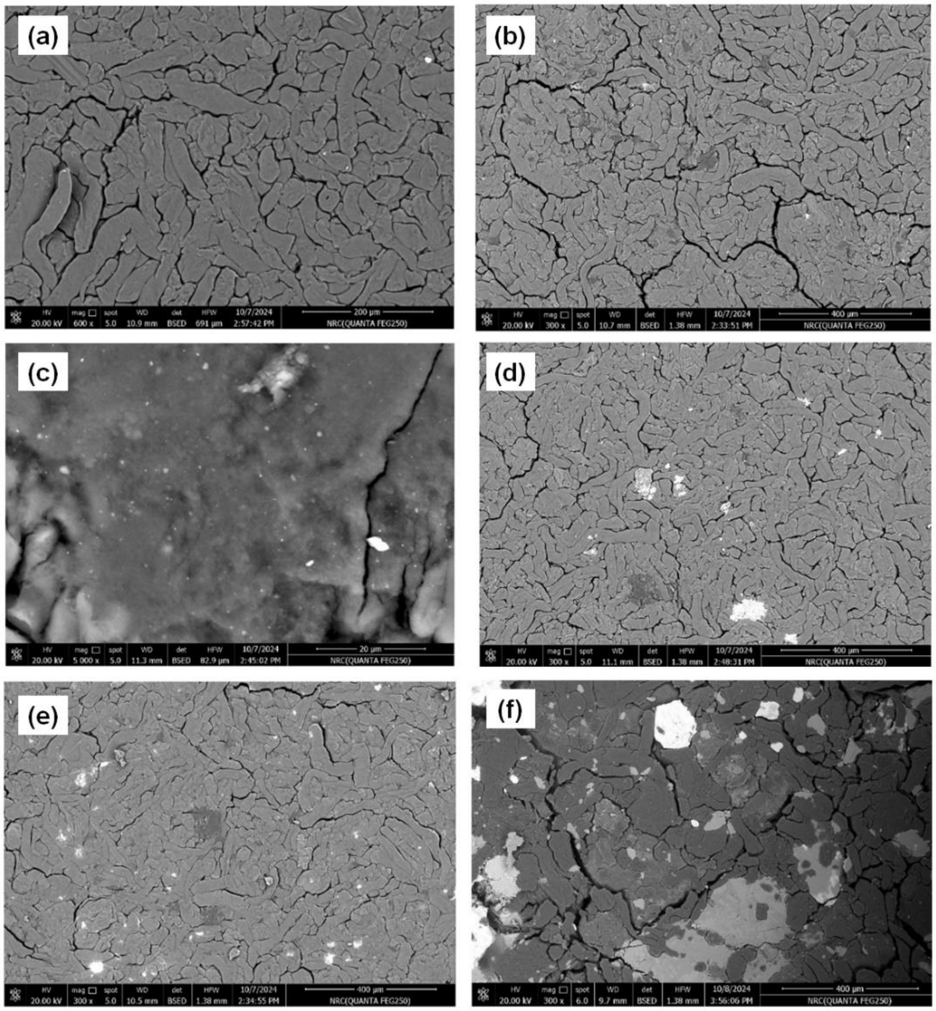

Figure 8. SEM images of (a) CMC (scale: 400 µm), (b) CMC/0.4% G (scale: 20 µm), (c) CMC/0.8% G (scale: 400 µm), (d) CMC/1% G (scale: 400 µm), (e) CMC/2% G (scale: 400 µm), and (f) CMC/3% G (scale: 400 µm). Reprinted with permission from [53]. 2025, MDPI.

Table 5. Summary of studies on green graphene-based cellulose nanocomposites and their key applications

Study	Composite Composition	Target/Application	Key Outcomes
Snari et al. [51]	Cellulose + Morin (natural ligand)	Fluorometric sensor for Al^{3+} detection	High luminescence with Al^{3+}; applicable in herbal tea QC and serum analysis
Chen et al. [52]	Cellulose/rGO aerogel	Chemical vapor sensor	Conductivity up to 1.9×10^{-5} S/cm at 8 wt% rGO; I(D)/I(G) ↑ to 1.56; good piezoresistive response
Aldaleeli et al. [53]	Sodium carboxymethyl cellulose (CMC) + 0.2–3 wt% Graphene	UV-protective biodegradable packaging	↓ UV transmittance, ↓ bandgap (5.27 → 4.81 eV), ↑ Urbach energy (0.34 → 0.94 eV), improved morphology

loading, reaching 1.9×10^{-5} S/cm at 8 wt%, surpassing the percolation threshold at just 3 wt% [52].

In another study, Aldaleeli et al. explored the influence of graphene loading (0.2 to 3 wt%) on the structural and optical properties of sodium carboxymethyl cellulose (CMC)-based films. Their findings showed that increasing graphene content modified surface morphology (Figure 8), reduced particle size, and enhanced optical properties. Notably, light transmittance in UV-C, UV-B, and UV-A regions decreased with higher graphene loading, while the optical bandgap narrowed from 5.27 to 4.81 eV. Conversely, Urbach energy increased from 0.34 to 0.94 eV, indicating enhanced disorder. These features render the nanocomposites promising for biodegradable, UV-protective food packaging applications [53]. Table 5 summarizes the Green Graphene-Based PLA and their key applications investigated in this study.

3. Applications of green composites

Green composites, derived from environmentally friendly materials, have emerged as viable alternatives to conventional materials across various applications, offering comparable or superior performance alongside notable environmental benefits. In energy storage, for example, supercapacitors fabricated with green composites such as graphite foil and sodium acetate have outperformed traditional systems that often rely on hazardous substances like fluorine and sulfur, enhancing both efficiency and safety [13]. In biomedical and packaging applications, bioactive films incorporating chitin nanocrystals stand out due to their excellent biocompatibility, biodegradability, and inherent antibacterial properties [3]. The automotive industry has also witnessed a shift toward sustainable alternatives, with hybrid composite materials, comprising components such as basalt, shell powder, and alumina, being employed in brake linings. These materials demonstrate reduced wear and lower water absorption compared to conventional asbestos-based counterparts, which pose significant environmental and health risks [14]. Additionally, advancements in casting technologies, including the use of low-temperature molds and non-stick coatings, have contributed to more sustainable aluminium alloy production by minimizing harmful emissions and enhancing casting quality [54]. In additive manufacturing, polylactic acid (PLA) reinforced with molybdenum disulfide (MoS_2) shows promise as a renewable, carbon-free substitute for petroleum-based resins, although further improvement in mechanical properties is necessary. Meanwhile, the polymer industry is actively developing biopolymers such as starch, cellulose, and chitosan to replace conventional synthetic polymers, thus addressing concerns related to long-term environmental degradation [55]. Collectively, these developments highlight the significant potential of green composites to advance sustainability without compromising on functionality or performance.

4. Challenges and opportunities

The development of green composites faces several important challenges alongside promising opportunities. A primary issue lies in ensuring effective compatibility between natural fibers and polymer matrices, as inadequate interfacial bonding can compromise mechanical performance. Improving this aspect is key to advancing their structural applications. Additionally, enhancing durability and resistance to environmental factors such as moisture, UV exposure, and temperature fluctuations is essential for extending their usability in long-term and outdoor settings.

Equally important is the implementation of comprehensive life cycle assessments to evaluate and validate the environmental benefits of green composites in comparison to conventional materials. Tackling these challenges opens up opportunities for innovation, improved performance, and greater integration of sustainable materials across various industries.

5. Conclusions and future outlook

Future progress in the field of green composites is closely tied to advancements in material science and manufacturing technologies. One promising direction is the integration of nanotechnology, specifically, the use of nanomaterials such as nanocellulose and nanoclays, which can significantly improve the mechanical strength, thermal stability, and barrier properties of these composites. Additionally, the adoption of biodegradable polymer matrices derived from renewable sources, such as polylactic acid (PLA) and polyhydroxyalkanoates (PHA), enhances the environmental sustainability of the final products, making them suitable alternatives to petroleum-based plastics. To enable widespread industrial adoption, it is equally important to optimize processing techniques. Methods like extrusion, compression molding, and injection molding must be refined for better scalability, cost-efficiency, and material consistency. Moreover, research into multifunctional green composites, those combining structural performance with properties like antimicrobial activity or self-healing, could further expand their applicability in fields such as biomedical engineering, packaging, construction, and automotive manufacturing. As environmental regulations become stricter and consumer demand for sustainable products grows, the development of high-performance, eco-friendly composites is likely to become a central focus in both academia and industry.

Conflict of Interest

The authors declare no conflict of interest.

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