Review Article

Nilofar Asim*, Mohd Sukor Su’ait, Marzieh Badiei, Masita Mohammad, Md. Akhtaruzzaman, Armin Rajabi, Nowshad Amin, and Mariyam Jameelah Ghazali*

Perspectives in biopolymer/graphene-based composite application: Advances, challenges, and recommendations

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Abstract: Graphene-based materials are widely applied due to their interesting physical and chemical properties, but their hydrophobic surface and toxicity to living creatures limit their application in some fields. Biopolymers are incorporated with graphene-based materials to overcome these issues and improve their biodegradability, biocompatibility, and ecological friendliness, and the synergetic effect enhances other properties as well. These properties make graphene-based materials a novel subject of interest in science and industry. In this study, the various applications of developed biopolymer/graphene-based composites are broadly addressed, and recent progress in the field is emphasized. Modification, stability, and compatibility are among the key merits for developing highly advanced composites with desirable properties. The major challenges and some recommendations in various applications based on reviewed studies are covered. However, the development of environmentally friendly, low-cost, high-quality, and large-scale biopolymer/graphene-based composites for specified applications is challenging. Studies based on application and trend are conducted. Opportunities and limitations can guide researchers in the field to solve challenges, provide directions for future studies, and optimize sustainable biopolymer/graphene-based composites for specified industrial applications.

Keywords: biopolymer, graphene-based composites, modification, applications, sustainability

1 Introduction

Since the discovery of graphene, its various derivatives are developed and extensively investigated for different applications. Graphene-based nanocomposites in different forms, such as films, 3D porous scaffolds, fibrils, hydrogel, and nacre-like structures, have been developed and studied [1,2].
Given the huge potential of utilizing waste materials as precursors for the preparation of graphene derivatives, the dependence of their morphology, structure, and properties on conversion methods and type and composition of waste materials should be considered [3,4].

The invention of graphene and its potential for making various composites have opened up new interesting research areas to tailor new materials with chemical and physical properties designed on the basis of their bonding mechanism [5]. Biopolymers are materials derived from biological sources, such as animals, microorganisms, and plants. Biopolymers, as potential materials to combine with graphene, have attracted huge attention because of their abundance, renewability, biodegradability, low cost, and various applications [5]. Figure 1 shows the various available biopolymers for combination with graphene-based materials.

According to Rouf and Kokini [5], biopolymer–graphene composites are synthesized through (1) solution intercalation, (2) melt intercalation, and (3) in situ polymerization. Other methods such as wet spinning, electrospinning, freeze-drying, microwave synthesis, molecular beam epitaxy drop casting, and layer-by-layer assembly are also utilized by researchers to prepare biopolymer–graphene-based composites [6,7]. The preparation method has important influences on the prepared nanocomposites’ physical and chemical properties because of their effect on homogeneity (distribution), interaction, and structure [8]. Figure 2 depicts the various available interactions between graphene derivatives and biopolymers.

Interfacial interactions, including covalent and noncovalent bonding, improve the mechanical properties of graphene-based nanocomposites. These different kinds of interactions usually work synergistically together, resulting in excellent integrated mechanical properties. Noncovalent bonding, including hydrogen bonding, ionic bonding, and π–π interactions, usually shows relatively weak strength compared with covalent bonding [10]. However, the mechanical properties of graphene-based nanocomposites have been enhanced via noncovalent bonding due to their easy construction with graphene nanosheets. Covalent bonding shows strong interfacial strength and can be tuned by adjusting the length of the molecule chain and the density of crosslinking. The use of small molecules as crosslinkers usually enhances strength and stiffness but results in relatively low toughness due to the restriction of sliding between adjacent graphene nanosheets. Recent investigations reveal that long-chain linear molecules are good for enhancing tensile strength and toughness. Compared with long-chain linear molecules, a polymer or macromolecules with active functional groups can achieve integrated strength, stiffness, and toughness due to their

Figure 1: Classification of biopolymers (reprinted with permission from refs [5,6]).
high density of covalent crosslinking [9]. Cellulose, the most abundant biopolymer on earth, is widely utilized for different applications. Despite the cheap and abundant resource for the preparation of cellulose from biomass, bacterial cellulose has attracted attention because the prepared cellulose does not attach to hemicellulose or lignin [11,12]. Bacterial cellulose can be utilized for various applications. Figure 3 shows the various preparation methods of graphene-based/bacterial cellulose nanocomposites. The flexible bacterial cellulose/graphene/polyaniline nanocomposite with excellent electrical conductivity is prepared via a facile two-step strategy and presents a high potential for application in electromagnetic shielding and flexible electrodes [13].

To overcome the utilization of the conventional chemical-based preparation method, Dhar et al. [14] prepared bacterial cellulose/graphene-based films via a single step in situ fermentation. The physicochemical and structural properties of the composite depend on reduced graphene oxide compositions and fermentation conditions. The excellent inherent properties of prepared composites suggest flexible electronic materials.

Luo et al. [15] developed a facile layer-by-layer assembly method to prepare bacterial cellulose/graphene oxide hydrogels with excellent mechanical properties. Hydrogels contain highly dispersed graphene oxide nanosheets bundled by 3D interconnected bacterial cellulose nanofibers. Considering that the properties of composites are highly related to the miscibility

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**Figure 2:** Common enabling interactions between graphene-based materials and biopolymers (reproduced with permission from refs. [9,10]).

**Figure 3:** Some utilized processing routes for the preparation of bacterial cellulose–graphene oxide nanocomposites [11].
and compatibility of the components, their modification can improve their properties dramatically.

2 Physical and chemical modifications of biopolymer, graphene-based materials, and their composites

The graphene derivatives’ properties can be modified for different applications by tailoring their structural changes, activation, and functionalization. Modification methods include covalent functionalization, noncovalent functionalization, substitutional doping of graphene, and hybridization with nanoparticles and nanowires [16]. Noncovalent modification can use π–π interactions, van der Waals forces, ionic interactions, and hydrogen bonding [17].

Various available functional groups, such as carboxyl, amino, or hydroxyl groups, on the biopolymer can establish effective interactions with functional groups on graphene derivatives and prepare new environmentally friendly composites with wide applications, which depend on their composition, interaction, and preparation conditions [5,6].

Xiong et al. [9] investigated structures, morphologies, functionalization, and processing applications of various bionanocomposites. Synergy effects are due to the combination of biopolymers with other materials, and their interfacial interactions and structures control their properties in different applications. Preparation methods have vital effects on the nanocomposites’ properties.

The biopolymer can be modified by grafting, crosslinking, blending, and coating [18,19]. The physical modification with or without the destruction of granular structure can modify some biopolymer properties [20].

The combination of the hierarchical structures of biopolymer nanofibrils, such as cellulose, chitin, silk, and collagen nanofibrils, and the unique strength and toughness of graphene derivatives by a simple preparation method are opening a new interesting research field to prepare new modified nanocomposites for specified applications [21].

The functionalization of available groups on biopolymer can provide wide opportunities for the preparation of tailored graphene-based nanocomposites. Cellulose has three active hydroxyl groups, including the primary hydroxyl of C-6 and the secondary hydroxyls of C-2 and C-3, which can undergo a series of modification reactions (including esterification, etherification, crosslinking, and graft copolymerization) [22]. These reactions produce cellulose derivatives with functional groups, such as silylates, sulfates, sulfonates, amines, hemiacetals, azides, and carbanilates. These functionalized cellulose derivatives find applications in different fields, such as medicine, agriculture, or textile. Moreover, many applications of chitosan have been limited by its low solubility in the aqueous solution. The efficient transformations of chitin and chitosan have also been limited by the lack of organosolubility due to its high molecular weight and interunit hydrogen bonding [23]. The chemical modification of these biopolymers through the introduction of different protecting groups at the hydroxyl and amino groups enhances good bioactivity properties and increases the solubility in organic and aqueous media (physiological conditions). The main reactions are acylation, alklylation, quaternization, sulfation, azidation, and phosphonation. Figure 4 presents various functionalization methods for nanocellulose and chitin or chitosan.

The addition of monosaccharides, disaccharides, and polysaccharides to graphene-based materials is considered a key modification approach for improving hydrophilicity and dispersibility in polar solvents and their biodegradability for biomedical and environmental applications [27]. Among the available various methods for the (poly)saccharide functionalization of graphene, click chemistry is considered one of the best methods [28]. Click chemistry is defined as numerous chemical reactions that have some important common characteristics, including high modularity, high chemical yield, no byproduct, and minimal sensitivity to oxygen, water, and solvent. Click chemistry enables the covalent attachment of functional groups with affinity to the π-conjugated surface of graphene. Accordingly, the modifications of graphene with polysaccharides either at its edges or at the basal plane become possible through different approaches. However, in some cases, other functionalization methods of graphene or graphene oxide are needed before click chemistry [27]. Therefore, various complementary methods to click chemistry that are typically performed on −COOH or −OH groups on the edge or epoxide groups on basal plane of graphene or graphene oxide are known. Oxygen-containing groups, including hydroxyl, carboxyl, and epoxy groups, on the surface of graphene oxide are used for common chemical reactions, such as esterification, isocyanation, carboxylic acylation, epoxy ring opening, diazotization, and addition [22]. The functionalization of carboxylic acid groups is performed through the addition of nucleophilic species, whereas the functionalization via epoxy groups is done...
through ring-opening reactions. Figure 5 presents other methods complementary to click chemistry. Li and Papa- 
dakis [27] reviewed various modi-
cfications of graphene-
based materials with (poly)saccharides through click 
chemistry. Figure 6 presents the three types of polysac-
charide/graphene modifications.

Functionalization and hybridization play important 
roles in dispersion, stabilization, reduction, and rein-
forcement of nanocellulose/graphene-based nanocom-
posites, which control their properties [29].

The interaction of graphene-based materials with biop-
olymers is due to available hydroxyl, epoxy, carbonyl, 
and carboxyl groups [30]. These interactions include 
covalent, electrostatic, hydrogen, and noncovalent bonds. 
Then, functionalization can modify graphene-based bio-
 polymer composites for specified applications [6,31,32]. 
The main categories of graphene-based biopolymer com-
posites include 3D porous scaffolds, electrospun mats, 
hydrogel, and nacre-like structures, which present dif-
ferent properties. However, preparation methods affect
the properties of prepared composites [1].

The vacuum-assisted self-assembly method is used 
to prepare casein phosphopeptide-biofunctionalized gra-
phone oxide nanoplatelets/cellulose nanofibers [33]. The 
prepared composites show high thermal conductivity and 
xcellent flame retardancy.

The microstructure-tunable cellulose nanofiber/ 
reduced graphene oxide aerogels developed via the con-
centration of cellulose nanofiber present lightweight and 
high-efficiency microwave absorption materials [34].

The functionalization of chitosan with sulfonated 
graphene oxide and polyvinyl alcohol results in poly-
mer electrolyte membrane with remarkable increment 
in electrochemical properties for high-temperature energy 
applications [35,36].

3 Applications of biopolymer/ 
graphene-based composites

Eco-friendly biopolymer/graphene-based composites with 
various physicochemical properties can be used for dif-
ferent applications. The following sections provide some 
references for other researchers utilizing biodegradable 
and biocompatible biopolymer/graphene-based com-
posite materials for various applications, which sometimes 
overlap.

3.1 Application in the construction sector

The construction industry has a huge influence on the 
global economic growth. Finding construction materials 
with high performance, low cost, and sustainability is 
the center of concern for researchers in this field. The 
Improvement of performance such as strength [37], bar-
rier and permeability properties [38,39], leaching [40], 
and durability [41] by the addition of graphene-based 
materials to cement has been studied. Despite wide studies 
on utilizing biopolymers and graphene-based materials 
in cement, the addition of biopolymer/graphene-based 
composite materials into cement is rarely studied [42].

Figure 4: (a) General paths for the modification of nanocellulose (reproduced from refs. [24,25]). (b) Regioselective and chemoselective modifications of chitosan [26].
Figure 5: Different methods for the functionalization of graphene (upper part) and graphene oxide (lower part) relevant/complementary to click chemistry (R1–R10, various substituents; Nu, nucleophile) [27].
Thermal isolation materials are widely used to improve energy saving in industries and buildings [43]. Modified cellulose nanofiber/graphene oxide/sodium montmorillonite aerogel materials present improved properties compared with traditional organic insulation materials and are considered as potential materials in building insulation, firefighting, and the chemical industry [44]. Cao and Yuan [43] developed doped ammonium polyphosphate/graphene oxide/cellulose nanofiber-based aerogel with excellent thermal isolation and flame-retardant properties via the freeze-drying method. Composites prepared by utilizing expandable graphite/chemically treated pulverized oil palm empty fruit bunch fiber-reinforced epoxy exhibit improved fire resistivity, thermal properties, and mechanical properties [45].

Wicklein et al. [46] developed lightweight anisotropic foams on the basis of cellulose nanofibers, graphene oxide, and sepiolite nanorods through the freeze-casting of suspensions. The produced foams have excellent thermal-insulating and fire-retardant properties, which can be utilized in buildings. Spatial confinement and freeze-drying methods are used to prepare the hierarchical graphene-confined zirconium phosphate/cellulose nanofiber aerogel [47]. The prepared hybrid aerogel, which is mechanically stable and has outstanding thermal insulation and flame retardancy, can be utilized as a thermal...
insulator for improving energy efficiency in buildings. Guo et al. [48] prepared phosphorus-hybridized graphene nanosheets/cellulose nanofibers foam with excellent fire retardancy through the freeze-drying method.

Ternary nanocomposites utilizing bentonite, graphene oxide, and galactomannan crosslinked and strengthened with borate with the hierarchical structure are developed and exhibit high tensile stress and toughness. The brilliant fire-retardant property of these nanocomposites makes them potential candidates in fire-protective insulation, packaging, and coating applications [49]. Efficient flame-retardant chitosan and graphene oxide in open-cell polyurethane foam coatings are prepared via layer-by-layer methods. The three bilayers exhibit 54% reduction in the peak of heat release rate, whereas six bilayers slow down the volatile release to low flammability limit [50].

Wang et al. [51] developed the multifunctional hybrid film through the reduction of graphene oxide nanosheets in the presence of montmorillonite nanoplatelets and cellulose nanofibers via vacuum filtration. The film has outstanding fire resistance, high thermal stability, and good flexibility. Its high dielectric constant and low dielectric loss show its potential as dielectric material in the capacitor field application.

Metal corrosion considers global challenges that seriously affect the economy, industry, and society [52]. Among various protection methods, anticorrosion coatings have attracted huge attention, and graphene-based anticorrosion coating materials are extensively studied [53–55]. However, chitosan, as a green, low-cost, and nontoxic natural biopolymer with good film-forming tendency, is used as anticorrosive coating for metals [56,57], and improved hydrophobicity by the incorporation of graphene-based materials leads to improved anticorrosion performance. Fayyad et al. [58] improved the anticorrosion performance of pure chitosan through the preparation of oleic acid-grafted chitosan/graphene oxide composite coating on carbon steel by 100-fold. The corrosion inhibition activities of some biopolymers as cheap, abundant, and eco-friendly counterpart in graphene-based composites have been studied to investigate the hybridizing effect. Graphene oxide–chitosan and graphene oxide–chitosan–ZnO composites have higher anticorrosion efficiencies by 83.8 and 85.6%, respectively, compared with graphene oxide [59]. The incorporation of modified gelatin and modified gelatin/graphene oxide in epoxy coating shows better anticorrosion performance on mild steel compared with pure epoxy coating [60]. Homogeneously dispersed graphene oxide improves the compactness and crosslinking degree of coating and increase the anticorrosion performance of coating by 59%. These results are promising for conducting research for the development of green and improved anticorrosion coating by incorporating biopolymer/graphene-based composites into usual coating materials. Linseed oil-based/reduced graphene oxide nanocomposites with anticorrosion properties exhibit the decreased corrosion rate by around 5,000 times when applied on mild steel [61].

Despite utilizing various phase-change materials in walls, floor heating, and building envelopes, which can consist of biopolymer/graphene-based composites for energy management in buildings, these composites will be reviewed under phase-change materials in the following subsections.

### 3.2 Biopolymer/graphene-based composites for catalyst applications

Despite developing various graphene-based composite for fuel cell application [62], biopolymer/graphene-based nanocomposites can be considered as green substrates for various catalyst materials. The synergetic effect of nanocomposite can promote the catalyst performance.

Pt/1,1′-dimethyl-4,4′-bipyridinium dichloride-functionalized graphene oxide/chitosan shows good catalytic activity for methanol [63] and ethanol electro-oxidation methods for direct ethanol fuel-cell application [64]. Pd-functionalized chitosan/reduced graphene oxide is developed as a catalyst with high activity and stability for formic acid electro-oxidation reaction in fuel cells [65].

The Pd/graphene oxide/chitosan/cellulose nanowhisker hydrogel presents good catalytic activity in the Mizoroki–Heck reaction to generate new C–C bonds [66].

Esmaeilzadeh et al. [67] synthesized Pd immobilized on the hybrid of magnetic graphene quantum dots/cyclodextrin-decorated chitosan as a hydrogenation catalyst. Chitosan/glutaraldehyde/reduced graphene oxide/Pd composite hydrogels are prepared via crosslinking and exhibit promising catalytic performance for the degradation of p-nitrophenol and o-nitroaniline. The developed hydrogels are considered as a potential catalyst for the reduction of organic pollutants in wastewater treatment [68]. The synthesis of porous Pd/chitosan/reduced graphene oxide microspheres via etching and freeze-drying methods leads to active and easily recyclable catalysts for Heck reactions [69]. Developed sodium alginate/sulfonated graphene
oxide using blending and casting methods [70] exhibit good results to be considered as a potential alternative membrane in direct methanol fuel cell application.

### 3.3 Biopolymer/graphene-based composites in medical applications

Graphene may be used for various applications, like biosensing, bioimaging, and medical and electronic devices [71]. For biological applications, the high dispersibility and stability of applied nanomaterials in biological fluids are important and considered as shortcomings for graphene nanomaterials because of their intrinsic properties. The tendency of graphene nanomaterials for the adsorption of proteins that make them prone to internalization by macrophages and clearance from the body due to shortage of their half-life in the blood circulation is another consequence of utilizing graphene-based nanomaterials in biological applications. Other drawbacks are toxicity, low biocompatibility, and in vivo distribution profile, which may cause adverse side effects [32].

Different modification and functionalization methods of graphene via covalent or noncovalent mechanism have been studied to improve the various properties of graphene, such as biodegradability, biocompatibility, and safety [16,17]. Consequently, various biocomposites, which utilize carbohydrates (such as cellulose, chitin, alginate, and cyclodextrins), are prepared and investigated [32,72]. For example, antibacterial graphene-reinforced composite coatings based on chitosan, hydroxyapatite, and antibiotic gentamicin are fabricated for bone tissue engineering [73]. Dacrory [74] recently reported the antimicrobial activity of dialdehyde cellulose/graphene oxide film against COVID-19. Many applications based on using graphene and biomolecules require biomolecules (such as peptides, proteins, and enzymes) to interact with graphene at one end while simultaneously exposing the other end to the surrounding medium for biosensing analytes in solution. For example, the molecular interaction of graphene nanoribbons with peptides and enzymes present in the body for nanobiotechnological applications is reported [75]. Moreover, Wang and coworkers [76] successfully probed the molecular interaction of graphene oxide and nitrogen-functionalized graphene with bone morphogenetic protein-2 as key proteins implicated in bone repair with highest adsorption energy. Functionalization can provide different features for the development of an efficient drug delivery system with different drug release profiles, such as water solubility, biocompatibility, stability, bioavailability, and enhanced loading capacity, and reduce the immune system activation. Hybrid materials comprising graphene oxide, β-cyclodextrin, and poly(amineamine) dendrimer are successfully prepared by covalent bonding and utilized as potential nanocarriers for anticancer drugs [77]. Moreover, Pooresmaeil et al. [78] utilized carboxymethyl cellulose as a coating agent for the encapsulation of magnetic graphene oxide coated with mesoporous silica for safe and sustained ibuprofen delivery. Shende and Pathan [72,79] extensively reviewed the interaction and application of various carbohydrate/graphene-based composites in biomedical fields, such as drug delivery and tissue engineering (Figures 7 and 8).

Functionalization can also provide the capability of targeting the nanocarrier, thereby improving the therapeutic efficiency (Figure 8). The modified graphene biocomposite tends to adsorb a wide range of hydrophobic drugs quickly and release them to the selected zone of organisms as a targeted drug delivery system. For example, galactosylated chitosan in combination with graphene oxide has been introduced as a new nanobiocomposite for doxorubicin delivery [80]. This strategy is considered advantageous because functionalized polymer–graphene biocomposites allow the targeting of high drug doses at the site of action, prevent the nonhomogenous biodistribution of therapeutic cargos, and overcome the adverse effects to the normal tissues caused by their interactions with anticancer therapeutics to an extent.

The biomedical application of graphene-based/biopolymer composites has been studied by Chen et al. [81]. Studies showed that graphene size and structure affect the properties of graphene–biopolymer composite properties [82,83]. Lalwani et al. [84] showed that the mechanical strength of biocomposite follows the order: graphene nanoplatelets > graphene nanoribbons > graphene nanotubes. These findings can extend their application in tissue engineering.

The antibacterial, antifungal, and antioxidant properties of graphene-based biopolymer composites make them interesting materials for active food packaging applications [85]. These properties are promoted because of the synergistic effect for free-radical inhibition (Figure 9). The amino and hydroxyl groups on biopolymers improve the oxidation inhibition by hydrogen donation [86].

Preparation conditions, such as ultrasonication time and heat treatment, affect the properties of chitosan/graphene oxide nanocomposite films. Optimum conditions lead to the preparation of films with complete degradation property, which can be used as green packing materials, in soil compost within 28 days [87].
Figure 8: Methods for targeted delivery by graphene-based materials decorated with different carbohydrate polymers [32].

Figure 9: Improvement of the scavenger activity of reduced graphene oxide in the presence of free radicals when combined with natural bioactive compounds [86].
Sonication mixing and solution casting methods are used to prepare cellulose nanowhisker/graphene nanoplatelet films [88]. The prepared films show high thermal, electrical, and mechanical properties with potential as green antistatic and electronic packaging materials.

To overcome the low mechanical strength of hydrogel as a drawback in nerve tissue regeneration application, Jafarkhani et al. [89] developed nanographene oxide/chitosan hydrogels with a 20% increase in the nerve cell growth. The developed poly(vinyl alcohol)/chitosan modified graphene oxide biocomposite presents improved antimicrobial and anti-inflammatory properties in vivo [90].

The design and preparation of graphene-based biomaterial scaffolds for neural regeneration application have been investigated by Reddy et al. [91]. Zhang et al. [92] synthesized antibacterial Ag-polydopamine/bacterial cellulose/reduced graphene oxide composite film, which shows excellent biocompatibility as wound dressing materials. Sodium alginate/graphene oxide/polyvinyl alcohol sponge nanocomposites prepared via the freeze–thaw cyclic process and freeze-dried molding show good water absorption, breathability, and mechanical properties. Investigation revealed that these nanocomposites are promising materials for wound healing applications. Despite the huge potential of chitosan as food packaging and wound healing materials, various modifications are required to tailor related properties, such as vapor permeability, optical transparency, and hydrophilicity. In this regard, the simple solvent casting method has been used to prepare flexible and transparent antimicrobial films by using chitosan/polyvinylpyrrolidone/graphene oxide nanosheets [93].

Bacterial cellulose/graphene-based composites show good potential in the biomedical application as well. Luo et al. [94] presented the improved cell behavior of graphene and graphene oxide in 3D nanofibrous bacterial cellulose scaffold compared with that of graphene. Rostami et al. [95] developed sharkskin-mimicked graphene oxide/chitosan membrane with improved antibacterial and cytocompatibility properties for diverse biomedical applications.

Qi et al. [96] prepared graphene oxide/poly-L-lysine composites films as bioscaffold coatings for potential application in stem cell research and developed a mild and inexpensive preparation method that can utilize other biomacromolecules, such as chitosan and gelatin in the composite preparation. The developed graphene nanoplatelet/hydroxyethyl cellulose graft poly(lactic acid) copolymer/polyurethane bionanocomposites with high thermal stability and water resistance show potential applications for electronics and medical areas [97].

Rebekah et al. [98] developed Fe nanoparticles decorated with graphene oxide/chitosan composite as a nanocarrier for protein delivery. The composite’s good drug loading and release profile make it a potential nanocarrier for clinical applications.

Functional 3D scaffold chitosan–graphite oxide nanoplatelet core–shell microparticles are synthesized via alkaline gelation and template-assisted assembly methods [99]. The outer shell protects the inner chitosan from enzymatic degradation and hydrophilic oxygen-containing functionalities on the outermost layer of graphene oxide, and its inner conductive graphitic core can maintain the bioactivity of the scaffold and promote the adhesion and growth of the neuronal cell.

### 3.4 Application as self-healing materials

Intelligent materials with the capability to respond to external stimulus, such as heat, electromagnetic wave, and mechanical force, and repair themselves after damage are self-healing materials. This ability allows them to refurbish their intrinsic properties and reach sustainable development by extending their lifetime, security, and saving cost [100]. Given their interesting graphene properties, self-healing graphene-based materials have attracted a huge interest. Considering the importance of preparation methods on the structure and properties of materials, various methods, such as solution mixing, in situ polymerization, layer-by-layer assembly, wet-fusing assembly, and hydrothermal method, are utilized to prepare graphene-based self-healing materials [100]. These materials can be utilized in different applications, such as biological applications, sensors [101], coatings [102,103], supercapacitors, flexible electronic devices and artificial skin, self-cleaning, adsorbent, biomimetic materials, and actuator. Figure 10 presents various forms of graphene-based self-healing composites. Graphene can be used as a reinforcement agent or a compound material in self-healing composites. The forms of graphene in self-healing materials are (a) graphene- [104], (b) graphene derivative- [105], and (c) crosslinked polymer/graphene-based [106] self-healing materials.

The self-healing chitosan/graphene oxide nanosheet hydrogel prepared by the self-assembly method shows potential in biomaterial, wastewater treatment, and smart material applications [107].
Lee et al. [108] fabricated a series of self-healing composites by utilizing graphene-based biopolymers for advanced electromagnetic applications. The modified cellulose graphene-based nanocomposite reveals fast electronic self-healing property, making it an interesting candidate in self-healing electronics, artificial skins, soft robotics, biomimetic prostheses, and energy storage applications [109].

The crosslinking of hydrogel to achieve reversible coordination bonding is considered a method for the preparation of stimulus-responsive and self-healing materials. Wang et al. [110] prepared mussel-inspired catechol-containing polyaspartamide/graphene oxide hydrogel with highly improved storage, loss moduli, and tensile and compressive strengths as soft matter for biomedical applications. The photo-crosslinked gelatin/reduced graphene oxide hydrogel exhibits potential in rapid bone regeneration application [111].

Samadi et al. [112] developed polyvinyl alcohol/agar/graphene nanocomposite hydrogels based on three construct dynamic networks with high strength, toughness, and quick self-healing property for biomedical applications. The stretchable, self-healing, and conductive TEMPO-oxidized cellulose nanofiber/graphene nanocomposite hydrogel has been developed via an in situ free-radical polymerization with good potential in self-healing wearable electronics applications [113]. Xie et al. [114] developed the multifunctional flame-retardant nacre-inspired functional cellulose/graphene oxide nanocoating by using a one-step self-assembly method. Its ultrasensitive fire warning and self-healing make the nanocoating interesting materials in building, transportation, and electrical equipment applications.

Chitosan/graphene oxide composite hydrogels incorporated with mussel-inspired protein polydopamine have been developed [115]. Covalent bonds, supramolecular interactions, hydrogen bonding, and π–π stacking in these polydopamine-based hydrogels provide adhesive, conductive, self-healing, and fast-recovering hydrogels with remarkable potential in electroactive tissue engineering applications.

Three-dimensional bioprinted phenol-rich gelatin/graphene oxide hydrogel as a component for a myogenesis-inducing material is considered as potential materials in muscle tissue engineering and regenerative medicine application [116]. Bioderived aliphatic hyperbranched polyurethane/3-aminopropyltriethoxysilane-modified graphene oxide nanocomposites produce smart materials with considerable enhanced mechanical and self-healing properties under microwave and sunlight. These materials have versatile application as self-healing and self-cleaning materials [117].

Despite the preparation of the self-healing bio-based elastomer [118] and possibility of the addition of graphene-based materials, these composites have not been studied yet.

### 3.5 Application in electronics

The abundance, biocompatibility, biodegradability, lightweight, strong electrical conductivity, excellent ion conduction, and mechanical flexibility of biopolymers make them interesting materials for developing flexible, wearable, implantable, and environment-friendly electronic materials [119].

Given their functional groups, such as hydroxyl, amino, amine, and carboxylic groups, various molecular engineering strategies can be used to modify and tailor optoelectrical properties [120,121]. Their abundant functional groups provide high solubility and dispersion properties, thereby giving the potential to adopt low-temperature manufacturing methods and avoiding degradation caused by other tough techniques used for semiconductor patterning methods [119]. It provides an opportunity to develop low-cost, large-area, and flexible bioelectronic devices by using conventional printing techniques [119, 122–124]. For example, the preparation of flexible nanofibrillated cellulose/reduced graphene oxide/carbon nanotube-conductive composite membrane via ultrasonic
dispersion and low-temperature hot pressing leads to composite membranes with potential in conductive membranes, electrode materials, and other electronic applications [125].

The development of environmental-friendly triboelectric nanogenerator biopolymer/graphene-based composites can prepare potential low-cost and biocompatible materials for various applications [126]. Gao et al. [127] prepared a super-elastic, lightweight chitosan/graphene oxide scaffold with potential in mechanical cushioning, energy damping, and flexible device applications via bidirectional freezing and annealing.

### 3.6 Application in fuel cells

The request for low-cost polymers with optimized proton conductivity, reduced methanol crossover, and thermal stability for fuel-cell membrane is increasing [128]. Chitosan, as a cheap, abundant, and eco-friendly biodegradable polymer with amino/hydroxyl groups, is considered an interesting biopolymer to prepare biopolymer/graphene-based composites as proton-exchange membranes for fuel cell applications [129]. Various functionalization methods can be considered to overcome the low proton conductivity of chitosan. Shirdast et al. [130] prepared sulfonated chitosan/sulfonated graphene oxide membranes for fuel cell applications. Nanocomposites showed high thermal/mechanical conductivity and selectivity (increases by 45% and 650%, respectively) properties compared with chitosan membranes for proton transport. Chen et al. [81] studied various preparation methods of advanced graphene-based/bio-based polymer composite membranes for fuel cell applications. Chitosan/sulfonated graphene oxide nanosheet membranes are developed as superior conduction proton-exchange membranes for fuel cells [131]. Pandey and Shahi [132] developed $N,N$-dimethylenephosphonic acid propylsilane graphene oxide/$N$-o-sulphonic acid benzyl chitosan with high water retention and conductivity for the preparation of multifunctional polymer electrolyte membranes.

Azli et al. [133] prepared potato starch/graphene oxide composite as the Li$^+$-conducting polymer electrolyte with high ionic conductivity and mechanical properties.

The layer-by-layer deposition method is used to prepare chitosan and graphene oxide on the membrane substrates of sulfonated poly(vinylidenefluoride) (PVdF) or sulfonated PVdF-co-hexafluoropropylene for utilization as proton-exchange membranes at high-temperature fuel cells [134]. Yaqoob et al. [135] developed cellulose-derived graphene/polyaniline nanocomposites as novel and natural anode materials for benthic microbial fuel cells. The modified anode has higher performance by four times and high removal of Cd(II) and Pb(II) heavy metals from wastewater through benthic microbial fuel cells.

The synthesis of a new anion-exchange membrane with high conductivity achieves the utilization of cellulose, graphene oxide, and polyphenylene oxide [136]. After functionalization with 1,4-diazabicyclo[2.2.2]octane, the mechanically stable membrane shows a hydroxyl conductivity of 215 mS cm$^{-1}$ at 80°C. Hosseinpour et al. [137] developed multilayer proton-exchange membrane utilizing cellulose nanocrystal for direct methanol fuel cell application. The developed membrane has presented a 20% increase in proton conductivity and 11% reduction in methanol crossover compared with Nafion N115.

### 3.7 Application in batteries

In the production of commercial Li-ion batteries as energy storage devices (including EV), active ingredient slurries are prepared using PVdF as a binder due to its adhesion properties and electrochemical stability. Unfortunately, some disadvantages, such as toxicity and nonenvironmental friendliness, are associated with the use of PVdF. $N$-Methyl-pyrrolidone (NMP) has been used as a solvent in the preparation of Li-ion electrodes, and both are costly [138–140]. The disadvantages of NMP are toxicity, nonvolatility (high melting point of about 200°C), and nonenvironmental friendliness because expensive solvents require high costs for purchase and waste maintenance [141]. Therefore, many researchers attempted to find other alternatives and investigate the suitability of other water-soluble, inexpensive, and environmentally friendly materials toward green binders to test as alternative binders (e.g., gelatin, chitosan tragacanth gum, and sodium alginate) [142]. Next, graphene can be considered as an active ingredient in preparing the biopolymer composite for Li-ion batteries due to its capability to participate in the energy storage intercalation mechanism (Figure 11). As reported by Raccichini et al. [143], an anode consisting of a single layer of graphene can host twice as many Li$^+$ than that consisting of conventional graphite. The storage of a lithium ion on each side produces graphene in Li$_x$C$_6$ stoichiometry, which provides a specific capacity of 744 mA h g$^{-1}$ that is twice that given by conventional graphite (372 mA h g$^{-1}$) [144–146]. Sandwiched pyrolyzed
bacterial cellulose/graphene oxide composite is prepared via a simple method [147]. Li–S batteries assembled with pure sulfur as cathodes and prepared composite interlayer exhibit around 600 mA h g\(^{-1}\) at 3 C and only 0.055% capacity decay per cycle, which can be observed in over 200 cycles.

Table 1 presents the data of some studies in bio-polymer/graphene-based composites in batteries.

3.8 Applications in supercapacitors

Supercapacitors with energy storage properties have fast charge–discharge process and outstanding cyclability and are considered environmentally friendly materials. Their performance depends on their surface area. Flexible supercapacitors require electrodes with high electrochemical performance and flexibility. The physical and chemical properties of nanocellulose and graphene-based materials make them interesting materials in supercapacitor application. Cellulose, as one of the most abundant natural organic polymers that originate from plants, woods, and bacteria, is a promising biopolymer with excellent mechanical properties, which can be improved by functionalization or structural modification [151].

Cellulose and its composites with different structures and properties can be utilized [21]. Graphene and graphene-based materials in various structures have been utilized extensively in supercapacitor application [155].

The use of nitrogen self-doped biopolymers, such as chitosan, in the preparation of graphene-based composite aerogels for high-performance supercapacitors also open new interesting research areas for the use of biopolymers in supercapacitors [156]. Various technologies, such as carbonization, biotemplate methods, and biocomplex technologies, can be used to prepare biopolymer-based composites for supercapacitors and other applications [157,158].

Xing et al. [159] reviewed the utilized design and preparation methods for nanocellulose/graphene-based composites in flexible supercapacitor application. Xing et al. [159] found out that controlling the distribution, structure, morphology, and quantity of nanoparticles on the surface; improving synergy in the composite; and simplifying the preparation process are the key factors for tailoring the properties of nanocellulose/graphene-based composites and their industrialization.

Liu et al. [160] developed 3D hierarchical porous glucose/graphene-based aerogels via hydrothermal and CO\(_2\) activation processes for gas adsorption and supercapacitor applications. Table 2 presents some studies conducted on biopolymer/graphene-based composites for supercapacitor application.
| Ref. | Composites | Electrolytes | Properties |
|------|------------|--------------|------------|
| [148] | Graphite:graphene:CMC carrageenan | 1 M LiPF<sub>6</sub>, EC:EMC:DMC (1:1:1, v/v/v) | ▪ Graphite:graphene vs roman in table 1: CMC carrageenan composite/Li achieves optimum discharge capacity and voltage range of 0.01–3.0 V.  
▪ Q<sub>d</sub> = 202.2 mAh g<sup>−1</sup> and CE of 94% for 100%:0%:10% composition  
▪ Q<sub>d</sub> = 314.3 mAh g<sup>−1</sup> and CE of 96% for 95%:5%:10% composition  
▪ Q<sub>d</sub> = 304 mAh g<sup>−1</sup> and CE of 93% for 90%:10%:10% composition  
▪ Improved capacity, cycle performance, and CE effect of using CMC carrageenan binder over PVdF |
| [148] | Graphite:graphene:CMC chitosan | 1 M LiPF<sub>6</sub>, EC:EMC:DMC (1:1:1, v/v/v) | ▪ Graphite:graphene:CMC chitosan composite/Li achieves optimum discharge capacity and voltage range of 0.01–3.0 V.  
▪ Q<sub>d</sub> = 324 mAh g<sup>−1</sup> and CE of 93% for 100%:0%:10% composition  
▪ Q<sub>d</sub> = 298 mAh g<sup>−1</sup> and CE of 95% for 95%:5%:10% composition  
▪ Q<sub>d</sub> = 386 mAh g<sup>−1</sup> and CE of 91% for 90%:10%:10% composition  
▪ Improved capacity, cycle performance, and CE effect of using CMC chitosan binder over PVdF |
| [148] | Pyrolyzed bacterial cellulose/graphene oxide | 1 mol LiTFSI and 4 wt% LiNO<sub>3</sub> dissolved in (TEGDME) | ▪ Sandwiched pyrolyzed bacterial cellulose/graphene oxide composite was prepared via a simple method. Li–S batteries were assembled with pure sulfur as cathodes, and the prepared composite interlayer exhibited around 600 mAh g<sup>−1</sup> at 3 C and only 0.055% capacity decay per cycle, which could be observed in over 200 cycles |
| [149] | (Si:graphite:graphene)/CB:biobinder BTR Co. Ltd, Shenzhen, China | 1 M LiPF<sub>6</sub>, EC:EMC:DMC (1:1:1, v/v/v) | ▪ 2%:8%:5% composition of Si:Graphite:Graphene/Li achieves optimum discharge capacity and voltage range of 0.01–2.0 V.  
▪ Q<sub>d</sub> = 820.7 mAh g<sup>−1</sup> and CE of 77.98% for 8%:1%:20% composition of (Si:graphite:graphene)/CB:BTR  
▪ Superior cycle performance improvement effect of graphene addition  
▪ Impedance reduction due to addition of graphene  
▪ The type of binder affects the capacitance performance of the LIB electrode |
| [148] | Sn:PVDF/LiSn:CMC/LiSn:CMC:AB/Li | 1 M LiClO<sub>4</sub>, EC:DEC (1:1, v/v) | ▪ Sn:PVDF/Li achieves optimum discharge capacity, voltage range of 0.01–2.0 V vs Li/Li<sup>+</sup>, Q<sub>d</sub> = 500 mAh g<sup>−1</sup>, and CE of 67.2% for 80%:20% composition.  
▪ Sn:CMC/Li achieves optimum discharge capacity, voltage range of 0.01–2.0 V vs Li/Li<sup>+</sup>, Q<sub>d</sub> = 800 mAh g<sup>−1</sup>, and CE of 65.4% for 80%:20% composition.  
▪ Sn:CMC:AB/Li achieves optimum discharge capacity, voltage range of 0.01–2.0 V vs Li/Li<sup>+</sup>, Q<sub>d</sub> = 800 mAh g<sup>−1</sup>, and CE of 76% for 60%:20%:20% composition.  
▪ Superior cycle performance improvement of CMC binder effect |
| [150] | Graphite:graphene oxide:CB/Li | 1 M LiPF<sub>6</sub>, EC:DMC:DEC (1:1:1, v/v/v) | ▪ Graphite:graphene oxide:CB/Li achieves optimum discharge capacity, voltage range of 0.01–3.0 V vs Li/Li<sup>+</sup>, and Q<sub>d</sub> = 450 mAh g<sup>−1</sup> for GCG (75%:17%:8%) |

(Continued)
3.9 Biopolymer/graphene-based composites as phase-change materials

Phase-change materials are promising for energy storage in smart thermal energy management, transfer, conversion (such as solar-to-thermal and electric-to-thermal), portable thermal energy, and advanced multifunctional areas [177]. Phase-change materials with low thermal conductivity can be utilized as a thermal insulator for buildings to maintain indoor thermal comfort and HVAC systems overload [178–182]. However, on the basis of their application overcoming their deficiencies, such as low thermal conductivity, low electrical conductivity, weak photo absorption, and shape instability, their performance should be increased. The incorporation of various carbon-based support materials, such as graphene-based nanostructure composites, is considered a versatile solution for these issues [177,183]. The modification can be done using various methods, such as physical blending, microencapsulation, spinning, and preconstruction 3D filler network [184].

Fatty acid/graphene and fatty acid/graphene oxide nanocomposites are promising materials for phase-change materials with high thermal conductivity [185]. Yuan et al. [186] prepared microcapsules of paraffin@ethylcellulose/methylcellulose/graphene oxide composite with improved photo-to-thermal conversion performance for solar–thermal storage application. The polyethylene glycol/cellulose/graphene aerogel is prepared via vacuum impregnation with high thermal conductivity, good shape stability, and large latent heat of fusion [187]. The incorporation of cellulose nanocrystals with graphene nanoplatelets and polyethylene glycol (PEG) control the leakage during phase transition and improve thermal conductivity [188].
The prepared composite has excellent light–thermal and electrothermal conversion capabilities. Thus, phase-change materials may provide stable temperature conditions for electronics operating in extreme environmental conditions. The new organic phase-change materials with a three-dimensional network are prepared by utilizing graphene nanoplatelet aerogel, melamine foam, cellulose nanofiber, and paraffin wax [189–191]. The developed phase-change material shows a 407% increase in thermal conductivity compared with paraffin wax and presents outstanding light–thermal and electrothermal transition capabilities. Considering the biomass waste management concept, the 3D, porous, reduced graphene oxide/modified spent coffee grounds with capability to adsorb 60% PEG is prepared and exhibits high leakage control performance [192]. The synthesized composite with high stability shows the excellent light–heat conversion rate, making the composite a good candidate for solar thermal storage in a short time.

Darzi et al. [193] prepared phase-change materials via the electrospinning of fatty acid ternary eutectic mixture with carbon fiber powder and graphene. The prepared
3.10 Application as sensor materials

The presence of various pollutant materials in air and water and the hazardous effects of creatures reveal the importance of sensors for their detection. Various fluorescence sensors for molecular recognition have been developed using functional nucleic acid-based composites due to their high flexibility [197]. Gao et al. [198] studied graphene-based DNA sensors. Lopez and Liu [199] reviewed the covalent and noncovalent functionalization of graphene oxide/DNA sensors. The advantages and disadvantages of each sensor are discussed.

Nanocellulose/graphene-based nanocomposites with outstanding mechanical properties and high electrical and thermal conductivity properties may be used in multisensing applications. These nanocomposites can respond to multiple stimuli, such as mechanical stimuli, environmental stimuli, and other human biosignals [29]. These nanocomposites can be combined with other materials to fabricate sensors. Various studies in this area are conducted and reviewed [29].

In biopolymer/graphene-based composite preparation, the homogenic dispersion of graphene-based materials in the polymer matrix is an important factor for the modification of its properties for various applications. Utilizing a facile, rapid, green, and inexpensive way for the large-scale preparation methods is important for commercialization and sustainability. Magerusan et al. [200] utilized the electrochemical exfoliation of graphite rods without the use of any organic solvent to prepare chitosan/graphene nanomaterial as a sunset yellow sensor. Table 3 lists some related studies in the sensor field.

3.11 Application in microextraction

Sample preparation in analytical chemistry is considered as the most time-consuming step and the main source of laboratory wastes. Microextraction methods with green solvents and sorbents are the solution to these issues. Biopolymers prove their potential as sorbents in microextraction methods due to their biodegradability, versatility, and easy functionalization [200]. Biopolymer/graphene-based composites also show promising performance as sorbent materials in microextraction. Table 4 presents samples for this application.

3.12 Biopolymer/graphene-based composites for CO₂ capture

The dramatic increase in atmospheric CO₂ as greenhouse gas due to rapid industrialization and its effect on climate change make it an environmental concern in recent years. CO₂ capture is considered a vital solution to overcome this issue [234]. Developing various materials for effective CO₂ capture is an interesting research area. Various adsorbents are investigated for improved performance. Various materials, such as carbonaceous materials [234,235], nanoadsorbents [236], and inorganic-based materials [237,238], can be utilized in CO₂ capture. Amine-rich solid sorbent materials are extensively studied for CO₂ capture application [239]. Porous materials, such as aerogel, are considered as potential adsorbent materials for various applications, such as CO₂ capture [240]. Chitosan, a low-cost, eco-friendly, and abundant natural polymer with amine groups, is considered an attractive material in CO₂ capture application. However, the mechanical properties and low surface area of chitosan limit its application as an adsorbent. Various modifications, such as doping, amine modification, physical/chemical activation, impregnation, and composite formation, are used to enhance the CO₂ capture performance of adsorbents [234]. The addition of various nanofillers, such as graphene-based
Table 3: Some biopolymer/graphene-based composites for sensor applications

| Composite                                                                 | Detection                                           | Ref.     |
|--------------------------------------------------------------------------|-----------------------------------------------------|----------|
| Graphene oxide and G-quadruplex DNA                                      | Ag⁺ and cysteine                                    | [201]    |
| Graphene oxide–DNA nanosystem                                            | Live cell imaging and detection                     | [202]    |
| DNA-grafted graphene oxide                                              | pH biosensors                                       | [203]    |
| Graphene oxide/G-rich single-stranded DNA                                | K⁺                                                  | [204]    |
| Graphene oxide–quadruplex DNA                                           | Pb²⁺                                                | [205]    |
| Graphene oxide–DNAzyme                                                   | Pb²⁺ and Hg²⁺                                       | [206]    |
| Cellulose/graphene-based nanocomposite                                   | Multiple sensing (temperature, humidity, stress/strain, and liquids) | [207]    |
| Cellulose/graphene nanoplate nanocomposite                               | Stretch and compress sensor                          | [208]    |
| Cellulose nanofibers/graphene oxide composite                            | Humidity sensor and moist-induced electricity generator | [209]    |
| Cellulose/reduced graphene oxide                                         | Chemical vapor sensors                              | [210]    |
| Cellulose paper coated with graphene                                      | Humidity and piezoresistive force sensor             | [211]    |
| Cellulose/graphene/polyamide electrospun nanofibers                      | Hg²⁺ sensor                                         | [212]    |
| Bacterial cellulose/graphene oxide composite                             | Nitrite sensor                                       | [213]    |
| 3D bacterial cellulose/nitrogen-doped graphene oxide quantum dot         | Iron ion sensor                                      | [214]    |
| Graphene/naflon/Pt NPs/chitosan/glucose oxidase                         | Glucose biosensor                                   | [215]    |
| Piezoresistive graphene–nanocellulose nanopaper                         | Strain sensor                                       | [216]    |
| Cellulose microfiber/reduced graphene oxide nanocomposite                | Nitrobenzene sensor                                 | [217]    |
| Graphene/sodium alginate aerogels                                       | Strain sensor                                       | [218]    |
| Reduced graphene oxide/cellulose films                                   | Flexible temperature sensor                         | [219]    |

Table 4: Some conducted studies on biopolymer/graphene-based composites in microextraction

| Composite                                                                 | Application                                                                 | Ref.     |
|--------------------------------------------------------------------------|-----------------------------------------------------------------------------|----------|
| Graphene oxide/chitosan                                                 | Analysis of sulfonamide residues in egg and honey in liquid chromatography–ultraviolet detection | [221]    |
| Hierarchically porous monolith polypyrrole/octadecyl silica/graphene oxide/chitosan cryogel | Extraction and preconcentration of carbamate pesticides in fruit juices  | [222]    |
| Hierarchical mercapto-grafted graphene oxide/magnetic chitosan           | Preconcentration and extraction of mercury ion from water samples            | [223]    |
| Magnetic chitosan and graphene oxide functional guanidinium liquid composite | Solid-phase extraction of protein                                           | [224]    |
| Graphene oxide–alizarin yellow R–magnetic chitosan nanocomposite        | Selective determination of aluminum in water samples                          | [225]    |
| Magnetic chitosan–Schiff base grafted on graphene oxide nanocomposite   | Extraction and quantification of lead ion in blood samples                    | [226]    |
| Molecular imprinted magnetic chitosan/graphene oxide                     | Selective separation/preconcentration of fluoxetine from environmental and biological samples | [227]    |
| Magnetic graphene/chitosan nanocomposite                                 | Nanoadsorbent for the removal of 2-naphthol from aqueous solution           | [228]    |
| Zn(ii)-imprinted polymer grafted on graphene oxide/magnetic chitosan nanocomposite | Extraction of zinc ions from different food samples                          | [229]    |
| Magnetic chitosan/graphene oxide composite                               | Solid-phase extraction of phenyl urea herbicides                            | [230]    |
| Magnetic graphene oxide/grafted chitosan copolymer                      | Adsorbent for catechin in magnetic solid-phase extraction                    | [231]    |
| 3D Chitosan/reduced graphene oxide composites                           | Solid-phase extraction adsorbents for pesticide analysis                     | [232]    |
| Alginate/graphene oxide biocomposite                                     | Microsolid-phase extraction for nonsteroidal anti-inflammatory drugs        | [233]    |

Materials and aerogel preparation, can be an effective solution. In this regard, Alhwaige et al. [241] prepared the chitosan aerogel composite with graphene oxide by using the freeze-drying method. They studied the effect of graphene oxide amount on the CO₂ capture performance of composites. The increase in graphene oxide amounts results in
increased surface area and CO₂ capture performance. The prepared composite exhibits superior CO₂ adsorption and high stability during prolonged cyclic operations.

Hsan et al. [242] prepared chitosan-grafted graphene oxide aerogels for CO₂ adsorption. The CO₂ capture of these aerogels reaches 0.257 mmol g⁻¹, which is much higher than that of chitosan, at 1 bar. The developed low-cost aerogel can decrease the anthropogenic CO₂ gas at favorable temperature and pressure. Kumar et al. [243] developed chitosan/graphene oxide nanocomposite film for environmental-friendly CO₂ adsorption and its catalytic conversion to valuable products for industrial application. Polyvinyl amine/chitosan/graphene oxide membrane is prepared using the surface coating method for CO₂ capture [244]. Results show that adding grafted graphene oxide as a nanofiller can enhance CO₂/N₂ selectivity of the membrane.

### 3.13 Biopolymer/graphene-based composites as adsorbent materials

The importance of water purification is known. Among various solutions, developing tailored nanocomposite membranes attracts huge attention. Natural abundant biopolymers, such as chitosan and cellulose, are considered due to their cost and biodegradability and are a concern in the development of new membranes.

Chen et al. [81] studied graphene derivative/bio-based polymer composite preparation methods and tailored their various properties for specified applications. Their utilization in separation applications, such as dye/organic separation and ion adsorption and separation, is studied.

Spioalá et al. reviewed [245] the effect of different nanomaterials on chitosan-based nanocomposite membranes’ water purification performance.

Various strategies can be utilized to expand the adsorption properties of graphene-based materials on the basis of their composition, surface area, surface electrical structure, and spatial effect toward specific pollutants. The interaction mechanisms between pollutants and adsorbents are critical parameters to consider for improved adsorption performance. Wang et al. [246] studied the material design, performance, regeneration, and reuse of graphene-based functional material adsorbents; their associated adsorption mechanisms with heavy metal ions, dyes, and oils; and co-adsorption of their mixture from water. Various interactions, such as hydrophobic, van der Waals, electrostatic, complexation, π-interaction, and hydrogen bonding interactions, take place in the adsorption process. Considering that the material composition, morphology, structure, and functional groups on the adsorbent play a critical role in removal performance, Kong et al. [247] reviewed available methods for optimizing the heavy metal ions and their compound pollutant adsorption performance of graphene-based materials. Various modification methods to functionalize graphene-based adsorbents with various functional groups are studied for adsorbent optimization [247].

Biomass materials present interesting performance as biosorbents in wastewater remediation. Researchers utilized various modification techniques to improve their adsorption performance for dyes and heavy metals. H-bonding, chelation, electrostatic interaction, and π–π interactions are used for pollutant adsorption [248] Ahmed et al. [249] investigated different chitosan/chitin–carbonaceous material composites for the adsorption of various water pollutants and their regeneration and reusability as adsorbents.

Bacterial cellulose/graphene-based nanocomposites can be utilized as an adsorbent for water–oil separation and metal ion removal in water purification [11]. Their adsorption performance depends on various physicochemical properties, such as density, surface tension, viscosity, polarity, hydrophobicity, porosity, and pore size [250].

The development of super amphiphilic polyurethane/cellulose nanowisker/graphene nanoplatelet foams with good reusability and durability leads to potential materials with an opportunity to be used in oil emulsion-purifying and catalyst-ancreating applications [251]. The development of a 3D magnetic bacterial cellulose nanofiber/graphene oxide polymer aerogel leads to an excellent adsorbent for malachite green with 93% removal performance [252]. Table 5 presents some recent studies conducted on the adsorbent application of biopolymer/graphene-based composites.

### 4 Perspectives, challenges, and recommendations

The industry is waiting to absorb and implement the latest innovations and start manufacturing batteries, solar panels, electronics, photonic and communication devices, and medical technologies utilizing eco-friendly and sustainable materials. Biomedical applications are considered as the first mass applications to pave the way to emerging
Table 5: Some recent studies on depollution application

| Composite                                      | Adsorbent                      | Preparation method          | Ref.  |
|------------------------------------------------|--------------------------------|-----------------------------|-------|
| Chitosan/graphene oxide composite aerogel      | Methyl orange                  | Hydrothermal method         | [253] |
| Cellulose nanofibril/graphene nanoplates hybrid aerogels | Cationic and anionic organic dyes | Freeze-drying              | [254] |
| Chitosan/graphene oxide nanofibrous            | Cu$^{2+}$, Pb$^{2+}$, and Cr$^{6+}$ | Electrospinning            | [255] |
| Immobilized horseradish peroxidase chitosan/graphene oxide nanocomposite | Ofloxacin                      |                              | [256] |
| Fe$_3$O$_4$–graphene oxide/chitosan composite  | Cr$^{6+}$                      | Immersion–evaporation, coprecipitation | [257] |
| Schiff base chitosan/graphene oxide            | Cu$^{2+}$ and Pb$^{2+}$        |                              | [258] |
| Chitosan/graphene oxide nanocomposite          | Pb$^{2+}$                      | NIPS process                | [259] |
| Chitosan/aminopropylsilane graphene oxide      | Pb$^{2+}$ and dyes             |                              | [260] |
| nanocomposite hydrogel                         |                                |                              |       |
| Chitosan-functionalized EDTA–silane/magnetic   | Cd$^{2+}$ and Pb$^{2+}$        | Solvothermal                | [261] |
| graphene oxide                                 |                                |                              |       |

high value-added materials. Then, graphene can be incorporated in ubiquitous commodities, such as adsorbents, batteries, and electronics. However, the growth of biopolymer/graphene-based composite’s market is restrained by the price of production, competitive technologies, market fragmentation, and technical limitations.

Even with the development of various biopolymer/graphene oxides for medical applications, high purity is required to avoid toxicity and contamination and should be considered carefully. Thus, one of the important challenges for medical application is increasing the purity of prepared composites to avoid the side effects of other intermediates. Increased studies on developing bacteria-based nanocellulose can be a solution.

Modification plays a critical role in developing and increasing the performance of tailored biopolymer/graphene-based composites. Various physical and chemical modification methodologies can spearhead efforts to make initiatives in their fabrication to ensure consistency and trustworthiness.

The main goal of all studies is to see biopolymer/graphene-based composites fully integrated in day-to-day products and manufacturing. Based on the achievements and opportunities, future research trends can focus on the following:

1) Various modifications for graphene-based materials, biopolymers, and their composites for tailoring their structure, composition, and properties by considering interfacial interactions and processes and modification strategies that add value throughout the value chain (from new materials to individual components and products) to fulfill future industrial needs should be studied.

2) Finding a facile, rapid, green, and inexpensive way for large-scale preparation methods is important for commercialization and sustainability. Although several production routes for laboratory-scale have been reported, developing new ways to leverage versatility and unique properties of biopolymer/graphene-based composites should lead to continuous market growth.

3) The size control, uniformity, and compatibility based on the properties and performance of biopolymer/graphene-based composites are important and should be investigated.

4) Some properties and performance limitations of biopolymer/graphene-based composites can address by adding another building block or metal or metal oxide functionalization. Metal or metal oxide nanoparticles possess unique features and can alter chemical, electrical, magnetic, and conducting characteristics of targeted composite desirably.

5) The life cycle assessment is an important research area for innovative biopolymer/graphene-based composite materials for their industrialization. Studies should focus on potential differences in energy use, blue water footprint, human toxicity, and ecotoxicity of production routes. The life cycle assessment studies of products at early stages of technological development should be conducted in the future.

6) New composites, such as bioelastomer/graphene-based materials, with superior mechanical, electrical, and/or thermal behaviors for use as a biocompatible elastomer that provides structures with critical strengths for self-healing and 3D-printed scaffold should be developed.

7) Studies on minimally investigated areas, such as the effect of incorporation of biopolymer/graphene-based
composites on cement, anticorrosion, and insulator coating materials, must be conducted.

5 Conclusion

Despite the different advantages of utilizing biopolymer/graphene-based composites in wide applications, such as medical, energy, adsorbent, electronic, and construction, their application is mostly limited to the laboratory scale. Research on understanding their various modification, size, and uniformity control and life cycle assessment should be carried out to overcome this issue and find easy, fast, green, and cheap methods for large-scale preparation. In the future, the increasing interest in developing engineered biopolymer/graphene-based composites with high performance and sustainability is expected to enhance their industrial application. Utilizing biomass wastes as precursors for developing biopolymer/graphene-based composites can enhance their cost effectiveness and eco-friendliness. We hope that this study provides some insight and inspiration for researchers in this field to develop green and sustainable biopolymer/graphene-based composites for various applications.

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