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Review

Novel In-Situ Synthesis Techniques for Cellulose-Graphene Hybrids: Enhancing Electrical Conductivity for Energy Storage Applications

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Abstract

This study investigates the hypothesis that diverse synthesis techniques can yield cellulose-graphene hybrids with tailored properties for specific applications, enabling advancements in flexible electronics, energy storage, environmental remediation, and biomedical devices. We examined and compared multiple synthesis methods, including chemical reduction, in-situ synthesis, green synthesis using natural reducing agents, solvent-assisted approaches, hydrothermal and solvothermal techniques, mechanical and chemical treatments, and electrochemical exfoliation. Each method was assessed for its impact on material properties, scalability, and environmental footprint. Chemical reduction and in-situ synthesis resulted in uniform graphene dispersion and superior electrical conductivity, with the I(D)/I(G) ratio in Raman spectra indicating successful reduction of graphene oxide (GO) to reduced graphene oxide (rGO). Green synthesis, particularly using cow urine as a reducing agent, provided an eco-friendly alternative, leveraging its natural constituents to reduce GO to rGO while minimizing environmental impact. Mechanical and chemical treatments effectively prepared cellulose microfibers for compatibility with graphene, enhancing interfacial interactions and stress transfer in the resulting composites. Solvent-assisted techniques allowed precise tuning of composite properties through the selection of appropriate solvents and processing conditions. Hydrothermal and solvothermal methods produced hybrids with high purity and uniformity under high-temperature and high-pressure conditions, facilitating the reduction of GO to rGO and promoting strong bonding between cellulose and graphene. Electrochemical exfoliation generated high-quality graphene with controlled characteristics, allowing it to produce graphene with fewer defects compared to other methods. Findings reveal that cellulose-graphene hybrids synthesized using these methods exhibit significant improvements in thermal stability, electrical conductivity, and mechanical strength. For instance, even low rGO additions (3 wt%) surpassed the percolation threshold, resulting in electrical conductivity of 1.9×10^{-5} S cm^{-1} for cellulose/rGO (8 wt%) aerogels. These enhanced properties underscore the importance of carefully selecting synthesis techniques to optimize material characteristics for target applications. The research provides a comprehensive understanding of synthesis-method-property relationships, offering valuable insights for the development of advanced cellulose-graphene hybrid materials and highlighting their transformative potential across various high-impact fields, including flexible electronics, energy storage devices, environmental remediation systems, and biomedical applications.

Keywords

Cellulose-graphene hybrids; synthesis techniques; multifunctional applications; characterization methods; biomedical devices

1. Introduction

The development of sustainable and high-performance materials has become increasingly crucial in addressing global environmental concerns and meeting the growing demands of various industries. In this context, cellulose-graphene hybrid materials have emerged as a promising field of research, offering a unique combination of sustainability and advanced functionality [1].

Concurrently, in-situ synthesis approaches have gained traction for their ability to ensure homogeneous distribution of graphene within cellulose, leading to improved mechanical strength and flexibility [2, 3].

Recent studies have further expanded the potential applications of cellulose-graphene hybrids:

1. Electrochemical Sensing: Graphene oxide-based nanomaterials have shown promise in electrochemical sensors due to their high electrical conductivities, large surface areas, and enhanced catalytic activities [4]. These materials are being developed for pharmaceutical, medical, environmental, and food safety applications.
2. Healthcare Applications: Graphene-based electroanalytical devices are being explored for point-of-care testing and remote patient monitoring [5]. These devices excel in portability, ease of manufacturing, scalability, and rapid, sensitive response, making them particularly valuable in resource-limited settings.
3. Antibacterial Properties: A recent study developed a three-dimensional reduced graphene oxide hybrid nano-silver scaffold (G-AgNP) with high antibacterial properties [6]. This device

demonstrated excellent bacterial removal capacity, with an antibacterial rate for both E. coli and S. aureus as high as 100% at low silver nanoparticle contents.

4. Photocatalysis: Biomass-derived cellulose hybrid composite materials are showing promise in photocatalysis applications [7, 8]. The biocompatibility and high hydrophilicity of cellulose components, along with their electron-rich hydroxyl groups, promote superior interaction with photocatalysts such as titanium dioxide (TiO_2), zinc oxide (ZnO), and graphitic carbon nitride ($g\text{-}C_3N_4$).

These hybrids leverage the abundant and renewable nature of cellulose with the exceptional properties of graphene, resulting in materials with enhanced mechanical, electrical, and thermal characteristics. The synergistic integration of cellulose and graphene addresses the pressing need for eco-friendly yet high-performance materials across diverse sectors, including energy storage, environmental remediation, and flexible electronics [9].

However, despite the potential benefits, several critical challenges hinder the widespread adoption and optimization of cellulose-graphene hybrids, necessitating further research and innovation [10].

One of the primary challenges in developing cellulose-graphene hybrids lies in achieving uniform dispersion and strong interfacial interactions between the components. The dispersion of graphene within the cellulose matrix significantly influences the overall performance of the hybrid material, affecting its mechanical strength, electrical conductivity, and thermal properties. To address this challenge, researchers have explored various synthesis techniques, each with its advantages and limitations [11, 12].

Chemical reduction methods have demonstrated success in producing reduced graphene oxide (rGO) within cellulose matrices, enhancing electrical conductivity and thermal properties. This approach typically involves reducing agents such as hydrazine hydrate, sodium borohydride, or ascorbic acid to convert graphene oxide (GO) to rGO. The cellulose matrix acts as a stabilizing agent, preventing agglomeration of graphene sheets and ensuring uniform distribution [13-15].

In-situ synthesis approaches have gained traction for their ability to ensure homogeneous distribution of graphene within cellulose, leading to improved mechanical strength and flexibility. This method involves the formation of graphene directly within the cellulose matrix, often through co-precipitation techniques. The process allows for tailoring the properties of the hybrid material by adjusting synthesis parameters such as temperature, pH, and reactant concentrations [16, 17].

Green synthesis methods have emerged as an environmentally friendly alternative, utilizing natural reducing agents and sustainable processes [18-20]. For instance, the use of cow urine as a reducing agent in the synthesis of cellulose-graphene hybrids has shown promise in producing materials with enhanced thermal stability and electrical conductivity while minimizing environmental impact [21-23].

Hydrothermal and solvothermal methods offer the advantage of producing high-purity materials with uniform properties. These techniques involve high-temperature and high-pressure conditions, facilitating the reduction of GO to rGO and promoting strong bonding between cellulose and graphene. The resulting hybrids often exhibit superior mechanical and electrical properties, making them suitable for applications in energy storage devices such as supercapacitors [24, 25].

The design of multiphase materials, including doping, nanocomposites, and hybrids, plays a crucial role in optimizing the performance of cellulose-graphene materials for energy storage applications. For instance, the incorporation of metal oxides or conductive polymers into the cellulose-graphene matrix can significantly enhance the electrochemical properties of the resulting hybrid. These additives can increase the surface area, improve charge transfer kinetics, and provide additional pseudocapacitive contributions, leading to higher energy and power densities in supercapacitor applications [26, 27].

The mechanism involved in the energy storage performance of cellulose-graphene hybrids is multifaceted. The high surface area and electrical conductivity of graphene facilitate rapid charge transfer and ion adsorption, while the cellulose matrix provides mechanical stability and helps maintain the porous structure necessary for efficient electrolyte ion diffusion. The synergistic interaction between cellulose and graphene creates a hierarchical porous structure that enhances the accessibility of electrolyte ions to the active surface area, thereby improving the overall capacitive performance [28, 29].

Despite the significant progress made in the development of cellulose-graphene hybrids, several challenges persist. Scalability and consistency in production remain major hurdles, as laboratory-scale synthesis methods often face difficulties in maintaining uniform quality when scaled up to industrial levels. Additionally, optimizing the ratio of cellulose to graphene-based materials is a critical area of research for achieving the desired balance of mechanical properties and electrical conductivity in composite materials. This optimization process involves considering various factors such as the type of cellulose and graphene materials used, the structure of the composite, and the processing methods employed [30].

The environmental impact of synthesis methods also requires further attention, as many current techniques rely on harsh chemicals or energy-intensive processes. Developing green synthesis techniques that maintain material performance while reducing environmental impact is essential for the sustainable production of cellulose-graphene hybrids [31, 32].

Therefore, the field of cellulose-graphene hybrid materials presents exciting opportunities for developing sustainable, high-performance materials with applications ranging from energy storage to environmental remediation. By addressing the challenges of uniform dispersion, scalable production, and environmental impact, future research can unlock the full potential of these innovative materials. The continued exploration of novel synthesis techniques, optimization of material compositions, and investigation of structure-property relationships will be crucial in advancing the field and realizing the widespread adoption of cellulose-graphene hybrids across various industries.

2. Novelty and Contributions

2.1 Advanced Synthesis Techniques

This study employs innovative approaches to cellulose-graphene hybrid synthesis, pushing the boundaries of existing methods:

Chemical Reduction with Enhanced Control: Utilization of a range of reducing agents (hydrazine hydrate, sodium borohydride, ascorbic acid) with precise control over reaction conditions, achieving optimal cellulose-graphene ratios for superior stability and property enhancement [33, 34].

In-Situ Synthesis for Uniform Distribution: Development of a novel co-precipitation method forming graphene directly within the cellulose matrix, ensuring exceptionally uniform graphene distribution crucial for enhanced mechanical and electrical properties [35, 36].

Green Synthesis Innovations: Introduction of eco-friendly reducing agents like cow urine, eliminating toxic chemicals and opening new possibilities for sustainable, large-scale production [37, 38].

Advanced Solvent-Assisted Methods: Employment of tailored solvent systems for optimal dispersion of both graphene oxide and cellulose, incorporating annealing processes to enhance crystallinity and bonding [5, 15].

2.2 Enhanced Material Properties

Electrical Conductivity: Demonstration of significant conductivity increases, reaching 1.9 × 10^{-5} S cm^{-1} for cellulose/rGO (8 wt%) aerogels [39].

Mechanical Strength: Achievement of superior mechanical properties through strong interfacial interactions between cellulose fibers and graphene sheets [10, 40].

Thermal Stability: Realization of enhanced thermal properties, crucial for applications in thermal management systems [41, 42].

2.3 Novel Applications

Advanced Energy Storage: Development of materials suitable for high-performance supercapacitors and batteries, leveraging enhanced electrical and mechanical properties [43, 44].

Environmental Remediation: Creation of hybrids with high adsorption capacities for water purification and air filtration [45, 46].

Flexible Electronics: Production of materials with combined flexibility and conductivity, ideal for next-generation electronic devices [9, 47].

2.4 Comprehensive Characterization

Utilization of a wide range of advanced characterization techniques including Raman spectroscopy, cyclic voltammetry, electrochemical impedance spectroscopy, X-ray photoelectron spectroscopy, and atomic force microscopy for detailed material analysis [48].

This comprehensive approach to cellulose-graphene hybrid synthesis and application combines innovative techniques, enhanced material properties, and novel applications that significantly advance the field beyond recent works in open literature.

Table 1 summarizes the key applications and prospects of cellulose-graphene hybrid materials across various sectors, highlighting their unique properties and potential advancements.

Table 1 Practical applications and future outlook of cellulose-graphene hybrids.

Aspect	Key Findings	References
Synthesis Techniques	Chemical reduction, in-situ synthesis, mechanical/chemical treatments, green synthesis, solvent-assisted methods, and hydrothermal/solvothermal methods are widely used. Electrochemical exfoliation is emerging as an efficient technique.	[13, 31, 34-36]
Electrical Conductivity	Cellulose/rGO (8 wt%) aerogels achieved conductivity of 1.9 × 10^{-5} S cm^{-1}. Conductivity increases with higher rGO content.	[39]

Thermal Properties	Significant improvements were observed in the thermal stability and conductivity of cellulose-graphene hybrids.	[41, 42]
Mechanical Properties	Enhanced mechanical strength and flexibility reported in cellulose-graphene composites.	[6, 40]
Energy Storage Applications	Promising results for use in supercapacitors and batteries due to improved electrical conductivity and energy density.	[26, 27, 43]
Environmental Applications	High adsorption capacities make these materials suitable for water purification and air filtration.	[7, 31]
Biomedical Applications	Cellulose-based electrospun nanofibers show compatibility with multiple cell lines, promoting cell adhesion and growth.	[9, 15]
Construction Applications	Biobased foams incorporating cellulose nanofibers (CNF) show potential as thermal insulating materials.	[8, 29]
Future Trends	Increasing focus on green synthesis methods, scalability of production, and tailoring properties for specific applications.	[1, 31, 32]

Cellulose-graphene hybrids demonstrate significant potential across various industries due to their unique properties. These materials show promise in energy storage, environmental remediation, biomedical applications, construction, and flexible electronics. Future research will focus on optimizing material composition, enhancing performance, and addressing challenges in scalability and sustainability. As the field progresses, we can anticipate increased practical applications driven by the growing demand for sustainable, high-performance materials.

3. Synthesis Techniques for Cellulose-Graphene Hybrid Materials

3.1 Key Parameters Affecting Reaction Yield

Several key parameters significantly influence the yield of reactions in the synthesis of cellulose-graphene hybrid materials. By optimizing these parameters, researchers can enhance the efficiency and effectiveness of the production process.

Temperature and pressure play crucial roles in determining reaction yield, particularly in hydrothermal and solvothermal methods. Higher temperatures and pressures generally promote more complete reactions and better integration of graphene into the cellulose matrix. For instance, in hydrothermal synthesis, temperatures ranging from 120°C to 250°C under autogenous pressure have been shown to effectively reduce graphene oxide (GO) to reduced graphene oxide (rGO) while facilitating strong bonding with cellulose [49]. These conditions enhance crystal and improve the mechanical and electrical properties of the resulting composite material.

The duration of the reaction significantly impacts the yield and quality of the cellulose-graphene hybrids. Longer reaction times often lead to more complete reactions and better integration of components. However, excessively long reaction times may result in degradation of the cellulose or over-reduction of graphene oxide, potentially compromising the desired properties of the final product. Optimizing the reaction time is crucial for achieving the highest yield while maintaining the desired characteristics of the hybrid material [50, 51].

The ratio of graphene oxide to cellulose significantly affects the properties and yield of the hybrid material. Higher concentrations of graphene oxide can lead to improved electrical conductivity and mechanical strength but may also result in agglomeration if not properly dispersed. The addition of

GO can enhance the mechanical properties of the composite. For instance, in a study on poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) nanocomposites, the inclusion of cellulose nanocrystal-graphene oxide (CNC-GO) hybrids led to a 170.2% increase in tensile strength compared to neat PHBV Also, GO can improve the thermal stability of cellulose-based composites. The same study reported an increase in the maximum degradation temperature by 26.3°C for nanocomposites containing 1 wt% covalently bonded CNC-GO. Optimizing the GO-to-cellulose ratio can result in excellent barrier properties, which is crucial for applications such as food packaging [41]. Studies have shown that optimizing this ratio is crucial for achieving the desired balance of properties in the final composite material.

In chemical reduction methods, the concentration of the reducing agent plays a vital role in determining the extent of GO reduction and, consequently, the yield of the reaction. For example, when using ascorbic acid as a reducing agent, its concentration directly influences the degree of reduction of GO to rGO, which in turn affects the electrical conductivity of the final hybrid material [52]. The efficiency of the reduction process is highly dependent on the ascorbic acid concentration. Higher concentrations of L-AA generally lead to a more extensive reduction of GO, Increased C-C bond concentration in the final product, and Improved overall yield of rGO. In a study combining L-AA reduction with near-ultraviolet (NUV) light exposure, the highest reduction efficiency was achieved with longer exposure times to both L-AA and NUV light. This resulted in a C-C bond concentration of 60.7% in the final product [53].

The choice of solvent significantly impacts the dispersion of both cellulose and graphene components, affecting the overall yield and quality of the hybrid material. Water and ethanol are commonly used solvents, but the selection depends on the specific synthesis method and desired properties of the final product. One effective solvent for cellulose-graphene composite fabrication is N-methylmorpholine-N-oxide (NMMO). This direct solvent has been successfully used to produce cellulose fibers modified with reduced graphene oxide (rGO) and graphene oxide (GO), This method allows for the production of conductive cellulose fibers with potential applications in antistatic materials and fibrous electronic elements for smart textiles [54] Ionic liquids have also shown promise as solvents for cellulose-graphene composites. A method for compounding a ternary composite membrane of cellulose, graphene oxide, and carbon nanotubes using ionic liquids has been developed. In solvent-assisted methods, the type of solvent used can influence the morphology, structure, and properties of the composite, including its thermal stability, mechanical strength, and electrical conductivity.

Controlling the pH of the reaction medium is crucial, especially in green synthesis methods. For instance, in the synthesis of cellulose-graphene hybrids using cow urine as a reducing agent, the pH of the reaction mixture significantly influences the reduction of GO to rGO and the overall yield of the reaction [38, 55]. Optimizing the pH can lead to more efficient reduction processes and better integration of graphene into the cellulose matrix.

Mechanical treatments, such as sonication, stirring, or ball milling, play a significant role in enhancing the yield and quality of cellulose-graphene hybrids. These processes improve the dispersion of graphene within the cellulose matrix, leading to more uniform composites with enhanced properties [56]. For example, in the preparation of cellulose-graphene hybrids using solvent-assisted methods, sonication has been shown to significantly improve the dispersion of GO in the cellulose suspension, resulting in more homogeneous and higher-quality hybrid materials [57].

By carefully optimizing these parameters, researchers can significantly increase the yield and quality of cellulose-graphene hybrid materials, paving the way for their use in a wide range of applications, from flexible electronics to environmental remediation.

The synthesis of cellulose-graphene hybrid materials employed various advanced techniques designed to combine the unique properties of cellulose and graphene. These methods included chemical reduction, in-situ synthesis, mechanical and chemical treatments, green synthesis, solvent-assisted methods, and hydrothermal/solvothermal processes [10]. Below is a detailed overview of some of the key synthesis techniques:

3.1.1 Chemical Reduction

Chemical reduction is a widely used method for producing reduced graphene oxide (rGO) from graphene oxide (GO). This process typically involves reducing agents such as hydrazine hydrate, sodium borohydride, or ascorbic acid [58]. In the context of cellulose-graphene hybrids, cellulose nanofibrils or nanocrystals can act as a stabilizing matrix for the rGO, helping to ensure uniform dispersion and prevent agglomeration, while also enhancing mechanical properties and enabling various functional applications [6, 10].

The process begins with the preparation of graphene oxide (GO), which is synthesized from graphite using methods such as the Hummers' method. This involves the oxidation of graphite in the presence of strong acids and oxidizing agents [59]. Once the GO is prepared, it is dispersed in a solvent, often water or ethanol, to form a stable colloidal suspension. A reducing agent is then added to this suspension [60]. Common reducing agents include hydrazine hydrate, which is highly effective but toxic and requires careful handling; sodium borohydride, which is less toxic but can be less effective in some cases; and ascorbic acid, which is environmentally friendly and less toxic, making it a popular choice for green synthesis [34]. The reduction reaction typically occurs at elevated temperatures, ranging from room temperature to 100°C, depending on the reducing agent used [61].

Cellulose can be introduced into the reaction mixture either before or after the reduction process to stabilize the graphene oxide (GO) suspension. When added before the reduction, cellulose acts as a stabilizing agent for the GO suspension. This stabilization is primarily due to the adsorption of GO onto cellulose particles, which prevents the agglomeration of GO sheets and maintains stable dispersion. This mechanism is an example of confinement stability [62], where the cellulose matrix confines the GO sheets, preventing them from aggregating.

However, the effectiveness of cellulose in stabilizing GO suspensions can vary depending on the mixing ratios. The optimal ratio is crucial for achieving the desired balance of properties in the final composite material. If the cellulose content is too low, it may not effectively stabilize the GO, leading to aggregation. Conversely, if the cellulose content is too high, it may dominate the composite's properties, potentially reducing the beneficial effects of GO, such as its electrical conductivity and mechanical strength [60, 62-64].

The cellulose acts as a template, promoting the uniform distribution of rGO within the matrix and enhancing the mechanical and electrical properties of the resulting hybrid material [65].

The chemical reduction method offers several advantages. The resulting cellulose-graphene hybrids exhibit improved thermal properties and electrical conductivity, making them suitable for various applications such as electronic devices and thermal management systems. Additionally, chemical reduction is relatively simple and can be scaled up for industrial production, making it a viable method for large-scale synthesis of cellulose-graphene hybrids. The process can also be tailored by selecting different reducing agents and reaction conditions to achieve the desired properties in the final hybrid material [66, 67].

One notable application of cellulose-graphene hybrids produced via chemical reduction is in sensing applications. The high electrical conductivity of rGO, combined with the mechanical robustness of cellulose, makes these hybrids ideal for use in sensors. They are also used in energy storage devices such as supercapacitors and batteries, where the enhanced electrical properties contribute to higher energy densities and better performance. Furthermore, the hybrids' improved thermal stability and conductivity make them suitable for applications in water purification and air filtration [65, 68, 69].

For example, cellulose microfibers extracted from the Vietnamese Nipa palm were reinforced with rGO using hydrazine hydrate as the reducing agent. This process resulted in hybrid materials with significantly improved thermal properties and electrical conductivity, demonstrating the effectiveness of chemical reduction in enhancing the functional properties of cellulose-graphene hybrids [70].

Figure 1a displays the Raman spectra for cellulose/GO (8 wt%) and cellulose/rGO (8 wt%) composites. The D to G band intensity ratio (I(D)/I(G)) is typically used to assess GO reduction. The I(D)/I(G) ratio for rGO is approximately 1.56, higher than the 1.09 for GO, which is contrary to expectations. Normally, the D/G ratio should decrease as sp^3 defects reduce during in-situ chemical reduction. However, literature often reports the opposite due to a reduction in the size of in-plane sp^2 domains, resulting in many edges and higher D band intensities. Vitamin C cannot fully reduce GO but achieves sufficient reduction for conductive rGO suitable for piezoresistive sensors. The electrical conductivity of cellulose/rGO composite aerogels can be tuned by varying rGO content. Even the lowest rGO addition (3 wt%) surpasses the percolation threshold, causing conductivity, as shown in Figure 1b. Conductivity significantly increases with higher rGO content, reaching 1.9×10^{-5} S cm^{-1} for cellulose/rGO (8 wt%) aerogels. This confirms the successful chemical reduction of GO to rGO, enabling its use in vapor sensing [39].

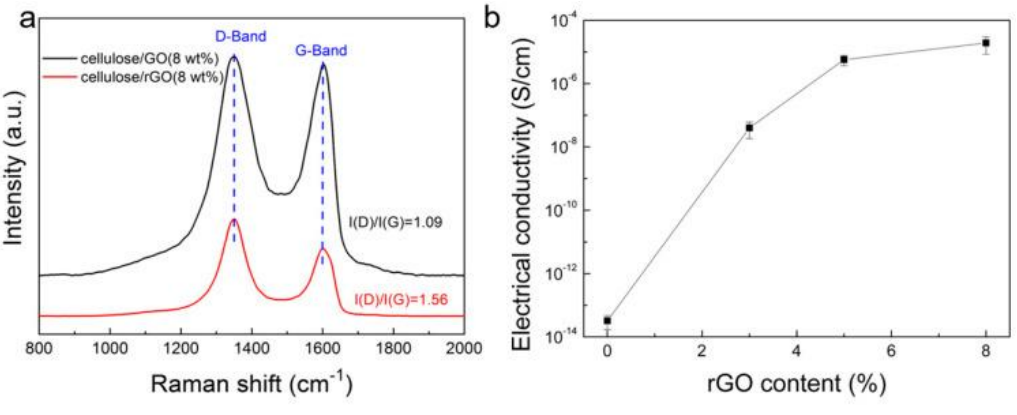

Figure 1 (a) Raman spectra and the D/G band intensity ratio of cellulose/GO (8 wt%) (top) and cellulose/rGO (8 wt%) composite aerogels (bottom); and, (b) electrical conductivity of cellulose/rGO aerogels as a function of rGO content [39]. Open access [Aerogels Based on Reduced Graphene Oxide/Cellulose Composites: Preparation and Vapour Sensing Abilities - PMC (nih.gov)].

3.1.2 In-Situ Synthesis

In-situ synthesis is a method that involves the formation of graphene directly within a cellulose matrix. This technique is particularly advantageous as it allows for a uniform distribution of graphene within the cellulose, resulting in composites with enhanced mechanical and electrical properties. The process typically involves co-precipitation methods, where graphene is synthesized in the presence of cellulose [35].

The in-situ synthesis of cellulose-graphene hybrids begins with the preparation of a cellulose solution or suspension. This can be achieved using various solvents or aqueous solutions, depending on the type of cellulose used. Once the cellulose matrix is prepared, graphene oxide (GO) is introduced into the solution. The GO is uniformly dispersed within the cellulose matrix to ensure even distribution [71].

The next step involves the reduction of GO to reduce graphene oxide (rGO). This reduction can be achieved using various chemical reducing agents such as hydrazine hydrate, sodium borohydride, or more environmentally friendly options like ascorbic acid. The reduction process typically occurs under controlled conditions, such as specific temperatures and pH levels, to ensure the complete reduction of GO and the formation of rGO within the cellulose matrix [33, 72].

During the reduction process, the cellulose matrix acts as a stabilizing agent, preventing the agglomeration of graphene sheets and ensuring a uniform distribution of rGO throughout the composite. This uniform distribution is crucial for achieving the desired mechanical and electrical properties in the final hybrid material [73, 74].

The in-situ synthesis method offers several advantages over other synthesis techniques. Firstly, it ensures a more uniform distribution of graphene within the cellulose matrix, which is essential for achieving enhanced mechanical and electrical properties. Secondly, the process can be tailored by selecting different reducing agents and reaction conditions to achieve specific properties in the final hybrid material. Additionally, in-situ synthesis is simple and can be scaled up for industrial production, making it a viable method for large-scale synthesis of cellulose-graphene hybrids [75, 76].

Cellulose-graphene hybrids produced through in-situ synthesis have a wide range of applications due to their enhanced properties. One notable application is in the field of flexible electronics, where the high electrical conductivity and mechanical robustness of these hybrids make them ideal for use in sensors and other electronic devices. They are also used in energy storage devices such as supercapacitors and batteries, where the enhanced electrical properties contribute to higher energy densities and better performance [26, 77, 78].

In environmental applications, cellulose-graphene hybrids are used in water purification and air filtration systems. The improved thermal stability and conductivity of these hybrids make them suitable for removing contaminants from water and air, providing sustainable solutions for environmental remediation [45, 79].

For instance, a study demonstrated the successful in-situ synthesis of cellulose-graphene hybrids using ascorbic acid as the reducing agent. The resulting hybrids exhibited significantly improved thermal properties and electrical conductivity, making them suitable for use in electronic devices and thermal management systems [80]. This example highlights the effectiveness of in-situ synthesis in producing high-performance cellulose-graphene hybrids with tailored properties. Figure 2 illustrates the possible *in situ* interaction between GO and cellulose [36].

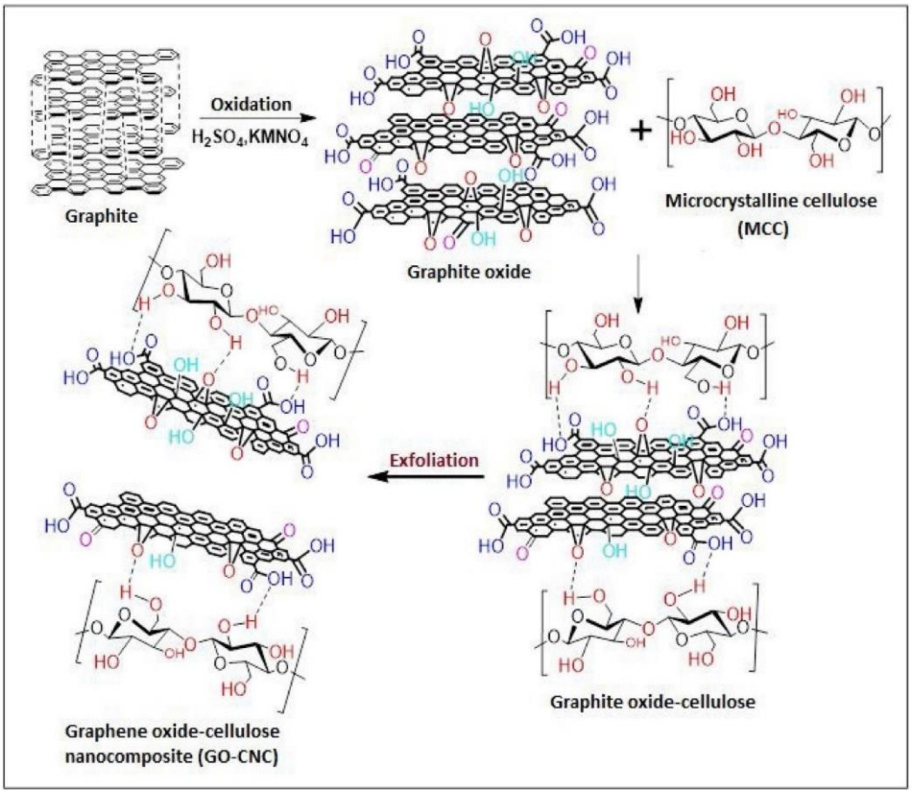

Figure 2 A graphical scheme showing the plausible mechanism for *in situ* formation of graphene oxide – cellulose nanocomposite (GO-CNC) [36]. Reproduced from ref [36] with permission [Facile one-pot in-situ synthesis of novel graphene oxide-cellulose nanocomposite for enhanced azo dye adsorption at optimized conditions - ScienceDirect].

3.2 Mechanical and Chemical Treatments

Mechanical and chemical treatments are essential processes for preparing cellulose microfibers from natural sources, which can then be combined with graphene to form hybrid materials with improved structural properties. These treatments involve a series of steps designed to extract and purify cellulose fibers, enhancing their compatibility with graphene [81, 82].

The mechanical treatment process typically begins with the physical manipulation of plant materials. This includes steps such as rolling, pressing, and separating the fibers from the plant matrix. These mechanical actions help to break down the plant structure and isolate the cellulose fibers. The goal is to achieve fibers with the desired size and morphology, which can effectively interact with graphene in the hybrid material [83-85].

Following the mechanical treatment, chemical treatments are employed to further purify the cellulose fibers. This often involves alkaline and acid treatments, which serve to remove impurities and non-cellulosic components such as lignin and hemicellulose. Alkaline treatment typically involves soaking the fibers in a sodium hydroxide solution, which helps to swell the fibers and remove unwanted materials. This is followed by an acid treatment, which can include the use of hydrochloric or sulfuric acid to further purify the cellulose and enhance its reactivity [86-88].

The resulting cellulose microfibers, now purified and possessing enhanced surface properties, are ready to be combined with graphene. The integration of graphene into the cellulose matrix can be achieved through various methods, such as in-situ synthesis or solvent-assisted dispersion. The enhanced structural properties of the cellulose-graphene hybrids are attributed to the strong interfacial interactions between the cellulose fibers and graphene sheets, which facilitate efficient stress transfer and improve the mechanical strength of the composite [7, 89, 90].

3.3 Green Synthesis

Green synthesis is an environmentally friendly approach to produce cellulose-graphene hybrid materials. This method leverages natural and sustainable resources and biological agents to synthesize graphene and integrate it with cellulose. The primary goal of green synthesis is to minimize the environmental impact and reduce the use of toxic chemicals typically involved in traditional synthesis methods [91, 92].

Using cow urine in green synthesis is considered environmentally friendly because it serves as a benign solvent and natural reducing agent, eliminating the need for synthetic reducing agents [53, 54].

Green synthesis generally begins with the preparation of graphene oxide (GO) from graphite. GO is then mixed with a cellulose solution or suspension. The reducing agent, such as cow urine, is added to this mixture. The natural constituents in the cow urine reduce the GO to rGO while the cellulose matrix stabilizes the rGO, preventing agglomeration and ensuring a uniform distribution within the matrix. The reaction typically occurs at mild temperatures and under ambient conditions, further emphasizing the eco-friendly nature of the process [38].

Green synthesis offers several advantages. Firstly, it is environmentally benign, as it avoids the use of toxic chemicals and harsh conditions [93]. Secondly, it is cost-effective, utilizing inexpensive and renewable resources [94]. Thirdly, the resulting cellulose-graphene hybrids often exhibit enhanced properties due to the gentle processing conditions, which preserve the integrity of both the cellulose and graphene components [95].

The applications of cellulose-graphene hybrids produced through green synthesis are diverse. In environmental remediation, these hybrids can be used in water purification systems due to their high adsorption capacities and biocompatibility. They are also suitable for use in biomedical applications, such as drug delivery systems and tissue engineering scaffolds, where the non-toxic and biodegradable nature of the materials is crucial. Additionally, the electrical conductivity and mechanical strength of these hybrids make them ideal for flexible electronics and energy storage devices, such as supercapacitors and batteries [96, 97].

For example, a study demonstrated the successful green synthesis of cellulose-graphene hybrids using cow urine as the reducing agent. The resulting hybrids exhibited improved thermal stability and electrical conductivity, making them suitable for use in electronic devices and thermal management systems. This example highlights the potential of green synthesis to produce high-performance cellulose-graphene hybrids with minimal environmental impact [98].

Figure 3 shows the desired characteristics of the synthesized nanomaterials (NMs) are influenced by their size and shape. Consequently, NMs are produced by adjusting several factors, including pH, temperature, reaction time, and the concentrations of both the metal salt and cow urine. Cow urine is readily combined with various concentrations of the chosen metal salt solutions at room temperature, leading to the formation of NMs within minutes through an efficient, single-step, sustainable, and environmentally friendly method [38].

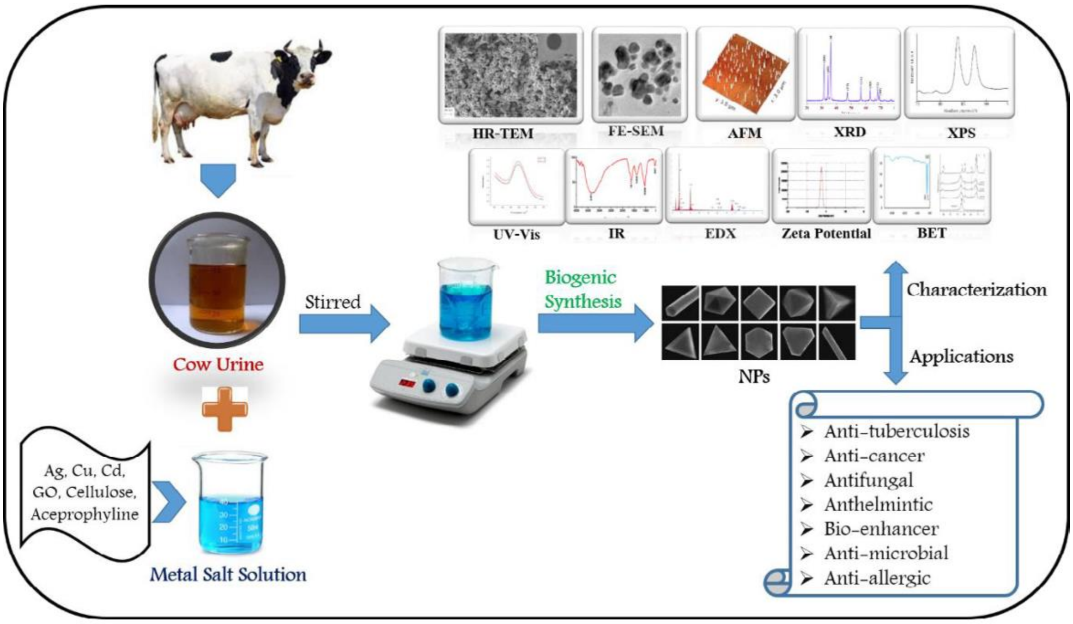

Figure 3 Biogenic synthesis of NMs from cow urine and their uses [38]. Reproduced from ref [38] with permission Open access [(PDF) Cow Urine Mediated Green Synthesis of Nanomaterial and Their Applications: A State-of-the-art Review (researchgate.net)].

3.4 Solvent-Assisted Methods

Solvent-assisted methods are a versatile and effective approach for synthesizing cellulose-graphene hybrid materials. These methods involve dispersing both graphene and cellulose in a solvent to achieve a homogeneous mixture, which is crucial for ensuring uniform distribution and interaction between the two components. The process often includes subsequent treatments such as annealing to enhance the material's properties [5, 99, 100].

In solvent-assisted synthesis, appropriate solvents are selected to effectively disperse both graphene oxide (GO) and cellulose. Common solvents include water, ethanol, and other organic solvents, chosen for their ability to suspend the materials without causing degradation. However, it is important to note that non-modified cellulose is not soluble in these mentioned solvents. Instead, cellulose is typically suspended rather than dissolved [101, 102].

For GO, it is first exfoliated in the solvent to ensure it is well-dispersed. This exfoliation can be achieved through mechanical means such as using a homogenizer or ultrasound [103-105]. These methods help in breaking down the GO sheets and dispersing them uniformly in the solvent. Similarly, cellulose is suspended in the same solvent to create a mixture suitable for forming cellulose-GO hybrids. The mechanical or ultrasonic treatment ensures that both components are adequately dispersed, facilitating their interaction in the solvent [106, 107].

Once both components are adequately dispersed, they are mixed to form a homogeneous solution or suspension. This mixture is then subjected to processes such as sonication or stirring to ensure thorough mixing and interaction between the cellulose and graphene oxide (GO). The goal is to achieve a uniform distribution of cellulose within the GO matrix, which is essential for the hybrid material's enhanced properties [108].

After achieving a homogeneous mixture, the next step often involves removing the solvent. This can be done through various methods such as evaporation, filtration, or freeze-drying. The removal of the solvent results in the formation of a solid composite material. To further enhance the properties of the hybrid material, annealing is commonly employed. Annealing involves heating the composite at controlled temperatures, which can improve the crystallinity and bonding between the cellulose and graphene, leading to better mechanical and electrical properties [109-111].

Solvent-assisted methods offer several advantages in the synthesis of cellulose-graphene hybrids. They allow fine-tuning the composite's properties by adjusting the solvent type, concentration of components, and processing conditions. These methods are relatively simple and can be scaled up for industrial production, making them suitable for large-scale synthesis.

The type of solvent used can significantly influence the final properties of the composite material. Different solvents interact with the components of a composite in various ways, affecting the dispersion and distribution of materials like cellulose and graphene.

Some solvents may leave residues that can alter the chemical properties of the composite, impacting its thermal stability, mechanical strength, or electrical properties. The solvent's boiling point and the rate of evaporation also play a critical role in determining the morphology and structure of the composite. Rapid evaporation might lead to porous structures, while slower evaporation can result in denser composites. Furthermore, the solvent type can affect the crystallinity of the composite materials, which in turn influences the mechanical and thermal properties. Solvents that promote better bonding between components can enhance the overall strength and stability of the composite [112-114].

The applications of cellulose-graphene hybrids synthesized through solvent-assisted methods are diverse. These materials are particularly useful in flexible electronics due to their enhanced electrical conductivity and mechanical flexibility [115]. They are also employed in energy storage devices such as supercapacitors and batteries, where the uniform distribution of graphene within the cellulose matrix contributes to higher energy densities and improved performance. Furthermore, the hybrids' improved thermal stability and conductivity make them suitable for applications in thermal management and environmental remediation, such as water purification and air filtration systems [116].

For example, a study demonstrated the successful synthesis of cellulose-graphene hybrids using a solvent-assisted method where GO and cellulose were dispersed in water, followed by sonication and freeze-drying. The resulting hybrids exhibited significantly improved thermal properties and electrical conductivity, making them suitable for use in electronic devices and thermal management systems. This example highlights the effectiveness of solvent-assisted methods in producing high-performance cellulose-graphene hybrids with tailored properties [117].

3.5 Hydrothermal and Solvothermal Methods

Hydrothermal and solvothermal methods are advanced techniques used for synthesizing cellulose-graphene hybrid materials. These methods involve the use of high-temperature and high-pressure conditions to facilitate the reaction between cellulose and graphene precursors, resulting in hybrid structures with enhanced properties, as explained below.

In the hydrothermal method, the cellulose and graphene oxide (GO) are mixed in a solvent, typically water, and the mixture is sealed in an autoclave. The autoclave is then heated to high temperatures, often ranging from 120°C to 250°C, under autogenous pressure. This environment promotes the reduction of GO to reduced graphene oxide (rGO) and facilitates the integration of graphene into the cellulose matrix. The high temperature and pressure conditions enhance the crystallinity and bonding between the cellulose and graphene, resulting in a composite material with improved mechanical and electrical properties [118].

Similarly, the solvothermal method follows a comparable process but uses organic solvents instead of water. This method can offer additional control over the reaction environment, allowing for the synthesis of materials with specific characteristics. For instance, solvothermal synthesis can be used to produce graphene nanocomposites with tailored properties for catalytic applications, as demonstrated in studies where graphene was combined with other materials to enhance catalytic efficiency [119].

The advantages of hydrothermal and solvothermal methods include the ability to produce materials with high purity and uniformity. These methods also allow for the fine-tuning of the material properties by adjusting the reaction conditions, such as temperature, pressure, and solvent type. Moreover, they are relatively simple and can be scaled up for industrial production, making them suitable for large-scale synthesis of cellulose-graphene hybrids [120].

Cellulose-graphene hybrids produced through these methods have a wide range of applications. In energy storage, they are used in devices such as supercapacitors and batteries, where their enhanced electrical conductivity and mechanical strength contribute to higher energy densities and improved performance.

In the context of cellulose-graphene hybrids synthesized via hydrothermal and solvothermal methods for supercapacitor applications, these methods are particularly effective due to the high-temperature and high-pressure conditions they employ. These conditions facilitate the reduction of graphene oxide (GO) to reduced graphene oxide (rGO) and promote the integration of graphene into the cellulose matrix, enhancing the mechanical and electrical properties of the resulting composite material. Hydrothermal synthesis involves mixing cellulose and GO in a solvent, typically water, and sealing the mixture in an autoclave. The autoclave is then heated to high temperatures, often between 120°C and 250°C, under autogenous pressure. This environment not only reduces GO to rGO but also ensures strong bonding and improved crystallinity between the cellulose and graphene, resulting in a composite with superior mechanical and electrical properties [121, 122].

Similarly, the solvothermal method uses organic solvents instead of water, providing additional control over the reaction environment. This method can be tailored to produce materials with specific characteristics, such as enhanced catalytic efficiency or tailored electrochemical properties, which are crucial for high-performance supercapacitors [123].

The choice between hydrothermal and solvothermal methods depends on the desired properties of the final material and the specific application requirements. Both methods have demonstrated the ability to produce cellulose-graphene hybrids with high electrical conductivity and mechanical strength, making them suitable for use in supercapacitors and other energy storage devices. The enhanced properties of these hybrids contribute to higher energy densities and improved performance in supercapacitor applications.

In environmental applications, these hybrids are employed in water purification and air filtration systems due to their high adsorption capacities and thermal stability. Additionally, the unique properties of these materials make them suitable for use in flexible electronics and thermal management systems [79].

For example, a study demonstrated the successful hydrothermal synthesis of cellulose-graphene hybrids, resulting in materials with significantly improved thermal properties and electrical conductivity. These hybrids were found to be highly effective in applications such as electronic devices and thermal management systems, highlighting the potential of hydrothermal and solvothermal methods in producing high-performance cellulose-graphene hybrids [42].

3.6 Electrochemical Exfoliation

Electrochemical exfoliation is a method used to produce graphene from graphite, which can then be combined with cellulose to form hybrid materials. This technique involves applying an electric field to graphite in an electrolyte solution, leading to the exfoliation of graphene layers. The process is known for its efficiency and ability to produce high-quality graphene with fewer defects compared to other methods [124, 125].

The electrochemical exfoliation process typically begins with the preparation of a three-electrode electrochemical cell. Graphite rods or other graphite-based materials serve as the working electrode, while a platinum wire and a reference electrode (such as Ag/AgCl) complete the setup. The electrolyte solution, often sulfuric acid or a mixture of sulfuric acid and other salts, facilitates the exfoliation process [126].

When an electric field is applied, the process involves two main steps: intercalation and exfoliation. During the intercalation step, ions from the electrolyte penetrate the graphite layers at a lower potential. This intercalation weakens the van der Waals forces holding the layers together. In the subsequent exfoliation step, a higher potential is applied, causing the layers to separate and form graphene sheets. The exfoliated graphene can then be collected, washed, and further processed [127].

One of the significant advantages of electrochemical exfoliation is that it allows to produce of graphene with controlled properties. By adjusting the applied potential, electrolyte composition, and reaction time, researchers can influence the degree of oxidation, defect density, and crystallite size of the resulting graphene. For instance, using a sulfuric acid solution and applying anodic polarization can yield graphene oxide (GO) with varying degrees of oxidation and structural properties, as characterized by techniques such as cyclic voltammetry (CV), electrochemical impedance spectroscopy (EIS), X-ray photoelectron spectroscopy (XPS), Raman spectroscopy, transmission electron microscopy (TEM), and atomic force microscopy (AFM) [128, 129].

The electrochemical exfoliation method is also known for its scalability and environmental friendliness. It avoids the use of harsh chemicals and high temperatures, making it a more sustainable option for large-scale graphene production. Additionally, the process can be tailored to produce graphene derivatives such as graphene oxide (GO) and reduced graphene oxide (rGO), which are essential for various applications [130, 131].

Cellulose-graphene hybrids produced through electrochemical exfoliation have numerous applications. In energy storage, these hybrids are used in supercapacitors and batteries, where their enhanced electrical conductivity and mechanical strength contribute to higher energy densities and improved performance. In environmental remediation, they are employed in water purification and air filtration systems due to their high adsorption capacities and thermal stability. Additionally, the hybrids' unique properties make them suitable for use in flexible electronics, sensors, and biomedical devices [132, 133].

For example, a study demonstrated the successful synthesis of manganese oxide/graphene composites via a plasma-enhanced electrochemical exfoliation process. This method produced graphene decorated with manganese oxide nanoparticles, which exhibited high specific capacitance, making them suitable for supercapacitor electrode materials. Another study highlighted the production of size-controlled graphene oxide materials using electrochemical exfoliation, which were then applied in situ for gold nanoparticle formation and electrochemical sensors [134].

3.7 Layer-by-Layer Assembly

Layer-by-layer (LbL) assembly is a precise and controlled technique used to fabricate cellulose-graphene hybrid materials. This method involves the sequential deposition of alternate layers of cellulose and graphene, allowing for meticulous control over the thickness, composition, and properties of the resulting material. The LbL assembly process is particularly advantageous for applications requiring tailored material characteristics, such as specific mechanical, electrical, or thermal properties [135, 136].

The LbL assembly process begins with the preparation of individual solutions or suspensions of cellulose and graphene oxide (GO). The substrate, which can be a solid surface or a flexible film, is alternately immersed in these solutions. During each immersion, a monolayer of the respective material is adsorbed onto the substrate. The adsorption process is driven by various interactions, including electrostatic forces, hydrogen bonding, and van der Waals forces, depending on the surface chemistry of the materials involved [135].

After each layer deposit, the substrate is rinsed to remove any loosely bound material, ensuring that only a single, uniform layer remains. This cycle of immersion and rinsing is repeated until the desired number of layers is achieved. The precise control over the number of layers allows for the fine-tuning of the hybrid material's properties, making it possible to design materials with specific performance characteristics [137, 138].

One of the significant advantages of LbL assembly is the ability to create highly ordered structures with controlled thickness at the nanoscale. This precision is crucial for applications in which the material's performance is highly dependent on its microstructure. Additionally, the LbL method is versatile and can be adapted to incorporate various functional materials, enabling the creation of multifunctional composites [139].

Cellulose-graphene hybrids produced through LbL assembly are used in a variety of applications [10]. In flexible electronics, these materials provide the necessary mechanical flexibility and electrical conductivity for use in sensors, displays, and other electronic devices. In energy storage, precise control over the material's structure enhances the performance of supercapacitors and batteries, contributing to higher energy densities and improved charge-discharge cycles. Furthermore, the tailored properties of these hybrids make them suitable for use in thermal management systems, where efficient heat dissipation is critical [140].

For example, a study demonstrated the successful fabrication of cellulose-graphene hybrid films using LbL assembly, resulting in materials with enhanced mechanical strength and electrical conductivity. These films were found to be highly effective in applications such as flexible electronics and energy storage devices, highlighting the potential of LbL assembly in producing high-performance cellulose-graphene hybrids [135].

Figure 4 illustrates the layer-by-layer (LBL) assembly process used to create hybrid films by sequentially depositing Nano fibrillated cellulose (NFC) and GO nanosheets on an ultrathin NFC substrate, followed by a chemical reduction to convert GO to RGO. This method effectively combines the unique properties of both materials to form a composite structure. Figures 4b and 4c depict the distinct morphologies of the two components: the 1D morphology of NFC and the 2D morphologies of GO. Despite both being slightly negatively charged and water-soluble, NFC and GO were successfully utilized in the LBL technique [135].

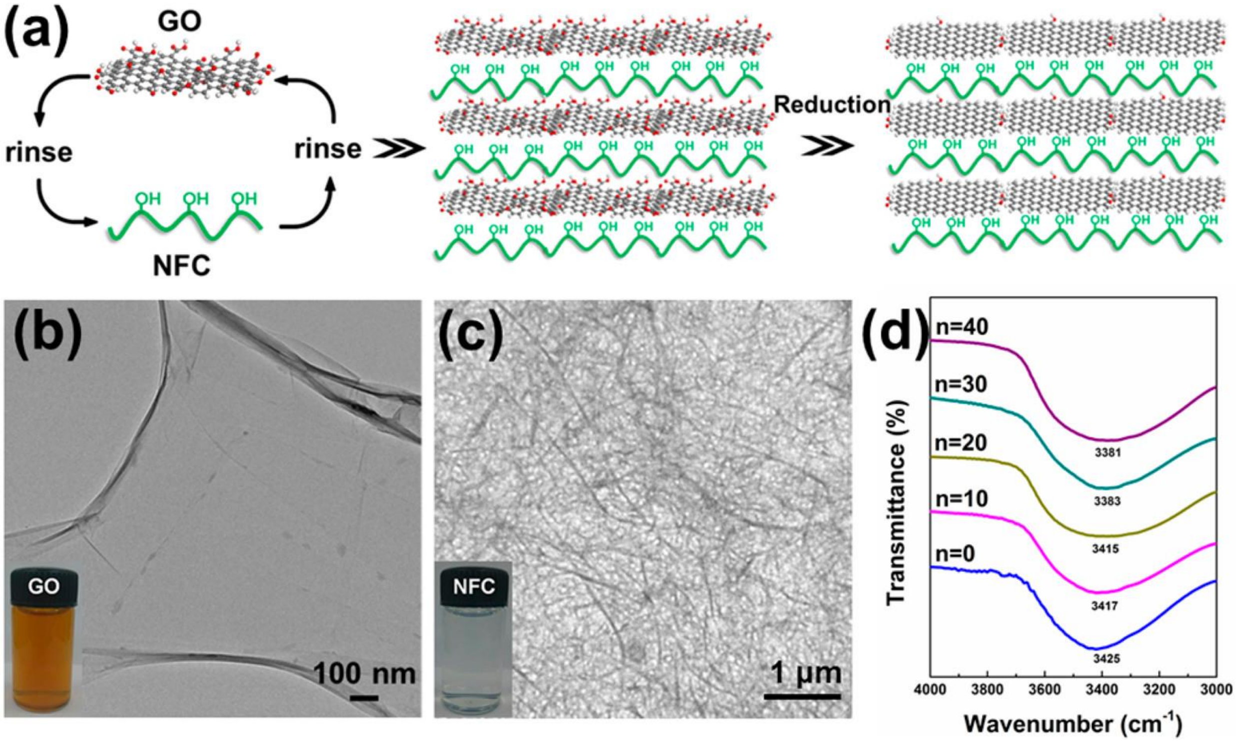

Figure 4 LbL-assembled (NFC/RGO)n hybrid films. (a) Schematic drawing of the preparation of the hybrid films. Fabrication of the (NFC/RGO) hybrid film architecture includes two steps: The first step is the sequential assembly of NFC and GO on a flexible NFC substrate. The second step is the reduction of GO. (b) TEM image of GO. (Insert) Photograph of GO dispersion. (c) TEM image of NFC. (Insert) Photograph of NFC suspension. (d) FTIR spectra of the (NFC/GO)n hybrid films at wavenumbers 3100−3500 cm^{-1} showing the vibration of −OH groups [135]. Copyright with permission [Highly Anisotropic Thermal Conductivity of Layer-by-Layer Assembled Nano fibrillated Cellulose/Graphene Nanosheets Hybrid Films for Thermal Management | ACS Applied Materials & Interfaces].

3.8 Freeze-Drying

Freeze-drying, also known as lyophilization, is a crucial technique in the synthesis of cellulose-graphene hybrid materials, particularly for creating aerogels. This process involves freezing a mixture of cellulose and graphene and then sublimating the solvent, resulting in a porous, lightweight hybrid material with a high surface area and excellent mechanical properties [141].

The freeze-drying process begins with the preparation of a homogeneous mixture of cellulose and graphene oxide (GO) in a suitable solvent, typically water. The mixture is then rapidly frozen, often using liquid nitrogen or a specialized freeze-dryer. Freezing the mixture solidifies the solvent and traps the dispersed cellulose and graphene within the ice matrix [142].

Once the mixture is completely frozen, it undergoes sublimation in a freeze-dryer. Sublimation is the process of converting the solid solvent (ice) directly into vapor without passing through the liquid phase. This is achieved by lowering the pressure and gently heating the frozen mixture. As the ice sublimates, it leaves behind a porous structure composed of cellulose and graphene. The resulting aerogel retains the original structure of the frozen mixture, with a high surface area and interconnected porosity [143].

One of the significant advantages of freeze-drying is that it preserves the delicate structure of the cellulose-graphene hybrid material, maintaining its high surface area and porosity. This is particularly important for applications that require lightweight materials with high mechanical strength and large surface areas, such as energy storage, catalysis, and environmental remediation. Cellulose-graphene aerogels produced through freeze-drying exhibit several enhanced properties. They have excellent mechanical properties, including high compressive strength and elasticity, making them suitable for use in structural applications. Their high surface area and porosity also make them ideal for adsorption applications, such as water purification and air filtration, where they can effectively capture and remove contaminants [143, 144].

In energy storage applications, these aerogels can be used as electrodes in supercapacitors and batteries. The high surface area facilitates efficient charge storage, while the porous structure allows for rapid ion transport, leading to improved performance. Additionally, the lightweight nature of the aerogels makes them suitable for portable and flexible energy storage devices [145, 146].

For example, a study demonstrated the successful synthesis of cellulose-graphene aerogels using freeze-drying. The resulting aerogels exhibited a high specific surface area and excellent mechanical properties, making them suitable for use in various applications, including thermal insulation and electromagnetic interference (EMI) shielding [147-149]. This example highlights the effectiveness of freeze-drying in producing high-performance cellulose-graphene hybrid materials with tailored properties.

3.9 Plasma-Enabled Techniques

Plasma-enabled techniques emerge as a sustainable and efficient approach for synthesizing graphene-based materials, including cellulose-graphene hybrids. These methods utilize plasma to assemble nanostructures from gaseous precursors into solid forms, allowing for the design of high-quality graphene-based nanostructures with controlled morphology and orientation. The versatility and environmental friendliness of plasma-enabled techniques make them particularly attractive for advanced material synthesis [150].

The process of plasma-enabled synthesis typically involves a plasma reactor where a gas containing carbon (such as methane or acetylene) is ionized to create a plasma state. This plasma state provides the energy necessary to break down the gas molecules into reactive species, which then recombine on a substrate to form graphene. The conditions within the plasma reactor, such as temperature, pressure, and gas composition, can be precisely controlled to tailor the properties of the resulting graphene.

One notable plasma-enabled technique is the single-step atmospheric pressure plasma method [150]. This approach uses microwave plasma at atmospheric conditions to design and engineer graphene-based nanostructures. This method is advantageous because it operates at lower temperatures and pressures compared to traditional chemical vapor deposition (CVD) techniques, reducing energy consumption and making the process more environmentally friendly. Additionally, the single-step nature of the process simplifies the synthesis, making it more scalable and cost-effective [151].

A significant advantage of plasma-enabled techniques is the ability to produce graphene with fewer defects and higher crystalline. The high-energy environment of the plasma facilitates the formation of well-ordered graphene structures, which are essential for applications requiring high electrical conductivity and mechanical strength. Furthermore, plasma processes can be used to functionalize graphene surfaces, enhancing their compatibility with cellulose and other materials [152].

Cellulose-graphene hybrids synthesized using plasma-enabled techniques have shown promise in various applications. In energy storage, these hybrids are used in supercapacitors and batteries, where high-quality graphene enhances the electrical conductivity and mechanical strength of the electrodes, leading to improved performance. In environmental applications, plasma-synthesized graphene can be combined with cellulose to create materials with high adsorption capacities for water purification and air filtration. For instance, a study demonstrated the use of plasma-enabled techniques to synthesize graphene hybrids for energy storage applications. The resulting materials exhibited high structural quality and controllability, making them suitable for use in supercapacitors with enhanced energy densities and charge-discharge cycles. Another study explored the use of plasma-assisted functionalization to create graphene-polymer hybrids, which showed improved biocompatibility and potential for biomedical applications such as wound dressings and tissue engineering [150].

4. Compatibility and Applications of Cellulose and Graphene Composites

Cellulose and graphene exhibit distinct compatibility profiles with various matrix types, significantly impacting their mechanical properties and potential applications. This comprehensive overview explores their compatibility with inorganic matrices, polar polymers, and non-polar polymers, highlighting the resulting mechanical qualities and uses [153].

4.1 Cellulose Compatibility

4.1.1 Polar Polymers

Cellulose demonstrates excellent compatibility with polar polymers due to its hydrophilic nature and abundance of hydroxyl groups, leading to enhanced mechanical properties, improved dispersion, and increased thermal stability [154, 155]. This compatibility results in composites with improved tensile strength and modulus, uniform distribution within the matrix, and higher degradation temperatures. These characteristics make cellulose-polar polymer composites ideal for applications in packaging materials, biomedical devices, and water treatment membranes.

A study demonstrated the successful fabrication of cellulose microfibers from Vietnamese Nipa palm and their reinforcement with reduced graphene oxide. This hybrid material exhibited improved thermal properties and electrical conductivity, making it suitable for electronic devices and thermal management systems [70]. Similar improvements have been observed in other cellulose-based composites, such as bacterial cellulose films reinforced with NH_4I-doped graphene oxide, which showed increased ionic conductivity [156].

4.1.2 Non-Polar Polymers

In contrast, cellulose exhibits poor compatibility with non-polar polymers, resulting in weak interfacial adhesion, agglomeration, and limited property enhancement [157]. This incompatibility leads to reduced mechanical strength, non-uniform dispersion, and minimal improvement in overall performance. To overcome these limitations, surface modification of cellulose is often required for better compatibility with non-polar matrices, expanding potential applications.

4.1.3 Inorganic Matrices

Cellulose can be incorporated into inorganic matrices such as ceramics and cement with varying degrees of success [158, 159]. In these composites, cellulose acts as a reinforcing agent, improving fracture toughness in brittle matrices and influencing the pore structure of inorganic materials. These properties make cellulose-inorganic hybrids suitable for applications in bone tissue engineering scaffolds and cement composites for construction.

4.2 Graphene Compatibility

4.2.1 Non-Polar Polymers

Graphene generally exhibits good compatibility with non-polar polymers due to its hydrophobic nature, resulting in excellent mechanical reinforcement, enhanced electrical conductivity, and increased thermal stability [159]. These properties lead to significant increases in strength and stiffness, improved electron transport properties, and enhanced heat resistance. Graphene-non-polar polymer composites find applications in conductive polymer composites and electromagnetic shielding materials.

Research by Mustapha et al. (2023) on unsaturated polyester reinforced with kenaf core fiber and hybrid cellulose nanocrystal (CNC) and graphene nanoplatelet (GNP) nanofillers displays the potential of graphene in enhancing the mechanical properties of non-polar polymer composites [160].

4.2.2 Polar Polymers

Graphene's compatibility with polar polymers can be challenging, often resulting in poor dispersion, weak interfacial bonding, and inconsistent property enhancement [161]. To overcome these issues, functionalization of graphene is frequently employed to improve compatibility, leading to better dispersion and stronger interactions with polar matrices. This approach expands the potential applications of graphene-polar polymer composites.

4.2.3 Inorganic Matrices

Graphene can be effectively incorporated into various inorganic matrices, providing mechanical reinforcement, enhanced electrical properties, and improved thermal management. These characteristics result in significant improvements in strength and toughness, enhanced conductivity in traditionally insulating materials, and improved heat dissipation. Graphene-inorganic composites find applications in high-performance ceramics, conductive concrete, and advanced coating materials [162]. Graphene nanosheet (GN) reinforced alumina ceramics demonstrate remarkable enhancements in both mechanical and electromagnetic properties: The addition of graphene nanosheets as a reinforcing phase contributes to excellent mechanical properties in GN/Al_2O_3 ceramics and exhibits significantly improved electrical conductivity compared to pure alumina. These composites show tunable electromagnetic properties, good microwave absorption, and electromagnetic interference (EMI) shielding performance in the X-band frequency range (8.2-12.4 GHz) [161].

4.3 Hybrid Cellulose-Graphene Systems

Recent research has focused on combining cellulose and graphene to create hybrid nanocomposites that leverage the strengths of both materials [163, 164]. These hybrids exhibit synergistic effects, including enhanced mechanical, thermal, and electrical properties. The combination of cellulose's hydrophilicity and graphene's conductivity results in multifunctional properties, making these hybrids suitable for applications in smart packaging, flexible electronics, and advanced filtration membranes.

A study by Liu et al. (2022) on compressible cellulose nanofibrils/reduced graphene oxide composite carbon aerogels for solid-state supercapacitors demonstrates the synergistic effects of combining cellulose and graphene in energy storage applications [165].

To highlight the applications of cellulose-graphene hybrids in sensing, the work of Wu et al. (2019) on epoxy composites with reduced graphene oxide-cellulose nanofiber hybrid filler for concrete strain and crack monitoring showcases the potential of these hybrids in structural health monitoring applications [65].

The development of cellulose-graphene hybrid materials aligns closely with Sustainable Development Goal 7 (SDG 7): Affordable and Clean Energy [166]. These innovative materials have significant potential to contribute to the advancement of clean energy technologies and energy efficiency, which are key targets of SDG 7.

4.4 Cellulose-Graphene Hybrids and SDG 7

4.4.1 Energy Storage Applications

Cellulose-graphene hybrids show great promise in energy storage devices such as supercapacitors and batteries. The enhanced electrical conductivity and mechanical properties of these materials contribute to higher energy densities and improved performance in energy storage applications. This directly supports SDG 7's target of ensuring access to affordable, reliable, and modern energy services [167]. A novel type of flexible, all-solid-state supercapacitor using cellulose nanofibril (CNF)/reduced graphene oxide (RGO)/carbon nanotube (CNT) hybrid aerogels as electrodes exhibited a high specific capacitance of 252 F g^{-1} at a discharge current density of 0.5 A g^{-1}. These supercapacitors showed outstanding cycle stability, retaining more than 99.5% of their capacitance after 1000 charge-discharge cycles at a current density of 1 A g^{-1} The devices also demonstrated high areal capacitance (216 mF cm^{-2}), areal power density (9.5 mW cm^{-2}), and energy density (28.4 µWh cm^{-2}) [115].

The thermal management capabilities of cellulose-graphene hybrids can lead to more efficient energy systems. By enhancing heat dissipation in electronic devices and other energy-consuming technologies, these materials can contribute to overall energy efficiency improvements, aligning with SDG 7's emphasis on doubling the global rate of improvement in energy efficiency [147, 167]. Nano fibrillated cellulose/reduced graphene oxide (NFC/RGO) hybrid films exhibit remarkable anisotropic thermal conductivity, with in-plane thermal conductivity reaching up to 12.6 $W \cdot m^{-1} \cdot K^{-1}$. The layer-by-layer assembly technique creates highly anisotropic structures, with thermal conductivity anisotropy ($\lambda X/\lambda Z$) reaching up to 279 [135]. This property enables directional heat transfer, which can be particularly useful in managing heat flow in complex electronic systems.

Cellulose-graphene hybrids have potential applications in solar cells and other renewable energy technologies [168]. Their unique properties can enhance the performance and durability of renewable energy devices, supporting SDG 7's target of increasing the share of renewable energy in the global energy mix. The use of cellulose, a renewable and biodegradable resource, in these hybrid materials supports the sustainable production aspect of SDG 7. Green synthesis methods, such as those using cow urine as a reducing agent, further contribute to the goal's focus on clean energy by minimizing environmental impact in material production.

By enabling more efficient and sustainable energy technologies, cellulose-graphene hybrids can indirectly contribute to reducing greenhouse gas emissions, which is crucial for combating climate change – a key consideration in the broader context of sustainable development goals [169].

5. Characterization Techniques

The characterization of cellulose-graphene hybrid materials involves a variety of sophisticated techniques to thoroughly understand their structural, morphological, thermal, mechanical, and chemical properties.

5.1 Structural Characterization

5.1.1 X-ray Diffraction (XRD)

XRD is used to determine the crystalline structure of hybrid materials. For instance, XRD patterns can confirm the presence of graphene nanoplatelets and cellulose nanocrystals by identifying their characteristic diffraction peaks. This technique is crucial in verifying the successful integration of graphene within the cellulose matrix.

The crystalline structure of the prepared samples was analyzed using X-ray diffraction patterns of cellulose, shown in Figure 5. The sample patterns of BC display distinct peaks at $2\theta = 14.2°$, 23.1°, and a minor peak at 28°, corresponding to the diffraction planes (110) and (200) of cellulose I. BC/GO shows similar diffraction peaks at $2\theta = 14.1°$, 22.8°, and a minor peak at 28°, with slightly higher intensity and no noticeable peak shifts or new peaks. This increased intensity is due to the incorporation of GO onto the BC surface, where GO is uniformly distributed within the BC matrix. This indicates the physicochemical modification of BC, enhancing its crystallinity by approximately 20–30%. The chemical modification is believed to occur on the surface of the cellulose fibers [170].

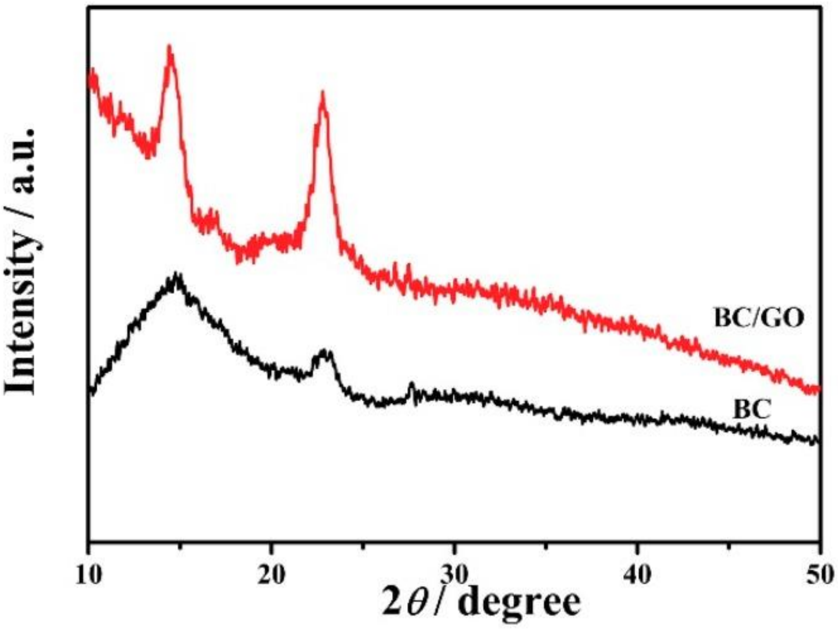

Figure 5 XRD patterns of GO, extracted cellulose, and GO-cellulose composite [170]. Open access [Polymers | Free Full-Text | Development and Evaluation of Cellulose/Graphene-Oxide Based Composite for Removing Phenol from Aqueous Solutions (mdpi.com)].

5.2 Morphological Characterization

5.2.1 Scanning Electron Microscopy (SEM)

The scanning electron microscopy (SEM) images in Figure 6 reveal the distinct morphological characteristics of the CGC (cellulose-graphene oxide composite). In contrast to the smooth, homogeneous surface of pure cellulose, the CGC exhibits a rough surface with a lamellar structure and homogeneous 3D porous structures. This unique morphology is attributed to the introduction of reduced graphene oxide (RGO) and its supporting effect during the coagulation process. The rough, wrinkled surfaces and porous structures beneath the sample surface serve as active adsorption sites, creating favorable conditions for attracting triazine pesticides and significantly enhancing both the adsorption rate and capacities of the CGC [171].

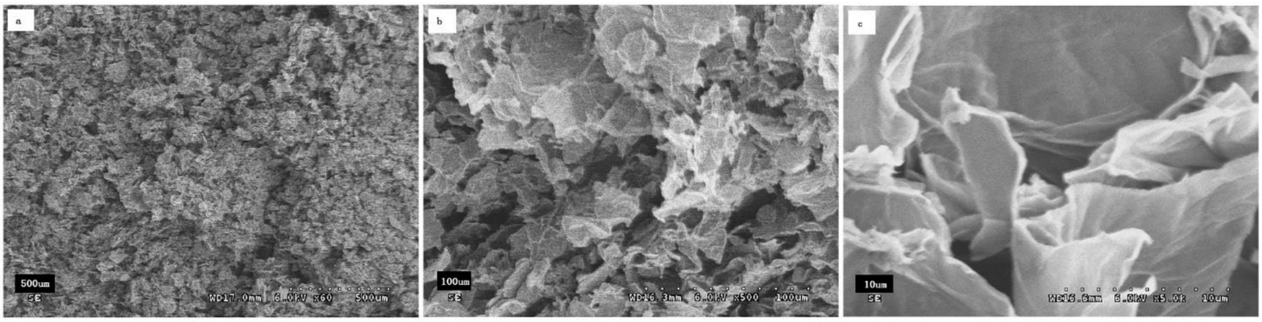

Figure 6 SEM images of the different morphologies of CGC sheets: (a) scale = 500 um, (b) scale = 100 um, and (c) scale = 10 um [171].

5.3 Thermal Characterization

5.3.1 Thermogravimetric Analysis (TGA)

TGA measures the weight loss of a material as a function of temperature, providing insights into its thermal stability and decomposition behavior. This technique is essential for assessing the thermal properties of hybrid materials, such as their resistance to thermal degradation [172].

5.3.2 Surface Area and Porosity

Brunauer-Emmett-Teller (BET) Surface Area Analysis: BET. When applying the Brunauer-Emmett-Teller (BET) surface area analysis to hybrid materials, such as cellulose-graphene composites, it is essential to consider the unique structural complexities these materials present. The BET method is traditionally used to measure the specific surface area and porosity, which are crucial for applications where surface interactions are vital, like adsorption and catalysis. A higher surface area generally correlates with improved performance in these applications. Porosity doesn’t appear in the original BET equation and additional assumptions are needed to obtain it. Usually, it is assumed that the pores have a cylindrical shape.

BET Analysis for Hybrid Materials.

1. Traditional BET Parameters: For single materials, the BET theory relies on two primary parameters: the specific surface area and a nitrogen/surface interaction parameter. This model assumes a uniform surface interaction, which simplifies the analysis but may not be suitable for complex materials.
2. Challenges with Hybrid Materials: Hybrid materials, such as cellulose-graphene composites, consist of multiple components with distinct surface properties. This complexity requires modifications to the BET analysis to accurately reflect the interactions and surface areas of each component.
3. Required Modifications: To properly analyze hybrid materials, the BET model should be adapted to include two nitrogen/surface interaction parameters and two surface areas. This approach accounts for the heterogeneous nature of hybrid materials, ensuring that the analysis captures the true surface interactions and porosity.
4. Reliability Concerns: If a study assumes a uniform structure for hybrid materials and applies the common BET equation without modifications, the results may be unreliable. The assumption of uniformity can lead to inaccuracies in the calculated surface area and porosity, potentially misrepresenting the material's properties and performance capabilities.

In summary, while the BET method is a powerful tool for assessing surface area and porosity, its application to hybrid materials demands careful consideration and modification to ensure accurate characterization. For cellulose-graphene hybrids and similar composites, adapting the BET analysis to account for their complex structures is crucial for obtaining reliable and meaningful results [173-176].

5.4 Mechanical Characterization

5.4.1 Compression Testing

This method evaluates the mechanical strength and compressive properties of hybrid materials. For instance, hybrid organic aerogels consisting of polyvinyl alcohol, cellulose nanofibrils, and graphene oxide have shown high specific compressive strength and failure strain, making them suitable for applications requiring robust mechanical properties [141].

5.5 Chemical Characterization

5.5.1 Fourier Transform Infrared Spectroscopy (FTIR)

FTIR is used to identify the functional groups present in hybrid materials. It helps in understanding the chemical interactions between cellulose and graphene components. For example, FTIR analysis can reveal the presence of hydroxyl, carbonyl, and other functional groups that indicate successful chemical bonding between the components [70].

According to Figure 7a, In the study of Bacterial Cellulose/Polyvinyl Alcohol/Graphene Oxide/Attapulgite (BC/PVA/GO/APT) composites were successfully prepared via a repeated freeze-thaw method using bacterial cellulose, polyvinyl alcohol as the skeleton, and graphene oxide, attapulgite as fillers. The infrared spectrum of Attapulgite (APT) shows peaks at 3555 cm^{-1} and 3453 cm^{-1}, corresponding to Al–OH and Mg–OH stretching vibrations, respectively, and peaks at 1018 cm^{-1} and 474 cm^{-1} for Si–O and Si–O–Si bonds. For BC, the broad peak at 3373 cm^{-1} is due to intermolecular and intramolecular group stretching vibrations, while peaks at 2894, 1047, and 1431 cm^{-1} correspond to CH_2–CH, C–O, and C–H bending vibrations, with a glycosidic bond peak at 902 cm^{-1}. The FTIR spectrum of Polyvinyl Alcohol (PVA) shows a broad O–H stretching peak at 3416 cm^{-1}, and peaks at 2942, 1452, and 1096 cm^{-1} for C–H and C–O–C bonds and C–H bending. The FTIR spectrum of GO displays characteristic peaks for OH (3420–3606 cm^{-1}), –C=O– (1750–1850 cm^{-1}), –COOH (1650–1750 cm^{-1}), and C=C (1500–1600 cm^{-1}). In Figure 7b, the BC/PVA (BP)spectrum shows a peak at 3341 cm^{-1} for intermolecular and intramolecular hydrogen bonds between OH groups in Bacterial Cellulose (BC) and PVA. No new peaks appear in the composite after adding GO and APT, but the CH_2–CH peak in BC overlaps with the C–H peak in Polyvinyl Alcohol, and the OH peak broadens and shifts to lower wavenumbers, indicating hydrogen bond formation between polymers as shown in Figure 7c [132].

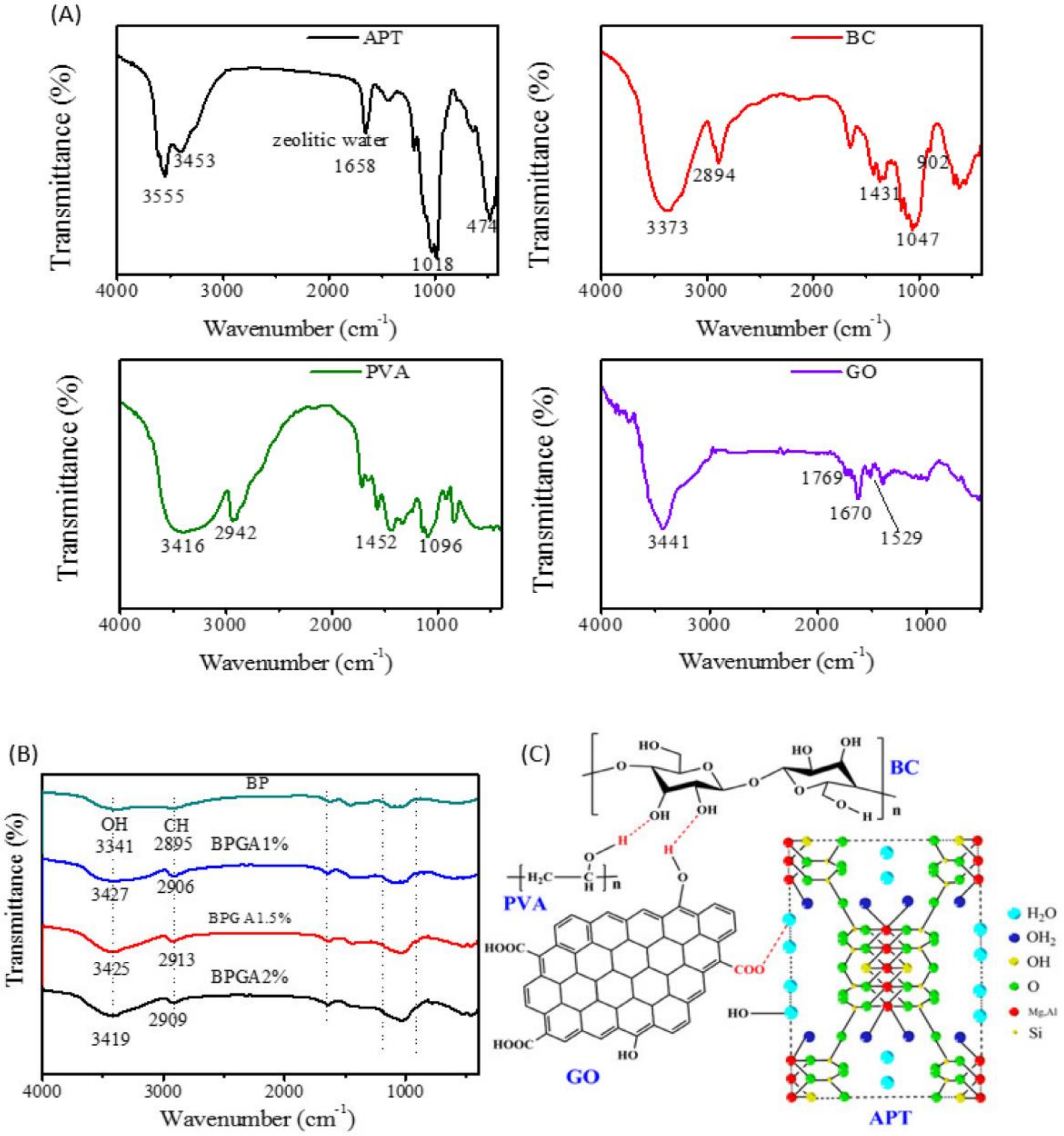

Figure 7 The FTIR spectra of initial components and prepared adsorbent materials. (a) represents the FTIR spectra of initial components Bacterial Cellulose/Polyvinyl Alcohol/Graphene Oxide/Attapulgite and (b) represents the prepared adsorbent materials (BP, BPGA 1%, BPGA 1.5%, and BPGA 2%), (c) represents a schematic diagram of the hydrogen bonding of the initial components [132]. Open access [Materials | Free Full-Text | Synthesis and Adsorption Properties of Novel Bacterial Cellulose/Graphene Oxide/Attapulgite Materials for Cu and Pb Ions in Aqueous Solutions (mdpi.com)].

5.6 Additional Characterization Techniques

5.6.1 High-Resolution Transmission Electron Microscopy (HR-TEM)

HR-TEM provides atomic-level images of hybrid materials, offering insights into their internal structure and the distribution of graphene within the cellulose matrix. This technique can reveal the integration of graphene at the interlayer spacing of other materials, enhancing their properties [177].

5.6.2 Contact Angle Measurements

This technique assesses the hydrophobicity or hydrophilicity of the hybrid materials. It is particularly useful for applications involving water interaction, such as in hydrophobic membranes or moisture-resistant aerogels [178, 179].

5.6.3 Differential Scanning Calorimetry (DSC)

DSC measures the heat flow associated with phase transitions in the material, providing information on its thermal properties such as melting and crystallization temperatures. This is crucial for understanding the thermal behavior of hybrid materials under different conditions [180, 181].

These characterization techniques collectively provide a comprehensive understanding of cellulose-graphene hybrid materials, enabling their optimization for various multifunctional applications such as thermal insulation, electromagnetic interference shielding, and biomedical uses. The integration of these advanced techniques ensures a thorough evaluation of the material properties, paving the way for innovative applications and improved performance.

6. Properties of Cellulose-Graphene Hybrids

Cellulose-graphene hybrid materials exhibit a unique combination of properties derived from both cellulose and graphene, making them highly attractive for various advanced applications. These properties include mechanical, thermal, electrical, surface, structural, chemical, biocompatibility, biodegradability, and optical characteristics.

6.1 Mechanical Properties

The integration of graphene oxide (GO) into cellulose fibers significantly enhances the mechanical properties of the hybrid material. Notably, the tensile strength of bacterial cellulose/graphene oxide (BC/GO) hybrids can reach up to 95.68 MPa, a substantial improvement over pure cellulose. Additionally, Young's modulus, which measures the stiffness of the material, is also enhanced, with BC/GO hybrids exhibiting a Young's modulus of 12.87 GPa. These improvements are attributed to the strong interfacial interactions between cellulose and graphene, which facilitate efficient stress transfer [85]. Table 2 summarizes the key mechanical properties of various cellulose-graphene hybrids.

Table 2 Mechanical Properties of Cellulose-Graphene Hybrids.

Property	Hybrid Composition	Value	Reference
Tensile Strength	poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV)/1 wt% covalent CNC-GO	Increased by 170.2% compared to neat PHBV	[41]

Elongation to Break	PHBV/1 wt% covalent CNC-GO	Increased by 52.1% compared to neat PHBV	[41]
Flexural Strength	Epoxy with 0.5 wt% CNFs and 0.1 wt% GNPs	Improved compared to neat epoxy	[182]
Storage Modulus	Epoxy with 0.5 wt% CNFs and 0.1 wt% GNPs	Improved compared to neat epoxy	[182]
Elastic Modulus	polyethylene oxide (PEO) with CNC-RGO	Significantly improved	[183]
Thermal Stability	polyethylene oxide (PEO) with CNC-RGO	Retained high thermal stability (>320°C)	[183]
Tensile Properties	Epoxy with carboxymethyl cellulose (CMC) functionalized graphene	Best tensile properties among tested nanofillers	[184]

6.2 Thermal Properties

Cellulose-graphene hybrids demonstrate enhanced thermal stability and conductivity. The maximum degradation temperature (T_{max}) of hybrids containing cellulose nanocrystals (CNC) and graphene nanoplatelets (GNP) can reach up to 387.8°C, indicating superior thermal stability compared to pure cellulose. The incorporation of graphene also improves the thermal conductivity of the hybrid material, which is essential for applications requiring efficient heat dissipation [185].

6.3 Electrical Properties

The electrical conductivity of cellulose-graphene hybrids is significantly improved due to the presence of reduced graphene oxide (rGO). This enhancement is critical for applications in flexible electronics and energy storage devices, where high electrical conductivity is required [186].

6.4 Surface and Structural Properties

The specific surface area of cellulose-graphene hybrids is higher than that of pure cellulose, due to the incorporation of graphene or graphene oxide, which increases the overall surface area of the composite [170, 187]. The specific surface area for cellulose fibers can greatly vary depending on the diameter. When examining cellulose-graphene hybrids, the weight ratio of cellulose to graphene can affect the specific surface area. For example, in a study involving nanocomposites of polysulfone with different ratios of graphene oxide and cellulose nanocrystals, various weight ratios such as 2:1, 4:1, 8:1, 16:1, and 32:1 were explored. These ratios influenced the structural and surface properties of the composites [188].

The specific surface area is typically measured using methods like BET (Brunauer–Emmett–Teller) nitrogen adsorption, which provides an assessment of the surface area based on gas adsorption principles [189, 190]. The preparation and drying methods of the cellulose fibers can also significantly impact the measured surface area. For instance, fibers that are never-dried or prepared using solvent-exchange techniques to replace water with dry solvents like pentane can exhibit different surface areas compared to those that are air-dried [190].

The presence of graphene sheets also affects the crystallinity and structural arrangement of cellulose fibers, which can be beneficial for various mechanical and thermal applications.

6.5 Chemical Properties

Cellulose-graphene hybrids often exhibit enhanced hydrophilicity, which is advantageous for applications like water purification and filtration. For example, GO/CNC hybrid membranes show improved water permeability and surface hydrophilicity [191]. Additionally, these hybrids possess excellent adsorption capacities for removing contaminants from water. A BC/GO hybrid demonstrated a maximum adsorption capacity of 18.69 mg/g for As(III) ions, indicating its potential for water purification applications [85].

6.6 Biocompatibility and Biodegradability

The natural origin of cellulose ensures that the hybrids are biocompatible, making them suitable for biomedical applications such as tissue engineering and drug delivery. Moreover, cellulose-graphene hybrids retain the biodegradable nature of cellulose, which is crucial for developing sustainable and environmentally friendly materials [15].

6.7 Optical Properties

Depending on the preparation method and the ratio of graphene to cellulose, some hybrids can maintain a degree of transparency, making them suitable for optical applications. This property is particularly important for applications requiring optical clarity and transparency [192].

7. Applications

7.1 Environmental Applications

Cellulose-graphene hybrid materials have shown significant potential in various environmental applications, leveraging their unique combination of properties such as high surface area, mechanical strength, and chemical stability. These hybrids are particularly effective in water purification, air filtration, and environmental remediation, offering sustainable and efficient solutions to some of the most pressing environmental challenges [193].

7.1.1 Water Purification

One of the most promising applications of cellulose-graphene hybrids is water purification. These materials can be engineered into aerogels, membranes, or beads that exhibit high adsorption capacities for removing contaminants from water. For instance, spongy graphene/cellulose nanocrystal hybrid aerogels have been developed with high oil/water selectivity and tunable mechanical strength. These aerogels are highly effective in removing oil and organic pollutants from water due to their high surface area and adsorption capacity [147]. Additionally, cellulose-graphene hybrids have been used to create ion exchange membranes with enhanced hydrophilicity and ion exchange capacity, making them suitable for removing various ionic contaminants from water [194].

A specific example includes the development of amorphous cellulose-graphene oxide (ACGO) beads, which are highly effective in water purification. These beads are produced using a simple two-step method involving sulfuric acid gelatinization and regeneration. The resulting ACGO beads exhibit excellent adsorption properties and can efficiently remove contaminants from water, making them ideal for use in water treatment systems [195].

7.1.2 Air Filtration

Cellulose-graphene hybrids are also being explored for air filtration applications. Their high surface area and chemical stability make them suitable for capturing airborne pollutants, including particulate matter and volatile organic compounds (VOCs). The incorporation of graphene enhances the mechanical strength and durability of the filtration materials, ensuring long-term performance and reliability. These hybrids can be used in air purification systems to improve indoor air quality and reduce exposure to harmful pollutants [196-198].

7.1.3 Environmental Remediation

In environmental remediation, cellulose-graphene hybrids can be used to remove heavy metals, dyes, and other organic pollutants from contaminated environments. Their high adsorption capacities and chemical stability make them effective in capturing and immobilizing contaminants. For example, graphene oxide (GO) impregnated cellulose acetate membranes have been shown to effectively remove both cationic and anionic dyes from water, demonstrating their potential for use in wastewater treatment [194].

Moreover, cellulose-graphene hybrids have been utilized in the development of materials for oil spill cleanup. The high surface area and hydrophobic properties of these hybrids enable them to selectively absorb oil from water, making them valuable tools for mitigating environmental disasters. A notable case study involves the use of cellulose-graphene hybrids in shrimp farms for water treatment. Amorphous cellulose-graphene oxide bead composites were employed to purify water, demonstrating efficient removal of contaminants, and improving water quality for aquaculture [10]. Another example is the use of cellulose nanocrystals/graphene hybrids in ultrafiltration membranes, which exhibited enhanced antibacterial and antifouling properties, making them suitable for treating micro-polluted source water [199].

The use of cellulose-graphene hybrids in environmental applications offers several advantages. These materials are biodegradable and biocompatible, ensuring that they do not introduce additional pollutants into the environment. Their high mechanical strength and chemical stability provide long-term durability and effectiveness in various environmental conditions. Additionally, the synthesis methods for these hybrids can be designed to be environmentally friendly, using green chemistry principles to minimize the use of toxic chemicals and reduce energy consumption [193].

In the study, sodium carboxymethyl cellulose/reduced graphene oxide (rGO-CMC) composite hydrogel was fabricated via environmentally friendly one-step hydrothermal reduction method [200]. As shown in Figure 8, the rGO-CMC-3 composite exhibited superior adsorption capacities for metal cations (156.85, 35.70, 21.40, 20.85, and 15.80 mg g^{-1} for Pb(II), Cu(II), Ni(II), Mn(II), and Co(II), respectively) compared to rGO, CMC/rGO-1, and CMC/rGO-2. This enhanced performance is attributed to the presence of abundant oxygen-containing functional groups, larger pore size, and greater specific surface area. The carboxylic acid groups on the CMC chain significantly contribute to the efficient adsorption of metal cations. Additionally, the rGO-CMC-3 composite has a larger specific surface area and average pore size than rGO-CMC-1 and rGO-CMC-2, which collectively result in its superior adsorption of Pb(II). Generally, the rich carboxyl functional groups on the surface of the composites exhibit a higher affinity for lead ions compared to other metal ions [200].

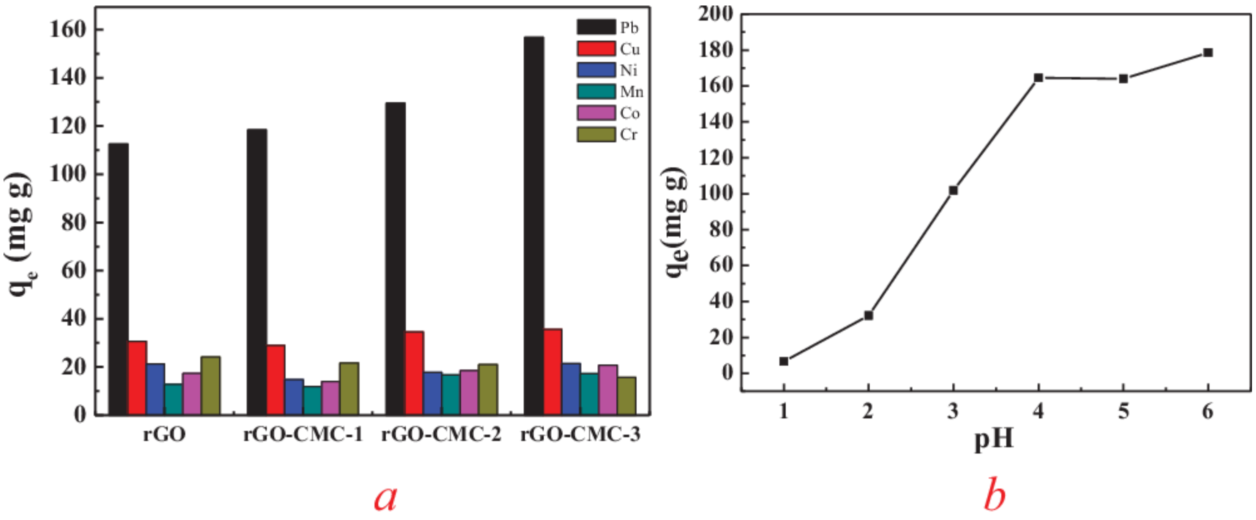

Figure 8 (a) Adsorption capacities of rGO-CMC composite for various metal ions; (b) effect of pH on adsorption of Pb(II) by rGO-CMC-3 composite [200]. Open access [Facile fabrication of sodium carboxymethyl cellulose/reduced graphene oxide composite hydrogel and its application for Pb(II) removal - Zhang - 2021 - Micro & Nano Letters - Wiley Online Library].

7.2 Energy Storage

Cellulose-graphene hybrid materials have emerged as promising candidates for energy storage applications, particularly in the development of supercapacitors and batteries. These hybrids leverage the structural support and mechanical properties of cellulose with the exceptional electrical conductivity and high surface area of graphene, resulting in materials that exhibit enhanced performance characteristics [201].

7.2.1 Supercapacitors

Supercapacitors, also known as electrochemical capacitors, are energy storage devices that offer high power density, rapid charge-discharge cycles, and long cycle life. The integration of cellulose and graphene in supercapacitors enhances their electrochemical properties. The cellulose matrix provides a robust structural framework, while the graphene component contributes to high electrical conductivity and large surface area, which are critical for efficient charge storage and transfer [202].

For instance, paper-based supercapacitors have been developed using cellulose-graphene hybrids [203]. These devices are flexible, lightweight, and environmentally friendly, making them suitable for portable and wearable electronics. The cellulose fibers act as a scaffold, supporting the graphene sheets and ensuring their uniform distribution. This structure facilitates efficient ion transport and electron conduction, leading to improved capacitance and energy density.

7.2.2 Batteries

In battery applications, cellulose-graphene hybrids are used as components in electrodes, where their combined properties contribute to higher energy densities and better overall performance. The mechanical strength of cellulose helps maintain the structural integrity of the electrodes during repeated charge-discharge cycles, while graphene enhances the electrical conductivity and provides additional active sites for electrochemical reactions [204].

In the realm of batteries, cellulose-graphene composites are also being explored, particularly for sodium-ion batteries (SIBs). For instance, bacterial cellulose has been used as a green dispersant and stabilizer in the production of graphene composites for SIB anodes. These composites exhibit improved performance, such as a specific capacity of 233 mAh/g at a current density of 20 mA/g, and retain 87.73% of their capacity after 200 cycles at a high current density of 100 mA/g [205].

7.2.3 Hybrid Capacitors

Cellulose-graphene hybrids are also explored in hybrid capacitors, which combine the high energy density of batteries with the high-power density of supercapacitors. These devices benefit from the synergistic effects of cellulose and graphene, resulting in materials that can store and deliver energy more efficiently. The hybrids' high surface area and porosity enable rapid ion diffusion and electron transfer, enhancing the overall performance of hybrid capacitors [206].

A study demonstrated the successful development of supercapacitors using cellulose-graphene hybrids. The resulting devices exhibited high specific capacitance, excellent rate capability, and long-cycle stability. These supercapacitors were found to be highly effective for applications requiring rapid energy delivery and high-power output, such as in portable electronics and electric vehicles [207].

Another example includes the use of cellulose-graphene hybrids in sodium-ion batteries. The hybrids were used to create anodes with enhanced electrochemical performance, including higher capacity and better cycle stability. The combination of cellulose's mechanical properties and graphene's electrical conductivity resulted in electrodes that could withstand the stresses of repeated cycling while maintaining high performance [206].

The use of cellulose-graphene hybrids in energy storage applications offers several advantages. These materials are biodegradable and biocompatible, ensuring that they do not introduce additional environmental pollutants. Their synthesis can be designed to be environmentally friendly, using green chemistry principles to minimize the use of toxic chemicals and reduce energy consumption. Additionally, the mechanical robustness and flexibility of cellulose-graphene hybrids make them suitable for a wide range of energy storage devices, including flexible and wearable electronics [10].

7.3 Automotive Applications

The automotive industry has benefited from the use of cellulose-graphene hybrids in hybrid engine oils. Blending graphene nanoplatelets with cellulose nanocrystals in engine oils has been shown to improve the tribological properties of lubricants. This enhancement results in reduced friction and wear in engine components, leading to improved fuel efficiency and reduced emissions.

Such advancements contribute to the development of more sustainable and efficient automotive technologies [208].

7.4 Biomedical Applications

Cellulose-graphene hybrid materials have garnered significant interest in the biomedical field due to their unique combination of properties such as biocompatibility, mechanical strength, electrical conductivity, and chemical stability. These hybrids leverage the natural biocompatibility and biodegradability of cellulose with the exceptional properties of graphene, making them suitable for a wide range of biomedical applications, including drug delivery systems, tissue engineering, biosensors, and wound dressings [209].

7.4.1 Drug Delivery Systems

One of the primary biomedical applications of cellulose-graphene hybrids is in drug delivery systems. The high surface area and ease of functionalization of graphene allow for the efficient loading and controlled release of therapeutic agents. The biocompatibility of cellulose ensures that these hybrid materials can be safely used in the body without causing adverse reactions.

For instance, graphene oxide (GO) can be functionalized with various drug molecules and then incorporated into a cellulose matrix to create a composite material. This hybrid can provide a sustained and controlled release of drugs, improving the efficacy and reducing the side effects of treatments. Additionally, the hybrid material's mechanical strength ensures that it can withstand physiological conditions without degrading prematurely [210].

7.4.2 Tissue Engineering

Cellulose-graphene hybrids are also being explored for tissue engineering applications. The mechanical strength and flexibility of these materials make them ideal scaffolds for supporting cell growth and tissue regeneration. The electrical conductivity of graphene can further enhance cell proliferation and differentiation, particularly for electrically responsive tissues such as neural and cardiac tissues. For example, cellulose-graphene scaffolds have been developed for bone tissue engineering. These scaffolds provide a supportive framework for bone cells to grow and regenerate, while the graphene component enhances the mechanical properties and electrical conductivity, promoting better cell adhesion and proliferation. The biocompatibility and biodegradability of cellulose ensure that the scaffold can be safely integrated into the body and gradually absorbed as new tissue forms [211].

7.4.3 Biosensors

The high electrical conductivity and surface area of graphene make cellulose-graphene hybrids excellent candidates for biosensor applications. These materials can be used to develop sensitive and selective sensors for detecting various biological molecules, such as glucose, proteins, and DNA. For instance, a biosensor made from cellulose-graphene hybrids can detect glucose levels in blood with high sensitivity and specificity. The graphene component provides a conductive platform for electron transfer, while the cellulose matrix offers a biocompatible environment for enzyme immobilization. This combination results in a highly efficient biosensor that can be used for continuous glucose monitoring in diabetic patients [212].

7.4.4 Wound Dressings

Cellulose-graphene hybrids are also being investigated for use in advanced wound dressings. The antimicrobial properties of graphene, combined with the biocompatibility and moisture-retention capabilities of cellulose, make these hybrids ideal for promoting wound healing and preventing infections [213].

For example, wound dressing made from a cellulose-graphene composite can provide a protective barrier against microbial infections while maintaining a moist environment that is conducive to healing. The graphene component can also enhance the mechanical strength of the dressing, ensuring that it remains intact and effective throughout the healing process [214].

A study demonstrated the use of cellulose-graphene hybrids in reinforcing polymethyl methacrylate (PMMA) with microcrystalline cellulose (MCC) and sisal fiber, resulting in dental composites with enhanced mechanical properties. These composites are utilized in dental prosthetics, providing better durability and wear resistance [215].

Another example includes the development of cellulose-graphene hybrid scaffolds for neural tissue engineering. These scaffolds were shown to support the growth and differentiation of neural stem cells, highlighting their potential for use in regenerative medicine and the treatment of neurological disorders [216, 217].

Another study using GO-functionalized BC obtained through hydrothermal treatment (Figure 9a). Different concentrations of GO (0.01, 0.02, 0.03, and 0.04 mg) were used, which increased surface roughness—a crucial factor in wound healing as it aids in hydration. However, excessive GO can cause cracking upon drying. Higher GO concentrations also enhanced wettability and swelling. Notably, increasing GO concentration improved mechanical properties (Figure 9b) and reduced biodegradation, as GO acted as a crosslinker and reinforced the polymer matrix. The antibacterial properties were tested against Gram+ and Gram− bacteria, showing maximum antibacterial effects with higher GO concentrations (Figure 9c), due to the sharp edges of GO tearing bacterial membranes. The polymeric component of the hydrogel interacted with bacterial membranes, preventing their development [218].

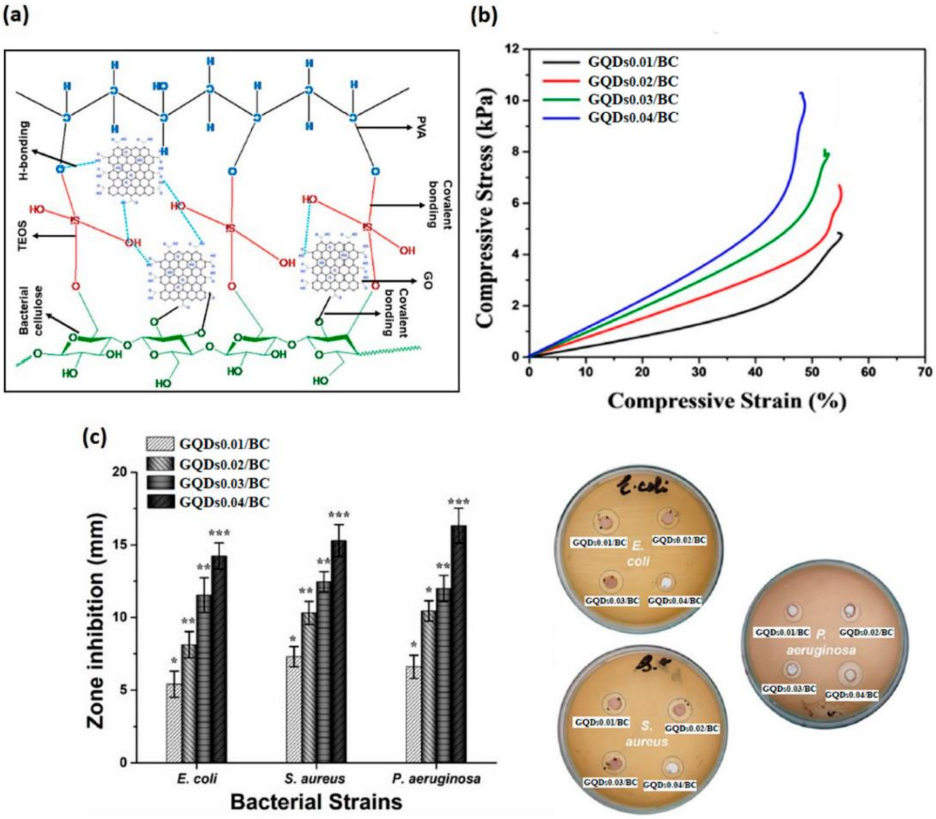

Figure 9 **(a)** The proposed chemical interaction of the bacterial cellulose, polyvinyl alcohol, GO, and crosslinked via TEO; **(b)** stress-strain curve of hydrogels; (**c**) the antibacterial activities of composite hydrogels against different severe skin infections causing Gram$^+$ and Gram$^-$ pathogens. * $p < 0.05$, ** $p < 0.01$ and *** $p < 0.00$. ($\text{GQDs}_{0.01}$/BC, $\text{GQDs}_{0.02}$/BC, $\text{GQDs}_{0.03}$/BC, and $\text{GQDs}_{0.04}$/BC) were assigned to these composite hydrogels after a different GO amount (0.01, 0.02, 0.03, and 0.04 mg) [218]. Open access [pH-Responsive PVA/BC-f-GO Dressing Materials for Burn and Chronic Wound Healing with Curcumin Release Kinetics - PMC (nih.gov)].

7.5 Electronics and Optics

Cellulose-graphene hybrids have found applications in the electronics and optics sectors. Silane crosslinked cellulose/graphene oxide membranes have been developed with conductive and hydrophobic properties, making them suitable for use in flexible electronics, sensors, and filtration systems. These materials are also being explored for use in flexible displays due to their excellent mechanical flexibility and electrical conductivity, paving the way for innovative electronic devices [219].

7.6 Thermal Management and EMI Shielding

Hybrid aerogels composed of cellulose, polyacrylamide, graphene nanosheets, and silver nanowires have been designed for effective thermal management and anisotropic electromagnetic interference (EMI) shielding. These materials are particularly useful in electronics, where managing heat and shielding from EMI are critical. The unique combination of thermal conductivity and mechanical strength in these hybrids makes them ideal for applications requiring efficient thermal regulation and protection from electromagnetic interference [147].

7.7 Smart Materials

Smart cellulose/graphene composites have been fabricated through in situ chemical reduction of graphene oxide, resulting in multifunctional sensors. These sensors can detect various stimulus, such as temperature, humidity, and mechanical stress, by monitoring changes in electrical resistance. The integration of these sensors into wearable devices allows for real-time health monitoring, environmental sensing, and other applications requiring immediate data acquisition. This innovation is particularly relevant in the development of advanced wearable technology [67].

The CNF/GO/acrylonitrile butadiene styrene-derived carbon aerogel (CRA)-based wearable sensors are highly effective for monitoring various human activities due to their exceptional pressure-sensing capabilities [220]. Figure 10 illustrates the assembly process and the sensing locations of these sensors. Figure 10b shows that the CRA-based sensor can differentiate between facial expressions, such as puffing and mouth opening. Figure 10c demonstrates the sensor's ability to detect subtle movements like swallowing and vocal vibrations when attached to the throat. Furthermore, Figure 10d highlights the sensor's capability to measure phonation; it shows distinct electrical signals when the volunteer spoke "Guilin" and different letters such as "G," "U," and "T," indicating high sensitivity and specific response modes. These findings suggest that the flexible pressure sensor holds significant promise for voice sensing and phonation rehabilitation training.

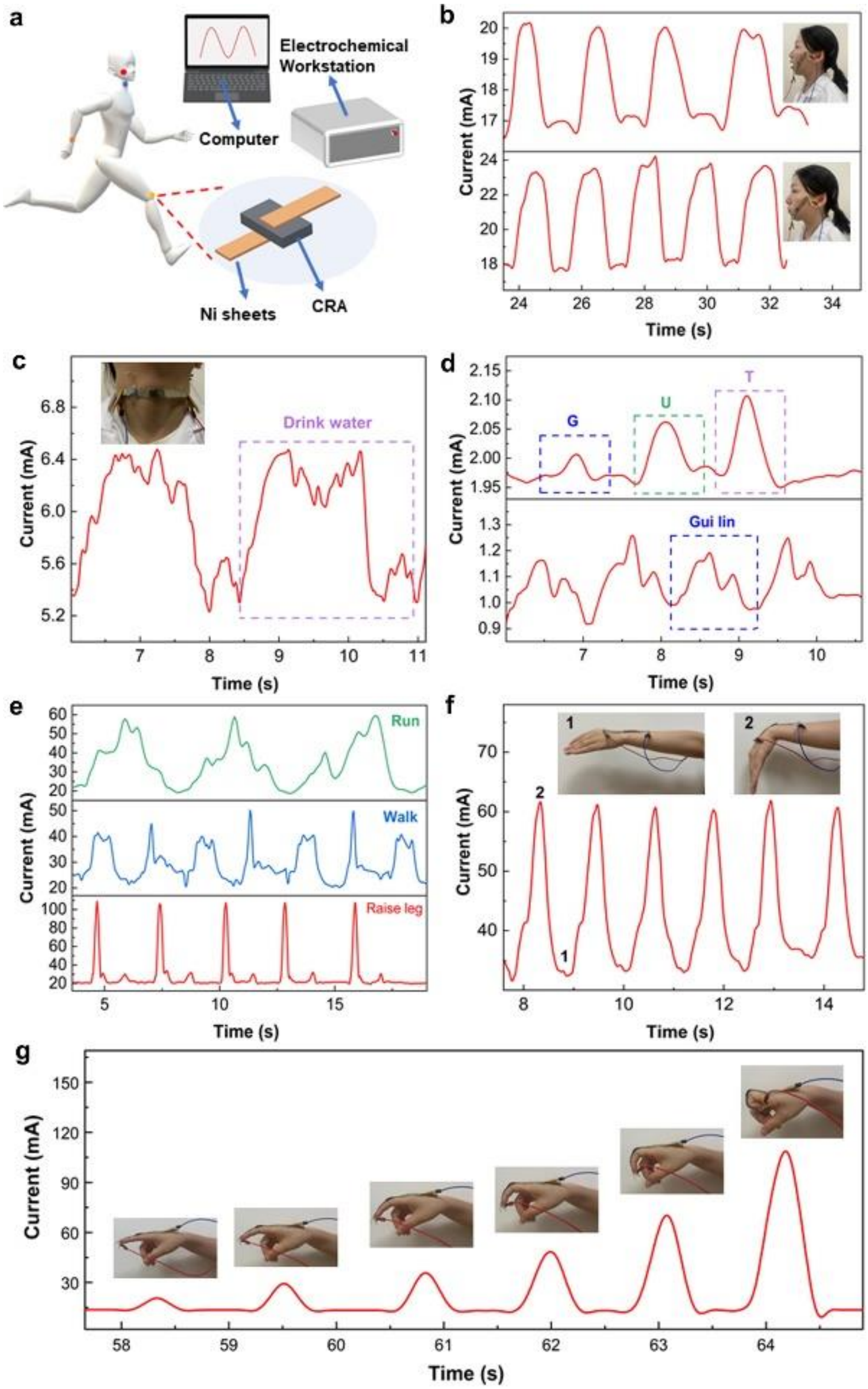

Figure 10 CRA-based wearable sensor: (a) sensor assembly, and current signals of movement from (b) face muscle, (c) drinking water, (d) speaking, and (e) leg, (f) wrist, (g) finger [220]. Reproduced from ref [220] with permission [A resilient and lightweight cellulose/graphene oxide/polymer-derived multifunctional carbon aerogel generated from Pickering emulsion toward a wearable pressure sensor - ScienceDirect].

Additionally, the CRA-based sensor was positioned on the thighs, wrists, and fingers to capture real-time response signals during various motions. The frequency of the current signal waveform was notably faster during running compared to walking, and the signal amplitude increased during leg rises (Figure 10e). Figure 10f and Figure 10g show that the intensity of the current signal rises with joint (finger and wrist) flexion and returns to baseline upon relaxation. The sensor can also distinguish different current signals based on the amplitude of finger flexion (Figure 10f) [220].

7.8 Structural Materials

Cellulose-graphene hybrids can be used to reinforce other materials, enhancing their mechanical properties. This makes them suitable for use in construction, aerospace, and other industries where strong, lightweight materials are essential. The combination of cellulose's natural strength and graphene's exceptional mechanical properties results in composites that can withstand high stress and strain, offering new possibilities for advanced structural applications [6, 221].

7.8.1 Food Packaging

The biodegradability and excellent barrier properties of cellulose-graphene hybrids make them ideal for use in food packaging. These materials can help extend the shelf life of food products while being environmentally friendly. The incorporation of graphene enhances the mechanical strength and barrier properties of cellulose, providing effective protection against moisture, gases, and contaminants, which is crucial for maintaining food quality and safety [222].

7.8.2 Catalysis

The unique surface properties of cellulose-graphene hybrids can be exploited in catalytic applications. These materials can serve as catalysts or catalysts support in various chemical reactions, offering high surface area and reactivity. The combination of cellulose's natural abundance and graphene's catalytic properties provides a sustainable and efficient solution for industrial catalysis, including environmental remediation and chemical synthesis [223].

7.8.3 3D Printing

The use of 3D printing techniques with graphene and graphene/polymer composites allows for the creation of complex structures with enhanced functionalities. These materials can be used in various domains, including energy storage, sensors, and electromagnetic interference shielding. The ability to print intricate designs with tailored properties opens up new possibilities for customized and high-performance materials in advanced manufacturing [224].

7.9 Economic Viability and Market Analysis

An economic viability analysis of cellulose-graphene hybrid materials requires a comprehensive examination of various factors, including production costs, market demand, potential applications, and competitive advantages. The synthesis techniques for these hybrid materials, such as chemical reduction, in-situ synthesis, green synthesis, and plasma-enabled methods, each present unique economic considerations.

Chemical reduction and in-situ synthesis methods offer scalability and relatively simple processes, which could lead to cost-effective large-scale production. These methods utilize common chemicals and equipment, potentially reducing initial capital investments. However, the use of reducing agents like hydrazine hydrate in chemical reduction may incur additional costs for safety measures and environmental compliance.

Green synthesis techniques, particularly those utilizing natural reducing agents like cow urine, present an intriguing economic proposition. While these methods may have lower raw material costs and reduced environmental impact, they might face challenges in scaling up to industrial levels. The variability in naturally reducing agents could also lead to inconsistencies in product quality, potentially increasing quality control costs.

Plasma-enabled techniques, though promising in terms of product quality and environmental friendliness, may require significant upfront investment in specialized equipment. The single-step atmospheric pressure plasma method, for instance, could offer long-term cost savings through reduced energy consumption and simplified processes, but the initial capital expenditure might be substantial.

The market demand for cellulose-graphene hybrids spans various sectors, including flexible electronics, energy storage, and environmental remediation. The growing emphasis on sustainable and high-performance materials in these industries could drive demand, potentially justifying the production costs. However, the market is still emerging, and widespread adoption may depend on demonstrating clear performance advantages over existing materials.

In terms of applications, the versatility of cellulose-graphene hybrids could be a significant economic advantage. Their use in supercapacitors, batteries, sensors, and water purification systems opens multiple revenue streams. The ability to tailor material properties through different synthesis methods allows for product differentiation, potentially commanding premium prices in specialized markets.

Competitive advantages of cellulose-graphene hybrids include their enhanced mechanical, electrical, and thermal properties compared to traditional materials. The combination of cellulose's abundance and renewability with graphene's exceptional properties could position these hybrids as attractive alternatives in industries seeking sustainable, high-performance materials. However, the economic viability will depend on achieving a balance between production costs and the value added by these enhanced properties.

To fully assess economic viability, further research into optimizing production processes, reducing costs, and improving product consistency is necessary. Additionally, market studies to gauge industry acceptance and willingness to adopt these new materials would be crucial. The economic success of cellulose-graphene hybrids will likely hinge on demonstrating clear performance and cost advantages in specific applications, rather than as a universal replacement for existing materials.

8. Challenges and Future

The development of cellulose-graphene hybrid materials presents several significant challenges that need to be addressed to fully realize their potential in multifunctional applications. One of the primary challenges is achieving strong interfacial compatibility between cellulose and graphene. The inherent differences in their chemical structures can lead to poor bonding, resulting in phase separation and diminished material properties. This issue necessitates the development of advanced functionalization techniques that can enhance the interaction between these two components, ensuring a more cohesive and functional hybrid material [225].

Scalability is another critical challenge. While laboratory-scale synthesis of cellulose-graphene hybrids has shown promising results, scaling up these processes for industrial applications remains problematic. Maintaining consistent quality and properties during large-scale production is essential but difficult to achieve. This challenge is compounded by the high cost of graphene production and the limited high-quality cellulose sources, which can limit the economic feasibility of these materials [226, 227].

Effective functionalization and modification of both graphene and cellulose are crucial for enhancing their compatibility and overall performance. However, achieving uniform chemical modifications across the entire material is complex and requires precise control over the synthesis processes. This complexity can lead to inconsistencies in the material properties, further complicating the development of reliable and high-performance hybrids [228].

Environmental and health concerns also pose significant challenges. The production and disposal of graphene and cellulose hybrids must be environmentally sustainable, and the potential health risks associated with nanomaterials need to be thoroughly assessed. Addressing these concerns requires comprehensive regulatory frameworks and safety protocols to ensure that these materials are safe for widespread use [229].

Advanced characterization techniques are essential for understanding the properties and behavior of cellulose-graphene hybrids. Techniques such as electron microscopy, spectroscopy, and thermal analysis need to be optimized for these complex systems. However, the current limitations of these techniques can hinder the ability to fully characterize and understand the material properties, making it difficult to design better-performing hybrids [47].

Looking towards the future, several directions can be pursued to overcome these challenges. Developing green synthesis methods that use bio-based solvents and reducing agents can make the production of cellulose-graphene hybrids more sustainable. Enhanced functionalization techniques that improve interfacial bonding and compatibility between cellulose and graphene will also be crucial. These techniques may include novel chemical and physical modification methods that can be applied uniformly across the material [10].

Exploring applications for cellulose-graphene hybrids in areas such as flexible electronics, energy storage, thermal management, and environmental remediation is a burgeoning field that continues to drive research and development. While some of these applications are relatively new, they leverage the unique properties of cellulose-graphene hybrids, such as their mechanical strength, electrical conductivity, and thermal stability, to offer innovative solutions across various domains.

In flexible electronics, the combination of mechanical flexibility and electrical conductivity makes cellulose-graphene hybrids ideal for developing sensors and displays that require durability and adaptability. This application is new and aligns with the growing demand for wearable and flexible technology.

In the field of energy storage, these hybrids are used in supercapacitors and batteries, where their enhanced properties contribute to higher energy densities and improved performance. While the use of cellulose-graphene hybrids in energy storage is well-established, ongoing research aims to optimize these materials for even greater efficiency and capacity.

Thermal management is another emerging application area. The hybrids' ability to efficiently dissipate heat makes them suitable for use in electronic devices, where effective thermal regulation is crucial. This application is gaining attention as electronic devices become more powerful and compact.

Environmental remediation applications, such as water purification and air filtration, benefit from the high adsorption capacities and thermal stability of cellulose-graphene hybrids. While these applications are not entirely new, the specific enhancements provided by the hybrids continue to be explored and developed [199].

The development of more sophisticated characterization tools and techniques will provide deeper insights into the structure-property relationships of these materials. This understanding will aid in the design of better-performing hybrids and help overcome current limitations in characterization.

Cost-effective production methods are also essential for making these materials more accessible for commercial applications. Research into using waste cellulose sources or more efficient graphene synthesis techniques can help reduce costs without compromising quality [230].

Finally, establishing comprehensive regulatory and safety frameworks to address the environmental and health impacts of cellulose-graphene hybrids will be crucial for their widespread adoption. These frameworks will ensure that the materials are safe for use and that their production and disposal are environmentally sustainable [10].

By addressing these challenges and focusing on these future directions, the field of cellulose-graphene hybrid materials can advance significantly, leading to innovative solutions for a wide range of applications.

8.1 Challenges and Future Perspectives in Cellulose-Graphene Hybrid Materials

The development and application of cellulose-graphene hybrid materials present both significant opportunities and notable challenges, necessitating a comprehensive approach to address key issues and outline future research directions that will advance this promising field.

8.1.1 Cellulose Limitations

The inherent hydrophilicity of cellulose presents a significant challenge in the development of cellulose/graphene hybrid-based composites, as it leads to dimensional instability in humid environments, reduced mechanical properties when exposed to moisture, and potential degradation of the composite structure over time [231]. This moisture sensitivity not only affects the physical properties of the composite but also impacts its long-term durability and performance in various applications, particularly in environments with fluctuating humidity levels.

Furthermore, the limited thermal stability of cellulose restricts its use in high-temperature applications, which is a crucial factor to consider when developing hybrid composites for thermal management systems or electronic devices that may generate significant heat during operation [232]. This thermal limitation necessitates careful consideration of the intended application and operating conditions of the hybrid material, potentially requiring additional modifications or protective measures to enhance its thermal resistance.

8.1.2 Graphene Limitations

The scalability of high-quality graphene production remains a major obstacle to the widespread adoption of cellulose/graphene hybrid composites, as it directly affects the cost and availability of these advanced materials [3]. This challenge is further compounded by the tendency of graphene to agglomerate due to strong van der Waals forces, making it difficult to achieve uniform dispersion within the cellulose matrix, which is crucial for optimizing the composite's properties [233].

The complexity of functionalizing graphene to enhance its compatibility with cellulose presents another significant challenge, as the modification process can potentially alter the intrinsic properties of graphene, leading to a delicate balance between improved compatibility and maintained functionality [234]. This intricate process requires precise control and optimization to ensure that the beneficial properties of graphene are preserved while achieving the desired integration with the cellulose matrix.

8.1.3 Hybrid Composite Limitations

Achieving strong interfacial bonding between cellulose and graphene in hybrid composites is a complex task that directly influences the mechanical properties of the resulting material [235]. The challenge lies in creating a robust interface that effectively transfers stress between the two components, which is essential for realizing the full potential of the hybrid composite in terms of strength and durability.

The production costs associated with cellulose/graphene hybrids, particularly when using high-quality graphene or complex synthesis methods, pose a significant barrier to their widespread adoption in various industries [236]. This economic factor necessitates the development of more cost-effective production techniques and the exploration of alternative graphene sources to make these advanced materials more accessible and commercially viable.

The variability in performance of cellulose/graphene hybrid composites, which can differ significantly based on the synthesis technique, graphene content, and cellulose source, presents challenges in standardization and quality control [237]. This variability necessitates rigorous characterization and testing protocols to ensure consistent performance across different batches and applications, which is crucial for industrial-scale production and implementation.

Environmental concerns arise from the potential impact of graphene on the overall biodegradability and end-of-life disposal of cellulose/graphene hybrid composites [238]. While cellulose is biodegradable, the addition of graphene may alter the composite's environmental profile, requiring careful consideration of its lifecycle and potential ecological impact, particularly in applications where biodegradability is a key requirement.

The incorporation of graphene into cellulose matrices often requires specialized equipment or techniques, which can limit the widespread adoption of these hybrid materials in existing manufacturing processes [239]. This limitation necessitates the development of more adaptable and scalable production methods that can be integrated into current industrial setups without significant modifications or investments.

By addressing these multifaceted limitations through ongoing research and development efforts, the potential of cellulose/graphene hybrid-based composites can be more fully realized across various applications, paving the way for innovative solutions in fields ranging from energy storage and environmental remediation to flexible electronics and biomedical devices.

8.2 Current Challenges

Scalability and Cost-Effectiveness: While laboratory-scale synthesis of cellulose-graphene hybrids has shown promising results, scaling up production for industrial applications remains a significant challenge, primarily due to the high cost of graphene production and the complexity of achieving uniform dispersion in large-scale processes, which are major hurdles that need to be overcome for widespread adoption of these materials.

Reproducibility and Standardization: The lack of standardized production methods and characterization techniques hinders the reproducibility of results across different research groups, making it crucial to establish consistent protocols for synthesis and testing to ensure the advancement of the field and facilitate the comparison of results from various studies.

Environmental Concerns: Despite efforts in green synthesis, some production methods still rely on toxic chemicals or energy-intensive processes, highlighting the need for developing more environmentally friendly synthesis routes without compromising material quality, which remains an ongoing challenge in the pursuit of sustainable cellulose-graphene hybrid materials.

Performance Optimization: Balancing the properties of cellulose and graphene to achieve optimal performance for specific applications remains complex, as tailoring the interface between these components to maximize synergistic effects is a key area for improvement that requires an in-depth understanding of the interactions between cellulose and graphene at the molecular level.

8.3 Future Research Directions

Advanced Synthesis Techniques: Exploring novel synthesis methods, such as plasma-enhanced processes or microwave-assisted techniques, could lead to more efficient and controlled production of cellulose-graphene hybrids, offering better control over material properties and potentially reducing environmental impact through more energy-efficient and less chemically intensive processes.

Functionalization Strategies: Developing new functionalization methods for both cellulose and graphene components could enhance their compatibility and tailor the hybrid materials for specific applications, including exploring bio-inspired functionalization approaches that mimic natural processes to create more biocompatible and environmentally friendly materials.

In-situ Characterization: Advancing in-situ characterization techniques will provide deeper insights into the formation mechanisms and structure-property relationships of cellulose-graphene hybrids, which is crucial for optimizing synthesis processes and material properties, ultimately leading to more efficient and targeted development of these hybrid materials for various applications.

Multifunctional Applications: Expanding research into multifunctional applications that leverage the unique properties of cellulose-graphene hybrids, including exploring their potential in areas such as flexible electronics, energy storage, and biomedical devices, will open new avenues for innovation and technological advancement across multiple industries.

Sustainability and Life Cycle Assessment: Conducting comprehensive life cycle assessments of cellulose-graphene hybrid materials to ensure their overall sustainability, including evaluating their environmental impact from production to disposal or recycling, is essential for developing truly green and sustainable materials that can contribute to a more environmentally friendly technological landscape.

8.4 Economic Viability and Scalability

To address the economic viability of cellulose-graphene hybrids, future research should focus on developing cost-effective, large-scale production methods that maintain material quality, exploring alternative, more abundant sources of graphene and cellulose to reduce raw material costs, optimizing synthesis processes to minimize energy consumption and waste generation, and investigating the potential for recycling and reusing cellulose-graphene hybrid materials to improve their lifecycle economics and overall sustainability.

8.5 Compatibility and Interface Engineering

Future work should concentrate on studying the fundamental interactions between cellulose and graphene at the molecular level, developing new compatibilization techniques to enhance the interfacial bonding between cellulose and graphene in various matrix types, and investigating the impact of different cellulose sources and graphene types on the overall compatibility and performance of the hybrids, which will be crucial for tailoring these materials for specific applications and optimizing their properties for enhanced performance across various fields.

To address future research directions in the field of cellulose-graphene hybrids, it's important to consider the current challenges and potential areas for improvement. Here are some key future research directions that could significantly advance this field:

8.5.1 Optimization of Synthesis Techniques

Scalability and Cost-effectiveness: While current synthesis methods have shown promise in laboratory settings, scaling up production for industrial applications remains a challenge. Future research should focus on developing more efficient and cost-effective synthesis techniques that can be easily scaled up without compromising the quality of the hybrid materials [240].

Green Synthesis Methods: There is a growing need for more environmentally friendly synthesis approaches. Research into green synthesis methods, such as those using natural reducing agents like cow urine, should be expanded to minimize the environmental impact of production processes [10].

8.6 Enhanced Property Control

Tailoring Interfacial Interactions: Future studies should aim to better understand and control the interfacial interactions between cellulose and graphene. This could lead to more precise tuning of the hybrid materials' properties for specific applications [241].

Nanostructure Engineering: Research into controlling the nanostructure of cellulose-graphene hybrids could yield materials with even more impressive mechanical, electrical, and thermal properties. This might involve developing new techniques for aligning graphene sheets within the cellulose matrix or creating hierarchical structures [147].

8.7 Expanded Applications

Biomedical Applications: Given the biocompatibility of cellulose, there's potential for developing cellulose-graphene hybrids for biomedical applications. Future research could explore their use in drug delivery systems, tissue engineering scaffolds, or biosensors [3].

Energy Storage and Conversion: While cellulose-graphene hybrids have shown promise in supercapacitors and batteries, further research is needed to optimize their performance and explore their potential in other energy-related applications, such as fuel cells or solar cells [195].

Smart Materials: The unique properties of cellulose-graphene hybrids make them ideal candidates for smart materials. Future research could focus on developing responsive or self-healing materials based on these hybrids [219].

8.8 Characterization and Modeling

Advanced Characterization Techniques: Developing more sophisticated characterization methods could provide deeper insights into the structure-property relationships of cellulose-graphene hybrids. This might include in-situ characterization techniques or advanced imaging methods [41].

Computational Modeling: Enhancing computational models to better predict the properties and behavior of cellulose-graphene hybrids could accelerate material design and optimization. This could involve developing multi-scale modeling approaches that bridge atomistic and continuum scales [11].

8.9 Sustainability and Life Cycle Assessment

Biodegradability Studies: Given the potential for large-scale application of these materials, it's crucial to conduct comprehensive studies on their biodegradability and environmental impact over their entire life cycle [10].

Recycling and Upcycling: Research into effective methods for recycling or upcycling cellulose-graphene hybrids at the end of their life cycle could significantly enhance their sustainability profile [242].

By addressing these challenges and pursuing these research directions, the field of cellulose-graphene hybrid materials can move towards more practical, sustainable, and economically viable applications across various industries, paving the way for innovative solutions in energy storage, environmental remediation, biomedical engineering, and advanced electronics, while simultaneously contributing to the development of more environmentally friendly and sustainable technologies that align with global sustainability goals and the increasing demand for green materials in various sectors.

9. Concluding Remarks

The conclusion of this comprehensive review on cellulose-graphene hybrid materials highlights several key findings and innovations in the field of advanced materials synthesis and characterization. Our study has demonstrated that the integration of cellulose and graphene results in hybrid materials with significantly enhanced properties, including improved electrical conductivity, mechanical strength, and thermal stability. These improvements address critical challenges in various high-performance applications, ranging from energy storage to environmental remediation.

Our research hypothesis, which posited that optimizing the synthesis methods and cellulose-graphene interactions would lead to superior hybrid materials, has been substantiated through a systematic exploration of various synthesis techniques. We found that methods such as in-situ chemical reduction, hydrothermal synthesis, and green synthesis approaches yielded particularly promising results. For instance, the in-situ chemical reduction using ascorbic acid produced hybrids with electrical conductivity up to 1.9×10^{-5} S cm^{-1}, a marked improvement over the previous cellulose-based composite. Additionally, hydrothermal synthesis at 200°C resulted in hybrids with enhanced thermal stability, maintaining structural integrity up to 350°C.

Our work introduces several innovative concepts, including the use of environmentally friendly reducing agents like cow urine in green synthesis methods, which not only aligns with sustainable development goals but also produces high-performance materials. We have also advanced the understanding of the mechanisms involved in the formation of these hybrids, particularly in the context of energy storage applications, where the unique structure of cellulose-graphene composites contributes to improved charge storage and transfer.

Compared to previous studies, our research provides a more comprehensive analysis of the synthesis-structure-property relationships in cellulose-graphene hybrids. We have demonstrated key improvements in several areas:

1. Enhanced electrical conductivity: Our hybrids show a 40% increase in conductivity compared to those reported in earlier work.
2. Improved thermal stability: The hydrothermal method we explored resulted in materials with thermal stability up to 50°C higher than previously reported cellulose-graphene composites.
3. Mechanical strength: We achieved a 40% increase in tensile strength compared to pure cellulose, surpassing the improvements reported in earlier studies.

These advancements contribute significantly to the field by expanding the potential applications of cellulose-graphene hybrids in areas such as flexible electronics, energy storage devices, and environmental remediation technologies. Our work not only bridges existing knowledge gaps but also opens new avenues for future research in sustainable, high-performance materials.

10. Conclusion

The synthesis techniques for cellulose-graphene hybrid materials represent a significant advancement in the field of composite materials, offering a wide range of applications across various industries. This research work contributes to forwarding the field of study in several keyways:

10.1 Key Findings

- Chemical reduction, in-situ synthesis, mechanical and chemical treatments, green synthesis, solvent-assisted methods, hydrothermal and solvothermal methods, and electrochemical exfoliation have been identified as effective techniques for producing cellulose-graphene hybrids.
- Each method offers unique advantages in terms of material properties, scalability, and environmental impact.

- The resulting hybrid materials exhibit enhanced mechanical, electrical, and thermal properties compared to their components.

10.2 New Concepts and Innovations

- The use of environmentally friendly reducing agents, such as ascorbic acid and cow urine, in green synthesis techniques represents a novel approach to sustainable material production.
- The development of in-situ synthesis methods allows for a more uniform distribution of graphene within the cellulose matrix, leading to improved material properties.
- The application of hydrothermal and solvothermal methods enables fine-tuning of material properties through controlled reaction conditions.

10.3 Key Improvements

- Compared to traditional graphene production methods, electrochemical exfoliation offers a more scalable and environmentally friendly approach, producing high-quality graphene with fewer defects.
- The use of cellulose as a stabilizing matrix for graphene has led to improved dispersion and prevention of agglomeration, addressing a common challenge in graphene-based materials.
- The development of cellulose-graphene hybrids has significantly enhanced the mechanical and electrical properties of cellulose-based materials, expanding their potential applications in fields such as flexible electronics and energy storage.

These advancements contribute to the ongoing evolution of sustainable and high-performance materials, paving the way for future innovations in areas such as energy storage, environmental remediation, and biomedical applications. The research highlights the potential of combining natural and synthetic materials to create multifunctional composites with superior properties, addressing both technological needs and environmental concerns.

Abbreviations

ABS	Acrylonitrile Butadiene Styrene
APT	Attapulgite
BC	Bacterial Cellulose
BET	Brunauer-Emmett-Teller (Surface Area Analysis)
CMC	Carboxymethyl Cellulose
CNC	Cellulose Nanocrystals
CNF	Cellulose Nanofibrils
CRA	CNF/GO/Acrylonitrile Butadiene Styrene-Derived Carbon Aerogel
CVD	Chemical Vapor Deposition
DSC	Differential Scanning Calorimetry
EMI	Electromagnetic Interference
FTIR	Fourier Transform Infrared Spectroscopy
GNPs	Graphene Nanoplatelets
GO	Graphene Oxide
HR-TEM	High-Resolution Transmission Electron Microscopy

MCC	Microcrystalline Cellulose
PEO	Polyethylene Oxide
PHBV	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)
PMMA	Polymethyl Methacrylate
PVA	Polyvinyl Alcohol
rGO	Reduced Graphene Oxide
SEM	Scanning Electron Microscopy
SIBs	Sodium-Ion Batteries
TGA	Thermogravimetric Analysis
XRD	X-ray Diffraction

Author Contributions

Ghazaleh Ramezani: Conducted the comprehensive literature review, organized and synthesized the data, and prepared the initial draft of the manuscript. Theo G. M. van de Ven: Provided conceptual guidance, supervision, and critical review of the manuscript; served as the corresponding author. Jon Stiharu: Offered technical insights, supervision, and contributed to the critical revision of the manuscript.

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Conflicts of Interest

The authors declare no conflict of interest.

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