encyclopedia

Review

Optimizing Graphene Oxide Content in Cellulose Matrices: A Comprehensive Review on Enhancing the Structural and Functional Performance of Composites

Ghazaleh Ramezani [1], Ion Stiharu [1,*], Theo G. M. van de Ven [2] and Vahe Nerguizian [3]

[1] Department of Mechanical and Industrial Engineering, Concordia University, Montreal, QC H3G 1M8, Canada; ghazaleh.ramezani@mail.concordia.ca
[2] Department of Chemistry, McGill University, Montreal, QC H4A 3J1, Canada; theo.vandeven@mcgill.ca
[3] Département de Génie Électrique, École de Technologie Supérieure, Montreal, QC H3C 1K3, Canada; vahe.nerguizian@etsmtl.ca
* Correspondence: ion.stiharu@concordia.ca

Abstract: The incorporation of graphene into cellulose matrices has emerged as a promising strategy for enhancing the structural and functional properties of composite materials. This comprehensive review provides a critical analysis of recent advances in optimizing graphene content in cellulose matrices and its impact on composite performance. Various optimization techniques, including response surface methodology, particle swarm optimization, and artificial neural networks, have been employed to identify optimal graphene concentrations and processing conditions. Quantitative analyses demonstrate significant improvements in mechanical properties, with notable increases in tensile strength and Young's modulus reported for graphene/microfibrillated cellulose composites. Substantial enhancements in thermal stability have been observed in lysozyme-modified graphene nanoplatelet–cellulose composites. Electrical conductivity has been achieved at low graphene loading levels. Additionally, barrier properties, biocompatibility, and functionality for applications such as energy storage and environmental remediation have been substantially improved. This review explores case studies encompassing the optimization of thermal conductivity, viscosity, durability behaviors, pollutant removal, and various other properties. Despite promising results, challenges remain, including uniform dispersion, scalability, cost-effectiveness, and long-term stability. Strategies such as surface functionalization, solvent selection, and protective coatings are discussed. Future research directions, including novel processing techniques like 3D printing and electrospinning, as well as the incorporation of additional functional materials, are outlined. This review synthesizes current knowledge, identifies emerging trends, and provides a roadmap for future research in the rapidly evolving field of graphene–cellulose composites.

Keywords: graphene; cellulose; composites; optimization; response surface methodology (RSM); particle swarm optimization (PSO); artificial neural networks (ANNs); mechanical properties

check for updates

Citation: Ramezani, G.; Stiharu, I.; van de Ven, T.G.M.; Nerguizian, V. Optimizing Graphene Oxide Content in Cellulose Matrices: A Comprehensive Review on Enhancing the Structural and Functional Performance of Composites. *Encyclopedia* **2024**, *4*, 1827–1856. https://doi.org/10.3390/encyclopedia4040120

Academic Editor: Raffaele Barretta

Received: 20 September 2024
Revised: 12 November 2024
Accepted: 25 November 2024
Published: 29 November 2024

1. Introduction

The integration of graphene oxide (GO) into cellulose matrices has emerged as a promising frontier in materials science, offering unprecedented opportunities to enhance the structural and functional properties of composite materials. This synergistic combination leverages the unique characteristics of GO—a two-dimensional carbon-based material with abundant oxygen-containing functional groups, high specific surface area, and excellent dispersibility in aqueous solutions—with the abundant, renewable, and biodegradable nature of cellulose [1,2]. Unlike pristine graphene, GO's oxygen-rich surface chemistry facilitates stronger interactions with cellulose, making it particularly suitable for composite applications. A study by Luo et al. (2018) reported the fabrication of bacterial cellulose–graphene oxide hydrogels using layer-by-layer assembly, resulting in extremely enhanced mechanical properties [3]. Another study by Song et al. (2017) demonstrated the creation of nanofibrillated cellulose (NFC)–reduced graphene oxide (RGO) hybrid films using layer-by-layer assembly, achieving a high tensile strength of 107 MPa along with outstanding flexibility [4]. Another study explored the synergistic effects of GO and CNC on the properties of poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) nanocomposites. The researchers prepared ternary nanocomposites consisting of PHBV and CNC-GO nanohybrids using a simple solution casting method. The study found that the ternary nanocomposites displayed significantly improved properties compared to neat PHBV and binary nanocomposites. Specifically, the ternary nanocomposites with 1 wt% covalent bonded CNC-GO exhibited excellent barrier properties, along with enhanced thermal stability, mechanical properties, and antibacterial activity [5]. These studies underscore the versatility of GO–cellulose composites in addressing challenges in materials engineering and environmental sustainability.

The optimization of GO content within cellulose matrices remains a critical challenge that significantly influences the final properties and performance of these composites. Factors such as the degree of oxidation of GO, the method of incorporation, and the processing conditions all play crucial roles in determining the overall efficacy of the composite. To address this challenge, researchers have employed various sophisticated optimization techniques. For instance, Basha et al. (2022) utilized response surface methodology (RSM) to optimize the adsorption of methylene blue dye using sulfonated graphene oxide impregnated cellulose acetate floated beads, demonstrating the effectiveness of statistical approaches in tailoring composite properties [6]. Building on this work, Khiam et al. (2022) applied particle swarm optimization (PSO) in conjunction with artificial neural networks (ANN) to optimize GO content in graphene oxide–chitosan composites for methylene blue adsorption, highlighting the potential of machine learning techniques in materials design [7]. Furthermore, Balasubramani et al. (2020) employed a similar ANN-PSO approach to model and optimize the removal of an antidepressant using graphene oxide–cellulose nanogel composites, further emphasizing the growing importance of these techniques in environmental applications [8].

Advancements in understanding the fundamental mechanisms underlying the enhanced properties of GO–cellulose composites have also been made. Recent advancements in understanding the fundamental mechanisms underlying the enhanced properties of graphene oxide (GO)–cellulose composites have provided significant insights into their remarkable performance and potential applications. These composites have garnered considerable attention due to their synergistic combination of the exceptional properties of GO and the abundant, renewable nature of cellulose. The enhanced properties of GO–cellulose composites can be largely attributed to the strong interfacial interactions between the two components. Recent studies have elucidated the nature of these interactions, revealing a complex interplay of hydrogen bonding, electrostatic interactions, and mechanical interlocking [9]. The presence of oxygen-containing functional groups on both GO and cellulose surfaces facilitates the formation of an extensive hydrogen bonding network, which significantly contributes to the improved mechanical properties of the composites [9,10]. Furthermore, molecular dynamics simulations have provided valuable insights into the nanoscale mechanisms of reinforcement. These simulations have demonstrated that the incorporation of GO into cellulose-based matrices can enhance the Young's modulus and tensile strength by up to 32.1% and 23.8%, respectively. This remarkable improvement is attributed to the formation of hydrogen bonds and the presence of Ca^{2+} ions near the interface, which play a crucial role in improving interfacial adhesion and facilitating load transfer between GO and the cellulose matrix [10].

Recent research has also highlighted the multifunctional nature of GO–cellulose composites, particularly in environmental remediation applications. These composites have shown exceptional performance in the removal of heavy metals, organic compounds, dyes, and microbial contaminants from wastewater [11]. The high surface area, superior mechanical strength, and notable biodegradability of nanocellulose composites, when combined with GO, create a powerful platform for addressing complex environmental challenges [10].

In the context of water treatment, GO–cellulose composites have demonstrated remarkable adsorption capacities for various pollutants. The integration of GO with cellulose-based materials has been shown to enhance the stability and processability of the composites, making them more suitable for large-scale industrial applications [12]. Moreover, these composites exhibit excellent regeneration potential, further enhancing their economic viability and sustainability in wastewater treatment processes [13].

D'Amora et al. (2023) conducted an extensive molecular dynamics study to elucidate the complex interfacial interactions between graphene oxide (GO) and cellulose nanofibrils [14]. Their research employed advanced simulation techniques to model the behavior of GO sheets and cellulose nanofibrils at the molecular level. The study revealed that hydrogen bonding plays a crucial role in the mechanical reinforcement of GO–cellulose composites. Specifically, they observed that the oxygen-containing functional groups on GO surfaces formed strong hydrogen bonds with the hydroxyl groups of cellulose chains. This interaction significantly enhanced the interfacial adhesion between GO and cellulose, leading to improved mechanical properties of the composite. Furthermore, the simulations demonstrated that the density and distribution of oxygen-containing groups on GO surfaces directly influenced the strength of these interfacial interactions. This finding provides valuable insights for optimizing GO functionalization to maximize its reinforcing effect in cellulose matrices.

Many studies explored on developed an innovative 3D printing methods for fabricating GO–cellulose aerogels with hierarchical porous structures [15–18]. Their approach utilized a custom-designed extrusion-based 3D printer capable of precisely depositing a GO–cellulose nanofiber (CNF) ink. The ink formulation was carefully optimized to achieve the right viscosity and shear-thinning behavior necessary for successful printing. The researchers employed a freeze-drying process post-printing to create the aerogel structure. The resulting 3D-printed GO–cellulose aerogels exhibited remarkable mechanical strength, with compressive moduli up to 150% higher than conventionally prepared composites. Additionally, the hierarchical porous structure, consisting of macropores from the printed architecture and mesopores within the GO-CNF network, led to enhanced electrical conductivity. The authors demonstrated the potential of these 3D-printed aerogels in various applications, including as high-performance supercapacitor electrodes and efficient oil absorption materials for environmental remediation.

Biswas et al. (2023) conducted a comprehensive study on the use of GO–cellulose nanocrystal (CNC) composites as efficient drug carriers for controlled release applications [19]. The researchers developed a novel synthesis method that involved the in situ reduction of GO in the presence of CNCs, resulting in a uniform dispersion of reduced GO (rGO) within the CNC matrix. They investigated the drug loading and release characteristics of these composites using doxorubicin, a common anticancer drug, as a model compound. The study revealed that the GO-CNC composites exhibited a remarkably high drug loading capacity of up to 85%, significantly higher than pure CNCs or GO alone. The researchers attributed this enhanced loading to the synergistic effects of π-π stacking interactions between the drug and rGO sheets, as well as hydrogen bonding with the CNC surface. Furthermore, the drug release profiles showed a sustained release behavior over 72 h, with the ability to modulate the release rate by adjusting the GO content in the composite. In vitro cytotoxicity studies demonstrated that the drug-loaded GO-CNC composites effectively inhibited the growth of cancer cells while showing minimal toxicity to healthy cells. The authors also conducted in vivo studies using a mouse tumor model, which showed improved tumor reduction compared to free doxorubicin administration.

This comprehensive study highlights the potential of GO–cellulose nanocomposites in developing advanced drug delivery systems for cancer therapy.

Another study made significant strides in developing GO–cellulose nanofiber composite electrodes for high-performance supercapacitors [20]. The research focused on optimizing the GO content and the nanostructure of the composite to maximize energy storage capacity and cycling stability. The team employed a layer-by-layer assembly technique to create highly ordered GO-CNF nanostructures with precisely controlled thickness and composition. They investigated the effect of GO content (ranging from 5 to 30 wt%) on the electrochemical performance of the composites. The study revealed that a GO content of 20 wt% resulted in the optimal balance between electrical conductivity and ion accessibility. These optimized GO-CNF electrodes exhibited an exceptionally high specific capacitance of 412 F/g at a current density of 1 A/g, which is among the highest reported for cellulose-based supercapacitors. Moreover, the electrodes demonstrated remarkable cycling stability, retaining 95% of their initial capacitance after 10,000 charge–discharge cycles. The researchers attributed this outstanding performance to the synergistic effects of GO's high electrical conductivity and the CNF's porous structure, which facilitated rapid ion transport and provided a large surface area for charge storage.

To further demonstrate the practical applicability of their GO-CNF supercapacitors, Kasprzak et al. fabricated a flexible, all-solid-state device using a gel electrolyte [21]. This device maintained excellent capacitive behavior under various bending conditions, showcasing its potential for use in wearable electronics and flexible energy storage systems.

These case studies illustrate the diverse and promising applications of GO–cellulose composites, highlighting the importance of continued research in this field to unlock their full potential in various technological domains.

From a chemical and physical perspective, graphene can be categorized into two primary classes: CVD-prepared graphene and oxidized graphene. CVD-prepared single- or multilayer graphene exhibits well-defined aromatic structures with minimal defects, as illustrated in Figure 1a,b. While these graphene sheets possess highly reactive surfaces suitable for bioelectrode applications, their poor solubility limits their use in nanomedicine or as nanocarriers. In contrast, graphene oxide (GO), depicted in Figure 1c, is a highly oxidized form of graphene that combines hydrophobic sp2- and sp3-bonded carbon with numerous hydrophilic functional groups, resulting in an amphiphilic colloid with enhanced dispersibility in aqueous and polar solvents. GO's unique structure enables π-π interactions, weak hydrogen bonding, and strong electrostatic interactions, making it suitable for various chemical modifications and interactions with biological systems. Reduced graphene oxide (RGO), shown in Figure 1d, offers a balance between the properties of graphene and GO, while graphene quantum dots (GQDs) and graphene oxide quantum dots (GOQDs), illustrated in Figure 1e,f, are nanometer-sized fragments with unique optical properties, opening up new possibilities for biological applications such as cell imaging and bioelectrodes [22].

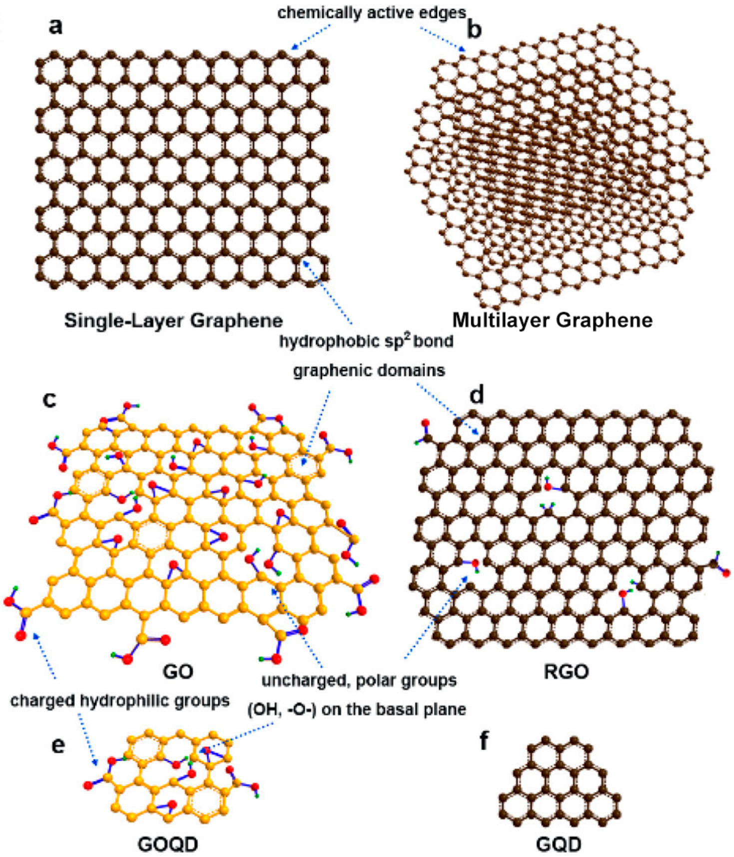

Figure 1. The forms of graphene and its derivatives that have been used for the construction of bioactive architectures: (**a**) single-layer graphene, (**b**) multilayer graphene, (**c**) GO, (**d**) RGO, (**e**) GOQD, (**f**) GQD. Adapted with permission from ref. [22]. Copyright 2017 ACS Publications.

2. Rheological Impact of Graphene on Polymeric Matrices

The incorporation of graphene into polymeric matrices induces profound rheological changes, fundamentally altering the physicochemical properties of the resulting nanocomposites. These modifications stem from the unique characteristics of graphene, particularly its high aspect ratio and surface area, which create a complex interplay with the polymer chains. The rheological impact manifests primarily through viscosity enhancement, non-Newtonian behavior, and altered viscoelastic properties, all of which are intricately connected and dependent on factors such as graphene concentration, polymer–graphene interactions, and processing conditions [23].

The addition of graphene typically leads to a significant increase in viscosity, attributed to the formation of a percolated network within the polymer matrix. This network restricts the mobility of polymer chains, resulting in a more viscous material. Concurrently, the nanocomposites exhibit pronounced non-Newtonian behavior, characterized by shear-thinning properties. This phenomenon is often described by models such as the Cross-Williams–Landel–Ferry (Cross-WLF), which capture the complex flow behavior of these materials. The shear-thinning nature arises from the alignment of graphene sheets and polymer chains under applied stress, facilitating easier flow at higher shear rates [23].

Viscoelastic properties, as indicated by storage modulus (G') and loss modulus (G''), undergo substantial changes with graphene incorporation. The enhanced storage modulus observed in many graphene–polymer systems signifies improved elastic behavior and structural integrity. These rheological modifications are intrinsically linked to the concentration of graphene, with studies showing that even small additions (0.5–1.0 wt%) can induce substantial changes in viscosity and viscoelasticity. As concentration increases, a more pronounced network structure forms, leading to more significant rheological alterations [24,25].

The nature and strength of polymer–graphene interactions play a crucial role in determining the rheological behavior of nanocomposites. Strong interfacial interactions, often evidenced by Fourier Transform Infrared (FTIR) spectroscopy studies, contribute to more effective stress transfer and enhanced rheological properties. These interactions are influenced by the processing conditions during nanocomposite preparation, including extrusion temperature, shear rate during mixing, and cooling rates, all of which affect graphene dispersion and, consequently, the rheological behavior of the composite [23,26]. The rheological changes induced by graphene incorporation have far-reaching implications for both material properties and processing considerations. The increased viscosity and enhanced viscoelastic response often correlate with improvements in mechanical properties, such as tensile strength, modulus, and hardness. These enhancements are attributed to the effective load transfer between the polymer matrix and the well-dispersed graphene sheets. Furthermore, the rheological modifications frequently coincide with enhanced thermal stability, as evidenced by shifts in melting temperatures and decomposition onset temperatures observed in thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC) studies [27].

While the increased viscosity of graphene–polymer nanocomposites can pose challenges for conventional processing techniques, it also presents opportunities for novel processing methods. The shear-thinning behavior, for instance, can be advantageous in extrusion-based additive manufacturing processes, allowing for improved flow through nozzles at high shear rates while maintaining structural integrity at rest. This characteristic exemplifies how the rheological impacts of graphene incorporation can be harnessed to enhance both material performance and processability [23].

Figure 2a–f presents a comprehensive analysis of the molecular characteristics, chemical structure, rheological behavior, and mechanical properties of PVBVA/graphene conductive patterns. Raman spectroscopy (Figure 2a) reveals the presence of graphene through characteristic D, G, and 2D bands, with a low ID/IG ratio indicating minimal structural defects. FTIR spectroscopy (Figure 2b) demonstrates hydrogen bonding between PVBVA and graphene, evidenced by a shift in hydroxyl peaks. Rheological studies (Figure 2c,d) show that the PVBVA/graphene ink exhibits higher viscosity and storage/loss moduli compared to pure PVBVA, resulting in improved printability and pattern precision. Tensile tests (Figure 2e,f) indicate significant enhancements in ultimate tensile strength and tensile modulus of the PVBVA/graphene patterns compared to pure PVBVA, with improvements of 98.7% and 177%, respectively. These results are contextualized within the existing literature, highlighting the exceptional mechanical reinforcement achieved with a relatively low graphene concentration [28].

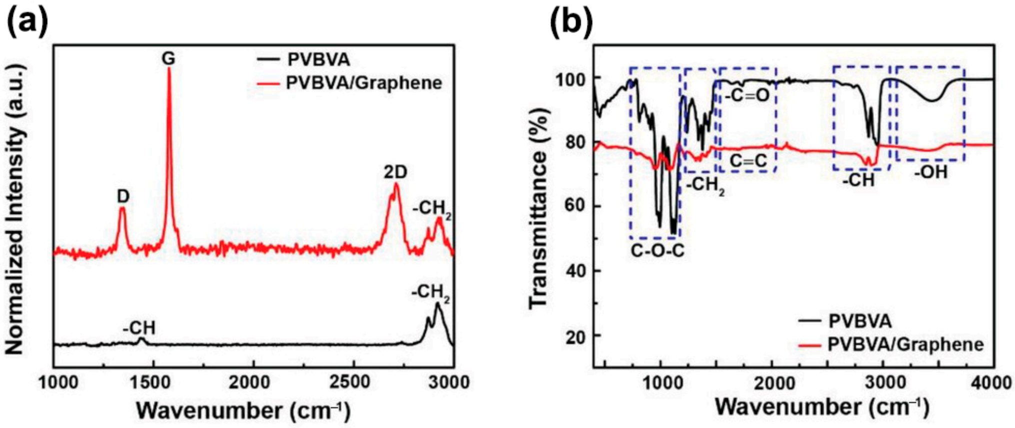

Figure 2. *Cont.*

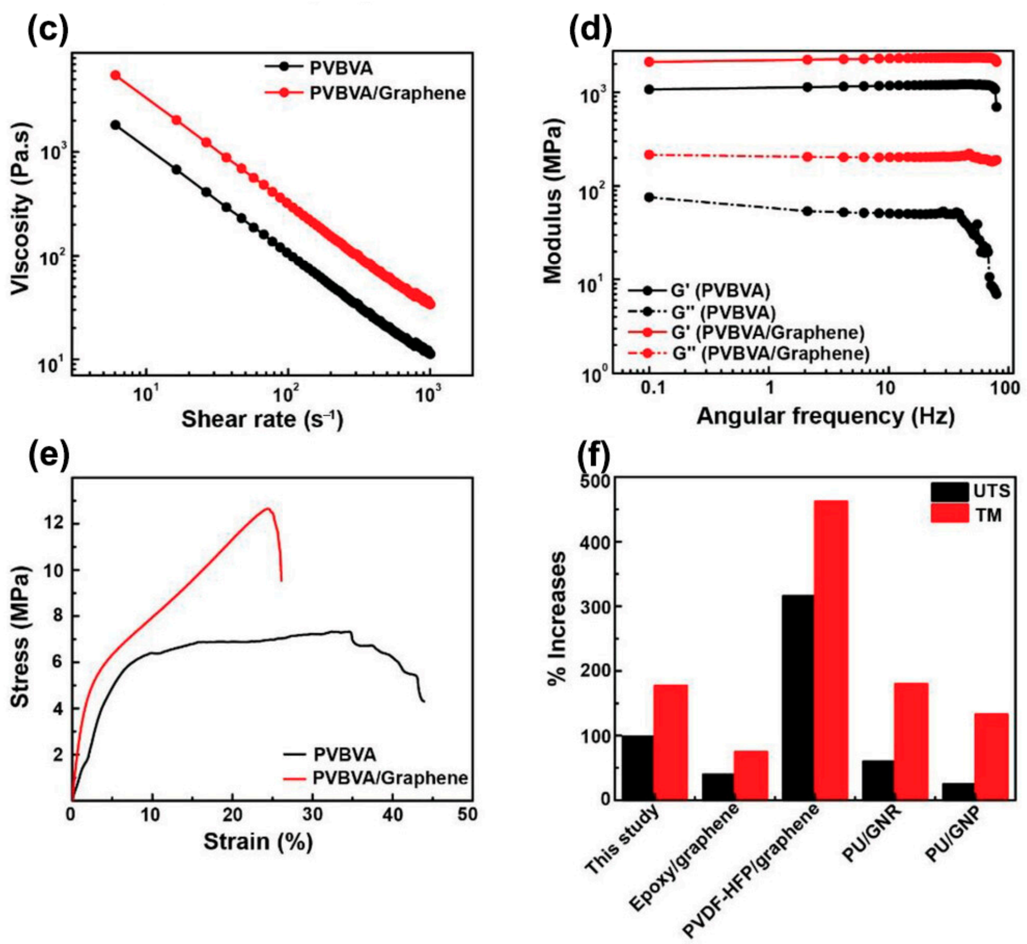

Figure 2. (**a**) Structural analysis of the PVBVA/graphene-based conductive ink using Raman spectra. (**b**) FTIR spectra indicating the interactions between the PVBVA polymer gel and graphene. Rheological behavior of the PVBVA/graphene-based conductive ink: (**c**) dynamic viscosity and (**d**) storage (G′) and loss moduli (G″) at various shear rates. (**e**) Tensile properties of the PVBVA/graphene-based conductive ink patterns: (**f**) comparison of the percentage increment in the ultimate tensile strength (UTS) and tensile modulus (TM) of the PVBVA/graphene-based nanocomposites with those reported in previous studies [28]. Reprinted with permission from ref. [28]. Copyright 2023 John Wiley & Sons.

3. Theoretical Framework

Nanocomposite theory provides a fundamental framework for understanding the complex interactions between graphene and cellulose matrices at the nanoscale level. This theory elucidates how the unique properties of graphene, particularly its nanoscale dimensions and high aspect ratio, profoundly influence the overall characteristics of the cellulose matrix. Recent advancements in research have shed light on the intricate mechanisms underlying nanocomposite theory in graphene–cellulose systems, offering deeper insights into their behavior and potential applications [24]. At the core of nanocomposite theory in graphene–cellulose systems are the interfacial interactions between the two components. These interactions, primarily hydrogen bonding and π-π interactions, occur between the oxygen-containing functional groups on the graphene oxide surface and the hydroxyl groups of cellulose nanofibrils. The significance of these interfacial interactions lies in their ability to facilitate efficient stress transfer between the cellulose matrix and graphene sheets, which is crucial for improving the overall mechanical strength and stiffness of the composite. Islam et al. (2024) conducted an extensive study using molecular dynamics simulations to investigate these interactions at the molecular level, revealing that the strength and nature of these bonds directly influence the composite's mechanical properties [29]. This understanding is crucial for optimizing the composite's performance, as it allows researchers to tailor the interfacial chemistry to achieve the desired mechanical characteristics. Also, the strength of these interfacial interactions directly influences the formation of percolation networks within the composite, another critical aspect of nanocomposite theory. He et al. (2022) made a significant breakthrough in this area by demonstrating that the percolation threshold in graphene–cellulose composites occurs at lower graphene concentrations than previously believed [30]. This finding has profound implications for the electrical properties of these composites, as the formation of a continuous pathway of graphene sheets throughout the cellulose matrix dramatically enhances electrical conductivity. The lower percolation threshold means that significant improvements in electrical conductivity can be achieved with smaller amounts of graphene, making these composites more economically viable and potentially expanding their range of applications. This discovery opens up new possibilities for developing lightweight, conductive materials for applications in flexible electronics, sensors, and electromagnetic shielding. Also, the mechanical reinforcement of cellulose matrices by graphene is a complex process involving several interrelated mechanisms, all of which are underpinned by the principles of nanocomposite theory. Jayatilaka et al. (2021) made significant strides in quantifying the stress transfer mechanism using in situ Raman spectroscopy, allowing for the direct measurement of stress transfer from the cellulose matrix to individual graphene sheets during mechanical loading [31]. Their findings revealed that the efficiency of stress transfer is highly dependent on the quality of interfacial bonding and the orientation of graphene sheets within the matrix, further emphasizing the importance of the interfacial interactions. This understanding is crucial for optimizing the composite's mechanical properties, as it allows researchers to design materials with enhanced load-bearing capabilities by controlling the orientation and distribution of graphene sheets within the cellulose matrix.

The recent breakthrough in visualizing crack propagation in graphene–cellulose composites in real time has been facilitated by advanced imaging techniques such as Digital Image Correlation (DIC). DIC has proven to be a powerful method for studying failure and crack propagation in advanced composite materials, including fiber-reinforced polymer (FRP) composites [32]. This technique allows for the determination of material deformation and damage, providing valuable insights into the behavior of composites under stress. In parallel, research on cellulose nanocrystal (CNC) matrices has shown that polymer nanocomposites containing self-assembled CNCs are ideal for applications requiring both strength and toughness, as the helicoidal structure of CNCs can deflect crack propagation while the polymer matrix dissipates impact energy [33]. Their study demonstrated the effectiveness of graphene sheets in deflecting cracks and enhancing fracture toughness by forcing cracks to follow a more tortuous path around the graphene sheets. This mechanism works in tandem with the pull-out mechanism studied by Ilyaei et al. (2021), who developed a novel method to quantify the pull-out energy of individual graphene sheets from the cellulose matrix [34]. The pull-out mechanism is a crucial energy dissipation process during fracture, contributing significantly to the overall toughness of the composite. These findings have important implications for designing materials with enhanced damage tolerance and resistance to crack propagation, which is particularly relevant for applications in structural materials and protective coatings. Also, integrating these various mechanisms, Zhu et al. (2024) introduced an innovative concept of hierarchical reinforcement in graphene–cellulose composites [35]. This multiscale approach involves using graphene to reinforce cellulose nanofibrils, which in turn reinforce the bulk cellulose matrix. The hierarchical structure allows for efficient stress transfer across multiple length scales, resulting in exceptional improvements in mechanical properties. This approach demonstrates how a comprehensive understanding of nanocomposite theory can lead to novel design strategies for high-performance materials. By leveraging the synergistic effects of reinforcement at different scales, researchers can develop materials with unprecedented combinations of strength, toughness, and lightweight characteristics.

Surface modification of graphene and cellulose plays a crucial role in enhancing their compatibility and tailoring the properties of the resulting composites, further expanding the applications of nanocomposite theory [36]. Cruz-Benítez et al. (2023) developed a green approach for the covalent functionalization of graphene oxide with cellulose nanocrystals using enzyme-mediated coupling [37]. This method resulted in improved dispersion of graphene in the cellulose matrix and enhanced mechanical properties of the composite. The covalent bonds formed between graphene oxide and cellulose nanocrystals create strong interfacial interactions, leading to more efficient stress transfer and better overall mechanical performance. This approach not only improves the composite's properties but also addresses environmental concerns by utilizing green chemistry principles. Also, complementing this covalent approach, Han et al. (2022) explored the use of bio-inspired polydopamine for the non-covalent functionalization of graphene [38]. This approach led to improved interfacial adhesion between graphene and cellulose, resulting in enhanced thermal conductivity in the composites. The non-covalent nature of this functionalization method allows for the preservation of graphene's intrinsic properties while still improving its compatibility with the cellulose matrix. This research highlights the importance of balancing the degree of functionalization with the preservation of graphene's unique properties to achieve optimal composite performance.

Further advancing our understanding of surface modification, Nekooei et al. (2023) investigated the use of cellulose derivatives for polymer wrapping of graphene [39]. Their study provided new insights into the molecular mechanisms of polymer adsorption on graphene surfaces, offering a way to improve the dispersion of graphene in cellulose matrices while potentially preserving the unique properties of graphene. This research highlights the importance of understanding molecular-level interactions in optimizing nanocomposite properties and demonstrates the potential for using cellulose-based materials to enhance the compatibility between graphene and cellulose matrices. Mohamed et al. (2018) developed a novel, environmentally friendly surfactant derived from cellulose for stabilizing graphene dispersions [40]. This approach led to improved colloidal stability and enhanced properties in the resulting composites. The use of cellulose-derived surfactants represents a sustainable approach to graphene functionalization, addressing concerns about the environmental impact of traditional synthetic surfactants while also improving the processing and fabrication of graphene–cellulose composites. This research demonstrates the potential for developing fully biobased composites with enhanced properties, aligning with the growing demand for sustainable materials in various industries.

Computational approaches, including molecular dynamics simulations and finite element analysis, have become increasingly important in predicting and optimizing the properties of graphene–cellulose composites [41,42]. Naddeo et al. (2018) proposed a hierarchical multiscale model that combined molecular dynamics simulations, micromechanical models, and finite element analysis to predict the mechanical properties of graphene–cellulose nanocomposites across multiple length scales [43]. This comprehensive approach demonstrates the power of computational techniques in advancing our understanding of these complex materials and provides a valuable tool for materials design and optimization. Petry et al. (2022) developed a machine learning model based on artificial neural networks to predict the thermal conductivity of graphene–cellulose aerogels [44]. Their model showed excellent agreement with experimental data and could be used to design composites with tailored thermal properties, highlighting the potential of data-driven approaches in materials design. This research demonstrates the growing importance of artificial intelligence and machine learning in materials science, offering new avenues for rapid materials discovery and optimization.

3.1. Theoretical Modeling and Simulation of Graphene–Cellulose Composites

The integration of graphene into cellulose matrices to form advanced composite materials has garnered significant attention in recent years due to the exceptional properties exhibited by these hybrid structures. To elucidate the complex interactions and resultant characteristics of graphene–cellulose composites, researchers have increasingly turned to sophisticated theoretical modeling and simulation techniques. These computational approaches offer invaluable insights across multiple scales, ranging from molecular-level interactions to macroscopic property predictions, thereby facilitating the rational design and optimization of experimental protocols.

3.2. Molecular Dynamics Simulations

Molecular dynamics (MD) simulations have been instrumental in understanding the interfacial interactions between graphene and cellulose at the atomic level [45]. Alqus et al. (2015) conducted MD simulations to study the interactions between hydrophobic and hydrophilic faces of ordered cellulose chains and a single layer of graphene in explicit aqueous solvent [46]. Their findings revealed that the hydrophobic cellulose face forms a stable complex with graphene, with the interface remaining solvent-excluded over the course of simulations. This study provided valuable insights into the amphiphilic nature of cellulose and its interactions with graphene in aqueous environments.

Figure 3 shows time series snapshots from a 300 ns molecular dynamics simulation of the graphene–cellulose (GC100) interface. Panel (a) provides a side view of the full system, illustrating the layered structure of cellulose on the graphene surface. Panels (b) and (c) offer top views of the graphene/layer 1 and graphene/layer 4 interfaces, respectively. These images demonstrate the stability of the hydrophobic cellulose face interacting with graphene, particularly in layer 1, while showing some flexibility in the more solvent-exposed layer 4. This figure highlights the strong complementarity between the hydrophobic cellulose face and graphene, which is crucial for understanding the behavior and properties of graphene–cellulose composites [46].

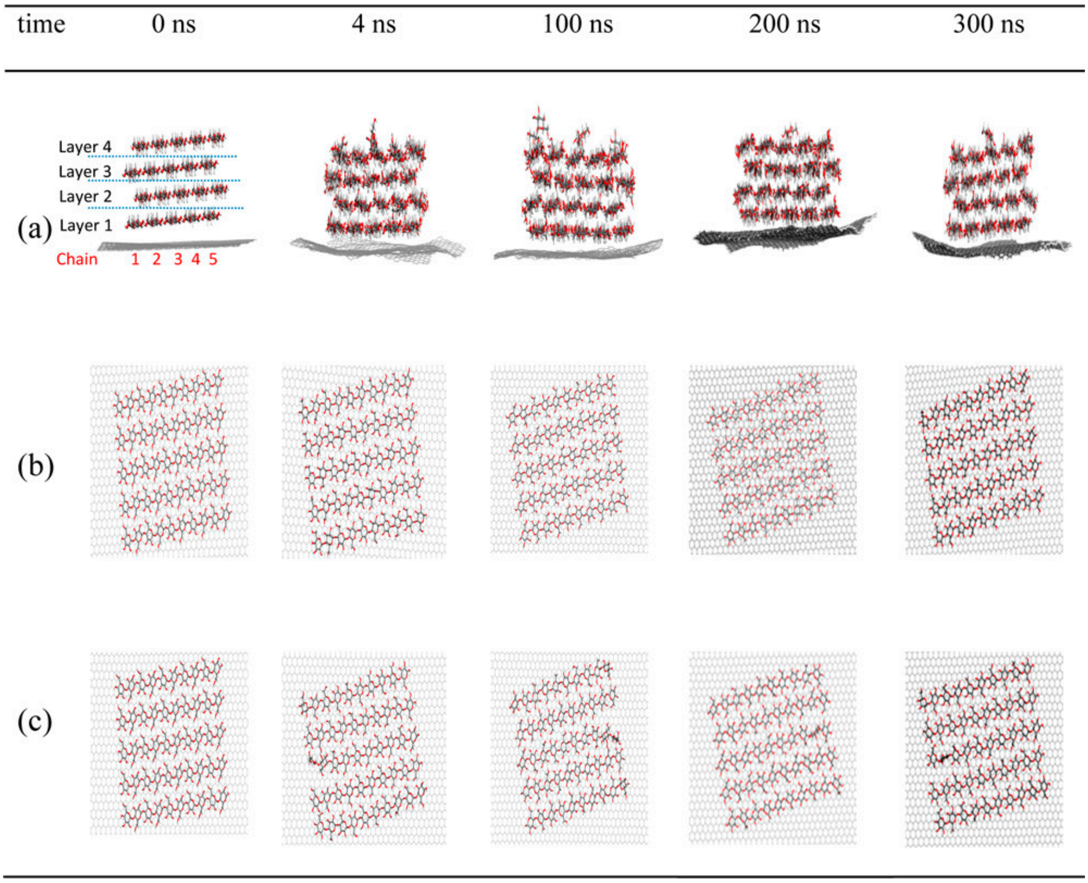

Figure 3. Time series of configurations sampled from 300 ns MD simulation of GC100. Structures shown are of (**a**) full system (side view) and top views of (**b**) graphene/layer 1 and (**c**) graphene/layer 4 [46]. Reprinted with permission from ref. [46]. Copyright ACS 2015 Publications.

3.3. Finite Element Analysis

The field of finite element analysis (FEA) for graphene–cellulose composites is an emerging area of research that combines the unique properties of graphene with the abundant and renewable nature of cellulose. While direct studies on the FEA of graphene–cellulose composites are limited, recent advancements in related areas provide valuable insights into potential developments in this field. S. Wang et al. (2023) demonstrated the application of FEA to graphene-reinforced composites by studying the properties of Ag-GNP silver–graphene composites and analyzing the electrical contact coupling field [47]. This approach could potentially be adapted for graphene–cellulose systems. In a related study, Wei et al. (2023) developed a simplified finite element method for analyzing the resistance response of graphene composites, considering size distribution and agglomeration [48]. This method could be valuable for modeling graphene–cellulose composites. Also, Nukala et al. (2023) conducted a simulation of wood polymer composites using finite element analysis [49]. This study is particularly relevant as wood is a cellulosic material, and the techniques used could be adapted for graphene–cellulose composites. Advancements in FEA for related composite materials have also been made. For instance, Wu et al. (2023) developed a size-dependent FEM for analyzing the 3D free vibration of functionally graded graphene platelet-reinforced composite cylindrical micro shells [50]. Similarly, Tayebi et al. (2023) applied a full layerwise finite element method to analyze the free vibration of functionally graded composite rectangular plates reinforced with graphene nanoplatelets [51]. In the realm of cellulose-based composites, Sathishkumar et al. (2023) conducted numerical buckling analysis of cellulose microfibrils reinforced polymer composites [52]. This study demonstrates the application of FEA to cellulose-based materials, which could be extended to include graphene reinforcement.

3.4. Multiscale Modeling

Multiscale modeling has emerged as a powerful approach for understanding and predicting the properties of complex materials like graphene–cellulose composites across different length scales. This hierarchical modeling strategy aims to bridge the gap between atomic-scale interactions and macroscopic properties. For instance, Rafiee and Rafiee et al. (2020) used multiscale modeling to predict Young's modulus in agglomerated graphene–polymer composites, focusing on capturing the agglomeration phenomenon [53]. C. Zhu et al. (2018) explored cellulose nanofiber–graphene oxide biohybrids using advanced microscopy and ReaxFF simulations to study self-assembly and copper-ion adsorption, providing insights into the interactions at the molecular level [54].

3.5. Density Functional Theory (DFT)

Density Functional Theory (DFT) calculations have indeed enriched recent studies on cellulose–graphene composites, providing valuable insights into their interactions and properties at the molecular level [55]. A study utilized DFT to investigate the electronic properties of graphene quantum dots with various substitutional impurities, which could be relevant for cellulose–graphene nanocomposites [56]. Another study on cellulose acetate–graphene oxide composites used DFT simulations to understand the adsorption mechanisms between these materials, finding that hydrogen bonding from hydroxyl groups facilitates the adsorption process [57]. Research by Tachikawa et al. used DFT to investigate hydrogen storage mechanisms in alkali-doped graphene nanoflakes, which could potentially be applied to cellulose–graphene systems [58]. A study on dialdehyde cellulose–graphene oxide films used DFT calculations along with molecular docking to investigate their potential antimicrobial activity against COVID-19 [59].

3.6. Machine Learning Models

Recent advancements in artificial intelligence have led to the development of machine learning models for predicting and optimizing the properties of graphene–cellulose composites. For example, Petry et al. (2022) explored the use of machine learning to analyze the microscopic interactions between graphene oxide and cellulose, enhancing the understanding of binding strengths [44]. Balasubramani et al. (2023) applied machine learning models to optimize processes involving graphene oxide and microcrystalline cellulose, focusing on pharmaceutical applications [60]. Additionally, Champa-Bujaico et al. (2023) utilized support vector machines to predict properties of bionanocomposites made from crystalline nanocellulose and graphene oxide, demonstrating the potential of machine learning in material design and optimization [61]. These studies highlight the transformative impact of AI in advancing the capabilities of graphene–cellulose composites.

4. Optimization of Graphene Content

Determining the optimal graphene content in cellulose matrices is crucial for achieving the desired structural and functional performance. Excessive or insufficient graphene loading can lead to suboptimal properties, making it essential to identify the optimal graphene concentration. Several studies have employed various optimization techniques, including response surface methodology (RSM), particle swarm optimization (PSO), and artificial neural networks (ANN), to identify the optimal graphene content and processing conditions [62–64].

4.1. Response Surface Methodology (RSM)

RSM is a widely used statistical technique for optimizing multiple variables simultaneously. In the context of graphene–cellulose composites, RSM has been employed to optimize various properties and processing conditions [6]. Figure 4 illustrates the effects of various experimental parameters on chlorpyrifos (CPF) removal using a magnetic graphene oxide–carboxymethyl cellulose (MGOC) composite. The 3D plots show the following: (A) CPF concentration and contact time: higher CPF concentrations decreased removal efficiency due to increased competition for adsorption sites. (B) CPF concentration and MGOC dosage: increasing adsorbent dosage improved removal efficiency by providing more adsorption sites. (C) MGOC dosage and contact time: removal efficiency increased with contact time, reaching equilibrium at 45 min. (D) Solution pH: lower pH improved CPF removal due to electrostatic interactions between the positively charged MGOC surface and the negatively charged CPF molecules [65].

The adsorption mechanism involves diffusion, electrostatic interactions, hydrogen bonding, surface complexation, and π-π interactions between CPF and the MGOC composite.

4.1.1. Optimization of Thermal Conductivity and Viscosity

A study employed RSM to optimize the thermal conductivity and viscosity of hybrid nanofluids containing graphene nanoplatelets (GNPs) and cellulose nanocrystals (CNCs). The researchers utilized a central composite design (CCD) to investigate the effects of temperature, volume concentration, and nanofluid type on the thermal properties. The results demonstrated that the optimal thermal conductivity and viscosity values were achieved at specific combinations of these factors, highlighting the effectiveness of RSM in optimizing the graphene content and processing conditions [66,67]. Another study employed RSM to optimize the durability behaviors of engineered cementitious composites (ECCs) by incorporating graphene oxide (GO) and polyvinyl alcohol (PVA) fibers. The researchers used a CCD to establish thirteen mixtures with varying GO and PVA concentrations and evaluated eight response variables, including compressive strength, weight loss, and rapid chloride permeability. The analysis of variance effectively designed and evaluated response models, with high Rˆ2 values ranging from 84% to 99%, indicating a good fit between the experimental data and the predicted values [66,68,69].

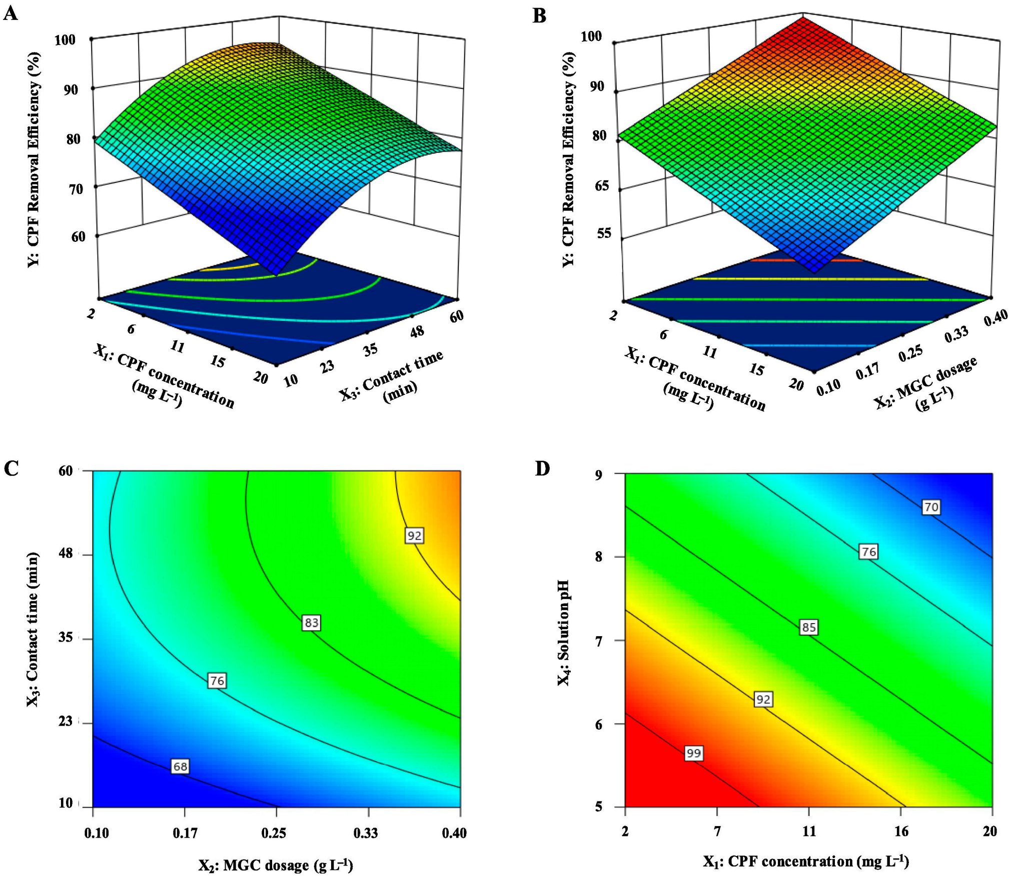

Figure 4. CPF removal efficiency as a function of (**A**) CPF concentration and contact time, (**B**) CPF concentration and MGOC dosage, (**C**) contact time and MGOC dosage, and (**D**) solution pH and CPF concentration (operating parameters set at their center points: CPF concentration of 11 mg/L, adsorbent dosage of 0.25 g/L, contact time of 35 min, and solution pH of 7). Reprint with permission from ref. [65]. Copyright 2022 Elsevier.

PSO is a computational optimization technique inspired by the social behavior of bird flocking or fish schooling. In the context of graphene–cellulose composites, PSO has been employed in conjunction with ANN to optimize various properties and applications [70,71]. One notable application of PSO in this field is the optimization of antidepressant removal from water using graphene oxide–cellulose nanogel composites. Researchers have employed PSO-based ANN modeling to fine-tune the composition and processing parameters of these composites, resulting in a more efficient removal of antidepressants like flupentixol from aqueous solutions [8]. The researchers employed a combination of particle swarm optimization (PSO) and artificial neural network (ANN) modeling to optimize the removal process [8]. This study presents a comprehensive approach to developing and optimizing a graphene oxide–cellulose nanogel composite for antidepressant removal. The researchers employed an artificial neural network (ANN) model to predict the composite's adsorption capacity, which was further enhanced through particle swarm optimization (PSO) to fine-tune input parameters. By optimizing process variables, including graphene oxide content, the team achieved improved removal efficiency for flupentixol. The successful application of the PSO-ANN methodology in tailoring graphene content demonstrates its potential for enhancing antidepressant removal in various applications, highlighting the synergy between advanced materials development and computational optimization techniques in environmental remediation efforts.

4.1.2. Optimization of Dye Removal

Another study by Khiam et al. utilized PSO and ANN to optimize the removal of methylene blue dye from aqueous solutions using a graphene oxide–cellulose nanocrystal composite. The researchers employed PSO to optimize the input parameters of the ANN model, which predicted the adsorption capacity of the composite. The optimized composite exhibited excellent dye removal efficiency, demonstrating the effectiveness of the PSO-ANN approach in optimizing the graphene content for environmental remediation applications [7]. A study concluded that the developed carboxymethyl cellulose–graphene oxide (CMC-GA-GO) biobased composite demonstrated high efficiency in removing methylene blue from wastewater, with maximum dye removal percentages ranging from 93 to 98%. The optimal conditions for maximum removal were found to be 20% GO weight percent in the composite, 25 min adsorption time, 25 °C adsorption temperature, 10–30 ppm initial MB concentrations, pH 7.0, and 0.2 g adsorbent dose. The adsorption process followed the Langmuir isotherm model and pseudo-second order kinetics, indicating monolayer adsorption and chemisorption, respectively. Thermodynamic analysis revealed the process was exothermic and spontaneous, working best at lower temperatures. The composite also showed good reusability, retaining 86% of its MB dye removal capacity after 10 cycles. Overall, the study concluded that the CMC-GA-GO composite shows promise as an inexpensive, reusable adsorbent for removing organic cationic dyes like methylene blue from industrial wastewater [12]. Another study employed ANN-PSO modeling to optimize methylene blue adsorption using graphene oxide–chitosan composites [7]. This approach allowed for precise prediction and optimization of the adsorption process.

Sulfo-functional 3D porous cellulose–graphene oxide composites have been developed as an innovative solution for water purification, specifically targeting the removal of methylene blue and tetracycline pollutants. These advanced materials leverage a three-dimensional structure to maximize surface area and enhance adsorption capabilities. The incorporation of sulfo-functional groups further augments the composites' affinity for the target contaminants. This dual-action approach enables the efficient elimination of both dye (methylene blue) and antibiotic (tetracycline) pollutants from water, showcasing the potential of these composites in addressing complex water treatment challenges [72]. Recent research in environmental remediation is exploring multifaceted approaches to pollutant removal [72]. Scientists are developing composites capable of simultaneously addressing multiple contaminants, including toxic metals and organic pollutants like phenol. These advanced materials, often incorporating graphene oxide, chitosan, and cellulose derivatives, demonstrate versatility in tackling various environmental challenges [73]. Additionally, the field is witnessing a growing integration of artificial intelligence and experimental design methodologies to optimize removal processes, signaling a shift towards more sophisticated and efficient pollution control strategies [74].

4.2. Artificial Neural Networks (ANN)

ANN is a machine learning technique that can model complex non-linear relationships between input and output variables. In the context of graphene–cellulose composites, ANN has been used in combination with PSO to optimize various properties and applications [8]. Figure 5 shows the optimal values of PSO parameters were obtained by conducting a sensitivity analysis for maximum iteration of 1000 [8].

In the study by Xiang et al., ANN was used in combination with PSO to optimize the removal of antidepressants using a graphene oxide–cellulose nanogel composite. The ANN model was trained on experimental data, and PSO was employed to optimize the input parameters, including the graphene oxide content, leading to improved adsorption performance [75]. This synergistic approach demonstrates the potential of ANN and PSO in optimizing the graphene content for specific applications. The optimization of dye removal processes has been extensively studied using various advanced techniques. One notable approach involves the use of response surface methodology (RSM) to optimize the adsorption of methylene blue dye using sulfonated graphene oxide impregnated cellulose acetate floated beads [6]. This method allows for the systematic evaluation of multiple parameters affecting dye removal efficiency. In another study, researchers developed a cost-effective and sustainable lemongrass membrane for dye removal, demonstrating excellent adsorption capabilities for methylene blue dye [76]. The kinetic study of this process revealed that the methylene blue adsorption followed a pseudo second-order model, providing valuable insights into the adsorption mechanism. Additionally, the incorporation of graphene oxide into the lemongrass membrane was explored, although the pure lemongrass membrane showed comparable dye adsorption performance [76]. These findings highlight the potential of using naturally sustainable materials in conjunction with advanced optimization techniques to enhance dye removal processes, offering promising solutions for wastewater treatment.

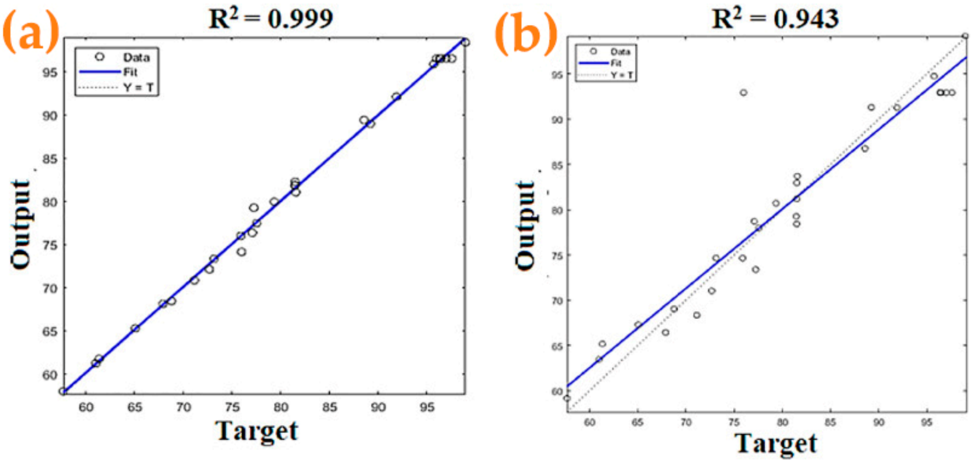

Figure 5. Regression plot for testing data (**a**) GO: S*pop* value of 50 and 4-8-1 network configuration (**b**) GOC: S*pop* value of 50 and 4-8-1 network configuration. Reprint from ref. [8]. Copyright 2020 Elsevier.

This flowchart (Figure 6) outlines the various optimization techniques used to determine the optimal graphene content in cellulose matrices. It includes the following key techniques and their specific applications:

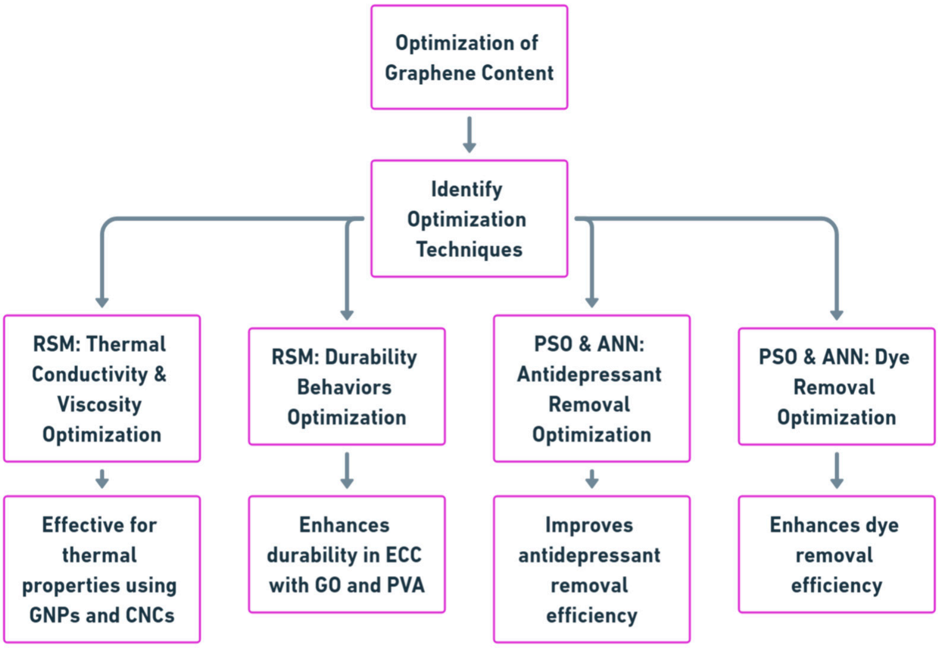

Figure 6. Optimization techniques for graphene content.

RSM (Response Surface Methodology): Used for optimizing thermal conductivity, viscosity, and durability behaviors. The flowchart highlights two branches of RSM applications: one for thermal properties using graphene nanoplatelets (GNPs) and cellulose nanocrystals (CNCs), and another for durability in engineered cementitious composites (ECC) with graphene oxide (GO) and polyvinyl alcohol (PVA) fibers [77,78].

PSO and ANN (Particle Swarm Optimization and Artificial Neural Networks): Employed together to enhance the removal efficiencies of pollutants such as antidepressants and dyes from aqueous solutions. These techniques optimize parameters in conjunction with ANN models that predict the adsorption capacity of graphene–cellulose composites.

This diagram visually communicates the strategic applications of these techniques in optimizing material properties and enhancing functional performance [79].

5. Quantitative Analysis of Structural and Functional Performance

The optimization of graphene content in cellulose matrices has been shown to significantly enhance the structural and functional performance of the resulting composites. Several studies have quantitatively analyzed the improvements in mechanical properties, thermal stability, electrical conductivity, barrier properties, and other relevant characteristics achieved through the incorporation of optimized graphene content [80,81].

5.1. Mechanical Properties

The addition of graphene to cellulose matrices has been demonstrated to improve the mechanical properties of the composites, such as tensile strength, Young's modulus, fracture toughness, and elongation at break [82,83]. Recent studies have demonstrated significant improvements in tensile properties of cellulose-based composites reinforced with graphene and its derivatives. A study on cellulose acetate–graphene oxide (CA/GO) films revealed that the addition of just 0.5 wt% GO led to a 64% increase in tensile strength and an 85% enhancement in strain-to-failure compared to pure cellulose acetate films. This remarkable improvement was attributed to the strong interfacial interactions between GO and the cellulose matrix, facilitated by hydrogen bonding [84]. Research has shown that carboxymethyl cellulose (CMC) functionalization can effectively improve the dispersion of graphene-based materials in polymer matrices. A study on epoxy nanocomposites reinforced with CMC-functionalized graphene nanoplatelets (GNPs) demonstrated enhanced tensile properties, with CMC-GNP reinforced composites exhibiting superior tensile strength compared to those reinforced with carbon nanotubes (CNTs) [85].

The incorporation of graphene-based materials has also been shown to enhance the compressive strength of cellulose composites. A recent study investigating the effects of graphene oxide (GO) and graphene nanoplatelets (GNPs) on cement-based composites found that the incorporation of 0.02 wt% GO and GNPs enhanced the flexural strength by 16.3% and 11.6%, respectivel [9]. The researchers attributed this improvement to the ability of GO and GNPs to fill cracks and form a more compact microstructure in cement mortars. The addition of graphene-based materials has been shown to significantly increase the Young's modulus of cellulose composites. A molecular dynamics simulation study on calcium silicate hydrate (C-S-H) composites, which are the main products of cement hydration, revealed that the incorporation of GO enhanced the Young's modulus by 32.1% [9]. This improvement was attributed to the formation of hydrogen bonds and the presence of Ca^{2+} ions near the interface, which improved interfacial adhesion and load transfer between GO and C-S-H. Graphene reinforcement has been shown to enhance the fracture toughness of cellulose-based composites. A study on chitosan-based composites containing optimized amounts of graphene oxide (GO) and cellulose nanocrystals (CNCs) observed a 216% improvement in toughness compared to pristine chitosan [86]. This significant enhancement was attributed to the synergistic effects of GO and CNCs, as well as their strong interfacial interactions with the chitosan matrix. The addition of graphene-based materials has been shown to improve the elongation at break of cellulose composites. A study on graphene oxide–polyacrylamide–carboxymethyl cellulose sodium (GO/PAM/CMC) nanocomposite hydrogels demonstrated improved elongation at break with the incorporation of GO [87]. The researchers found that hydrogen bonding between the oxygen-containing groups of GO and the N-H bonds of PAM resulted in a dense structure, leading to enhanced mechanical properties.

Figure 7 illustrates the mechanical properties of cellulose nanofiber–graphene oxide (CNGO) hybrid filaments: (a) The stress–strain curves demonstrate how the addition of graphene oxide (GO) affects the mechanical behavior of the filaments. As the GO concentration increases, the curves generally show higher stress values but lower strain at failure, indicating increased strength but reduced ductility. (b) This graph displays the relationship between GO concentration and two key mechanical properties: Young's modulus and tensile strength. Both properties show a similar trend, increasing significantly as GO content rises up to 5 wt%, then slightly decreasing at 7 wt%. This suggests an optimal GO concentration around 5 wt% for maximizing mechanical performance. (c) This plot compares the tensile strength of the CNGO hybrid filaments produced in this study to other reported values in the literature, with respect to filament diameter. It demonstrates that the CNGO filaments achieve competitive strength values, especially considering their relatively large diameter. The graph also indicates a general trend of increasing strength as filament diameter decreases, likely due to improved molecular alignment in thinner fibers [80].

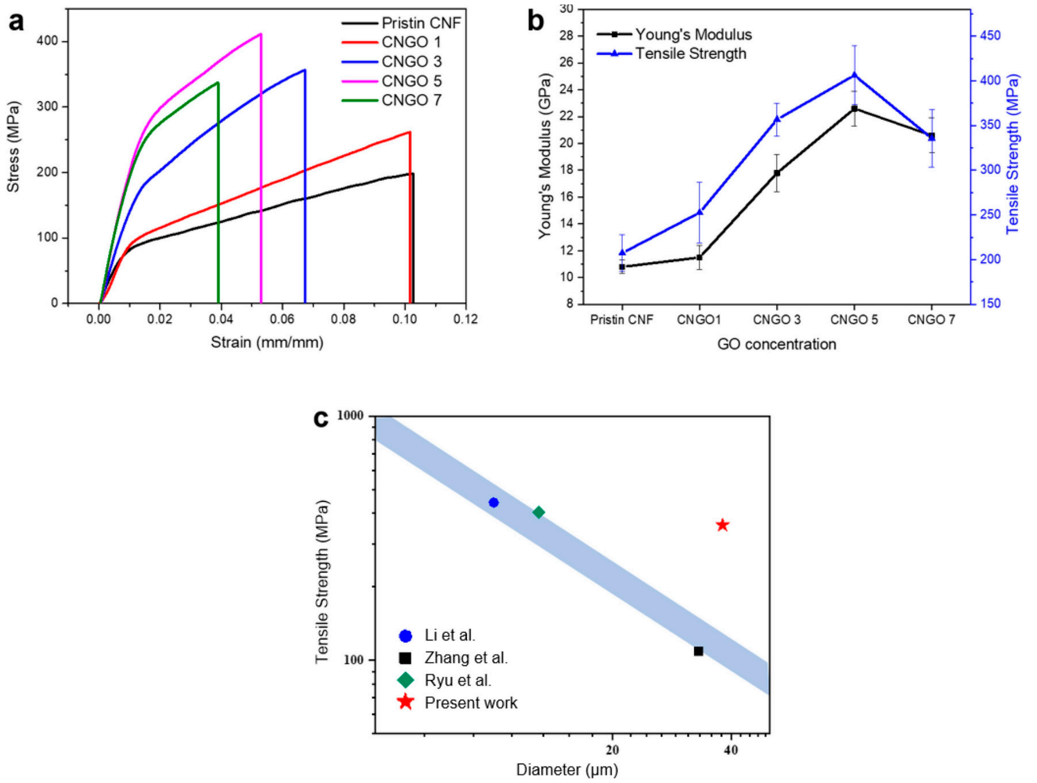

Figure 7. Mechanical properties of CNGO hybrid filaments: (**a**) stress–strain curves, (**b**) variations of Young's modulus and tensile strength with the GO concentration, (**c**) comparison of tensile strength with other reports in terms of filament diameter. Reprint from [80].

5.2. Thermal Stability

Graphene has been shown to improve the thermal stability of cellulose matrices by acting as a barrier to heat transfer and enhancing the char formation during thermal degradation. A study by Phiri et al. demonstrated that the incorporation of graphene into microfibrillated cellulose (MFC) composites resulted in a 22% enhancement in thermal stability at 9 wt% graphene loading [88]. The researchers observed that the graphene sheets formed a network structure within the MFC matrix, effectively hindering the movement of polymer chains and delaying the onset of thermal degradation. Another study by Shen et al. demonstrated that the incorporation of lysozyme-modified graphene nanoplatelets (LmGNP) into cellulose-based composites significantly enhanced the thermal conductivity, with a 297.3% increase observed for composites containing 8 wt% LmGNP compared to pure cellulose [89]. The researchers attributed this enhancement to the high thermal conductivity of graphene and the effective heat transfer pathways provided by the well-dispersed LmGNP in the cellulose matrix. Recent work by NI et al. investigated the thermal properties of unsaturated polyester composites reinforced with kenaf core fiber and hybrid nanofillers of cellulose nanocrystals (CNC) and graphene nanoplatelets. The study revealed that the incorporation of these nanofillers significantly improved the thermal stability of the composites. The maximum degradation temperature (T_{max} increased to 387.8 °C for samples containing both CNC and GNP, compared to the control samples without nanofillers [90].

Figure 8 presents the TG and DTG curves for NFC/GO-C and NFC/GO-E composites. The thermogravimetric analysis up to 800 °C shows a final weight loss of approximately 32.0% to 34.0%, primarily due to the decomposition of NFC and GO, including cellulose/hemicellulose and oxygen-containing functional groups (C=O, C-O-C, C-O-H). Above 500 °C, the weight-loss peak becomes less distinct due to biochar formation and the breakdown of C-C and C-H bonds. For NFC/GO-C composites, minor and major mass losses occur at 200 °C and 307 °C, while NFC/GO-E composites exhibit shoulder peaks at 205 °C and 241 °C. The thermal degradation process can be divided into two stages: 100–220 °C and 220–380 °C [91].

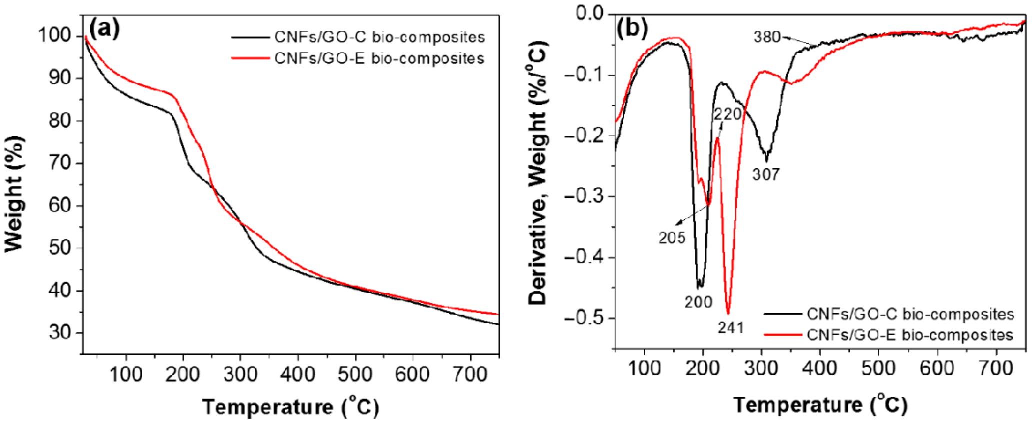

Figure 8. Thermogravimetric curves (TG), (**a**) and differential thermogravimetric curves (DTG), (**b**) of NFC/GO composites. Reprint from [91].

An innovative application of graphene–cellulose composites in thermal energy storage was explored by Hekimoğlu et al. The researchers developed a shape-stabilized microcrystalline cellulose/methyl stearate/graphene nanoplatelet composite for thermal energy storage applications [92]. The addition of graphene nanoplatelets not only enhanced the thermal conductivity of the composite but also improved its thermal stability and energy storage/release performance. The thermal stability of graphene–cellulose composites is significantly influenced by the concentration and dispersion of graphene within the cellulose matrix, as well as the interfacial interactions between these components [93]. While increasing graphene content generally enhances thermal stability, an optimal concentration exists beyond which further additions may not yield substantial improvements or may even result in detrimental effects due to agglomeration. Recent research has focused on developing innovative methods to improve graphene dispersion and strengthen interfacial interactions, such as surface modification techniques, the use of compatibilizers, and chemical functionalization of graphene or cellulose surfaces. These strategies aim to optimize the thermal stability of the composite by ensuring uniform distribution of graphene and maximizing the synergistic effects between the graphene and cellulose components [84].

5.3. Electrical Conductivity

The incorporation of graphene into cellulose matrices can impart electrical conductivity to the otherwise insulating materials. The graphene/microfibrillated cellulose (MFC) composite films demonstrated enhanced electrical, thermal, and mechanical properties. These composites were prepared using a simple method of co-exfoliation of graphite in an MFC suspension by high-shear exfoliation, resulting in homogeneously dispersed pristine graphene in the MFC matrix without chemical treatment. The composites achieved a high electrical conductivity of 2.4 S/m, although the specific graphene loading for this conductivity was not mentioned. At 0.5 wt% graphene loading, the specific surface area of the composites increased from 218 to 273 m^2/g, while the tensile strength and Young's modulus improved by 33% and 28%, respectively [88]. A review by Pottathara et al. explored the potential of graphene-based composites for flexible electronics, highlighting their superior mechanical strength, conductivity, and extraordinary thermal stability. The researchers discussed various strategies for incorporating graphene into polymer matrices, such as solution mixing, in situ polymerization, and layer-by-layer assembly, and their impact on the electrical and mechanical properties of the resulting composites [94]. Figure 9 illustrates the relationship between electrical conductivity and reduced graphene oxide (rGO) content in cellulose fibers. The experimental data are fitted using a percolation equation, with the derived parameters displayed in the inset. The percolation threshold (pc) of 2.46 wt.% exceeds typical values for cellulose-GO composites, likely due to the oriented nature of the fibers. The conductivity of the conductive phase (σo) is determined to be 73.3 $S \times cm^{-1}$, consistent with the values for rGO found in the literature. The critical exponent (t) of 3.5, while higher than theoretical predictions for 3D networks, is justifiable considering the potential non-uniform dispersion and orientation of rGO flakes within the fiber structure [95].

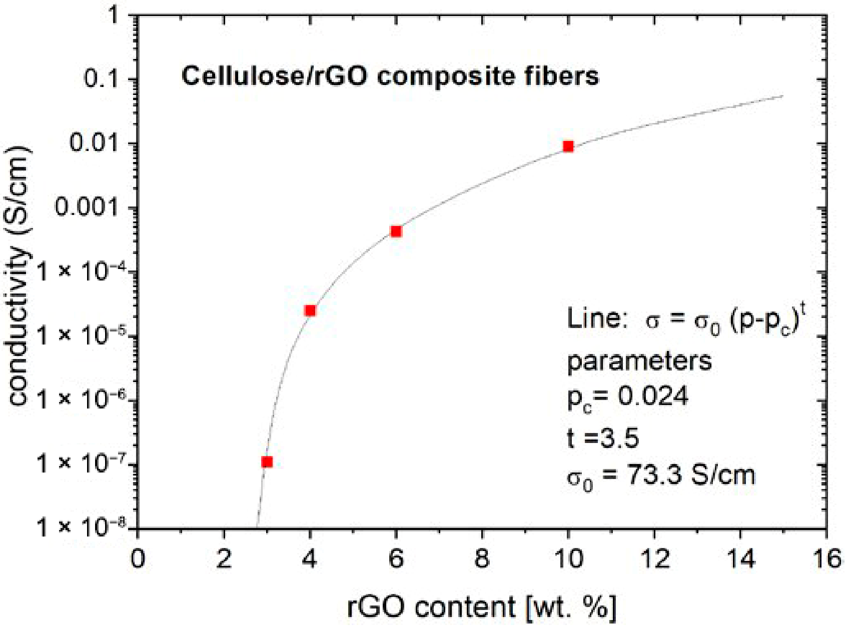

Figure 9. Dependence of electrical conductivity on the rGO content in cellulose fibers. The line represents theoretical values obtained using the percolation equation with the parameters shown in the inset. Reprinted from [95].

5.4. Barrier Properties

Graphene and its derivatives have been shown to improve the barrier properties of cellulose matrices, particularly against water vapor and gas permeation. In a study by Santillo et al., the researchers demonstrated that the presence of graphene oxide slightly increased the water barrier properties of chitosan-based composites, while the incorporation of CNCs and borate significantly reduced the water vapor permeability by about 50% compared to pristine chitosan. The researchers attributed this enhancement to the tortuous path created by the impermeable GO and CNC fillers, which hindered the diffusion of water vapor through the composite [96]. Another study by Qing et al. investigated the barrier properties of graphene oxide (GO)–cellulose nanofiber (CNF) composites for food packaging applications. The researchers found that the incorporation of GO significantly improved the oxygen barrier properties of the CNF films, with a 50% reduction in oxygen permeability observed for composites containing 1 wt% GO. The researchers attributed this enhancement to the high aspect ratio and impermeable nature of GO, which created a tortuous path for gas diffusion [97].

5.5. Biocompatibility and Tissue Engineering Applications

The biocompatibility and biodegradability of cellulose-based composites make them attractive candidates for tissue engineering applications. A review by Oprea et al. explored the potential of cellulose composites with graphene for tissue engineering, highlighting their ability to promote cellular attachment, growth, proliferation, and stem cell differentiation. The study discussed the preparation methods and biological performance of various cellulose-based composites, including cellulose paper, bacterial cellulose, and cellulose derivatives, with graphene, graphene oxide, and reduced graphene oxide [98]. These composites have shown promising results in developing multifunctional scaffolds for cell culture, bone, and neural tissue regeneration. A study by Y et al. investigated the potential of graphene oxide (GO)–cellulose nanofiber (CNF) composites for bone tissue engineering applications. The researchers found that the incorporation of GO into CNF scaffolds enhanced the attachment, proliferation, and osteogenic differentiation of human mesenchymal stem cells (hMSCs). The GO/CNF composites exhibited improved mechanical properties, electrical conductivity, and biocompatibility, making them promising candidates for bone tissue regeneration [99]. A notable study conducted in 2024 focused on the development of an injectable carboxymethyl chitosan (CMC)/hydroxyethyl cellulose (HEC)/β-tricalcium phosphate (β-TCP)/graphene oxide (GO) hydrogel for bone tissue engineering applications. The researchers employed a hydrothermal method to synthesize GO-functionalized bacterial cellulose, which was subsequently crosslinked with CMC and HEC. This innovative approach resulted in hydrogels with exceptional swelling properties and improved mechanical characteristics, particularly with the addition of 1 mg/mL GO. The study also investigated the osteogenic differentiation of rat bone marrow mesenchymal stem cells (rBMSCs), demonstrating significant promotion of osteogenesis, with the best effect observed at a GO concentration of 2 mg/mL [100].

The biocompatibility of these hydrogels was thoroughly evaluated through cell viability and proliferation assays. The results demonstrated good cell viability and proliferation, indicating the potential of these materials for tissue engineering applications. Furthermore, the hydrogels exhibited promising characteristics for bone tissue engineering, suggesting their potential use in clinical settings [100]. Another significant study in 2024 explored the development of polymer nanocomposites based on carbon nanotubes, graphene, and cellulose for fused deposition modeling (FDM) applications [101]. This research aimed to enhance the properties of FDM-printed parts by incorporating various nanofillers. The study highlighted the astonishing properties, biocompatibility, and ability to tailor the final performance of FDM-printed nanocomposite parts using these nanofillers. The researchers provided a comprehensive review of the processing, characterization, and potential applications of these nanocomposites in industrial settings [101]. The potential applications of graphene oxide in medical implants have also been the subject of recent research. A comprehensive review published in 2024 examined the use of nanomaterials, including graphene, in various medical implant applications [102]. The review focused on the advancements and challenges in employing nanomaterials to enhance the functionality of medical implants. Graphene, due to its unique electronic properties, was found to be particularly promising for applications such as electrical stimulation in neural implants.

The study highlighted the potential improvements in mechanical strength, biocompatibility, and drug delivery capabilities of implants incorporating nanomaterials like graphene [102]. In the realm of biosensors, a 2024 study utilized graphene oxide in the development of an electrochemical biosensor for the early detection of pancreatic cancer [103]. The researchers coated a gold working electrode with graphene oxide nano colloidal solution to create a nanoengineered biosensor capable of detecting pancreatic cancer-specific biomarkers. This innovative approach demonstrates the potential of graphene-based materials in advancing medical diagnostics and early disease detection [103].

5.6. Energy Storage and Environmental Remediation Applications

Graphene–cellulose composites are used to create high-performance supercapacitor electrodes. These composites offer high electrical conductivity and capacitance, making them suitable for flexible energy storage systems. For instance, chemically bonded polypyrrole/bacterial cellulose/graphene composites have demonstrated high volumetric and gravimetric capacitance with excellent cycling stability [104]. These composites serve as host matrices in lithium–sulfur batteries, enhancing the electrochemical performance by providing a stable structure and improving conductivity [105]. Flexible electrodes made from graphene–cellulose composites are used in lithium-ion batteries. They provide mechanical support and improved electrical conductivity, contributing to the overall efficiency and flexibility of the battery [106]. The morphology and characteristics of the self-supporting LFP/G/NFC electrode are vividly presented in Figure 10, offering a comprehensive view of its structure and properties. Figure 10a reveals the intricate composition of the electrode, where $LiFePO_4$ particles, graphene sheets, and nanofibrillated cellulose (NFC) fibers are clearly discernible. The graphene and filamentous NFC have combined to form a porous conductive framework, within which the homogeneous $LiFePO_4$ particles are well dispersed, albeit slightly aggregated.

A cross-sectional view of the flexible electrode is provided in Figure 10c, showcasing a clear and neat morphology. The absence of significant structural defects in this cross-section indicates stable binding between the components. This arrangement has resulted in a mesh-like three-dimensional structure with good electronic conductivity, which enhances the electronic contact at the electrode/electrolyte interface. The surface characteristics of the prepared LFP/G/NFC electrode are displayed in Figure 10b. Notably, the surface appears smooth and free from any flaws or peeling, suggesting a well-integrated structure. The electrode's flexibility is demonstrated in Figure 10d, where it exhibits high resilience through repeated bending. The electrode maintains its integrity even when bent to a curvature radius below 5 mm for 100 cycles, a testament to the sturdy frame provided by the graphene and NFC components. Figure 10e,f offer insight into the wettability of the electrode. These images compare the water contact angles of pure $LiFePO_4$ (68.9°) and the flexible LFP/G/NFC electrode (47.2°). The noticeably lower contact angle of the LFP/G/NFC electrode indicates superior electrolyte wettability compared to conventional $LiFePO_4$ electrodes. This enhanced wettability, combined with the electrode's flexibility and stable structure, suggests promising performance potential for this self-supporting LFP/G/NFC electrode in lithium-ion battery applications [106].

The high adsorption capacity and nanoporous structure of graphene–cellulose composites make them effective in filtering and purifying water. They can remove contaminants and pollutants efficiently due to their large surface area and reactivity [107]. These composites are used in phase change materials for solar energy applications. They enhance thermal conductivity and flexibility, which are crucial for efficient thermal energy storage and management [108].

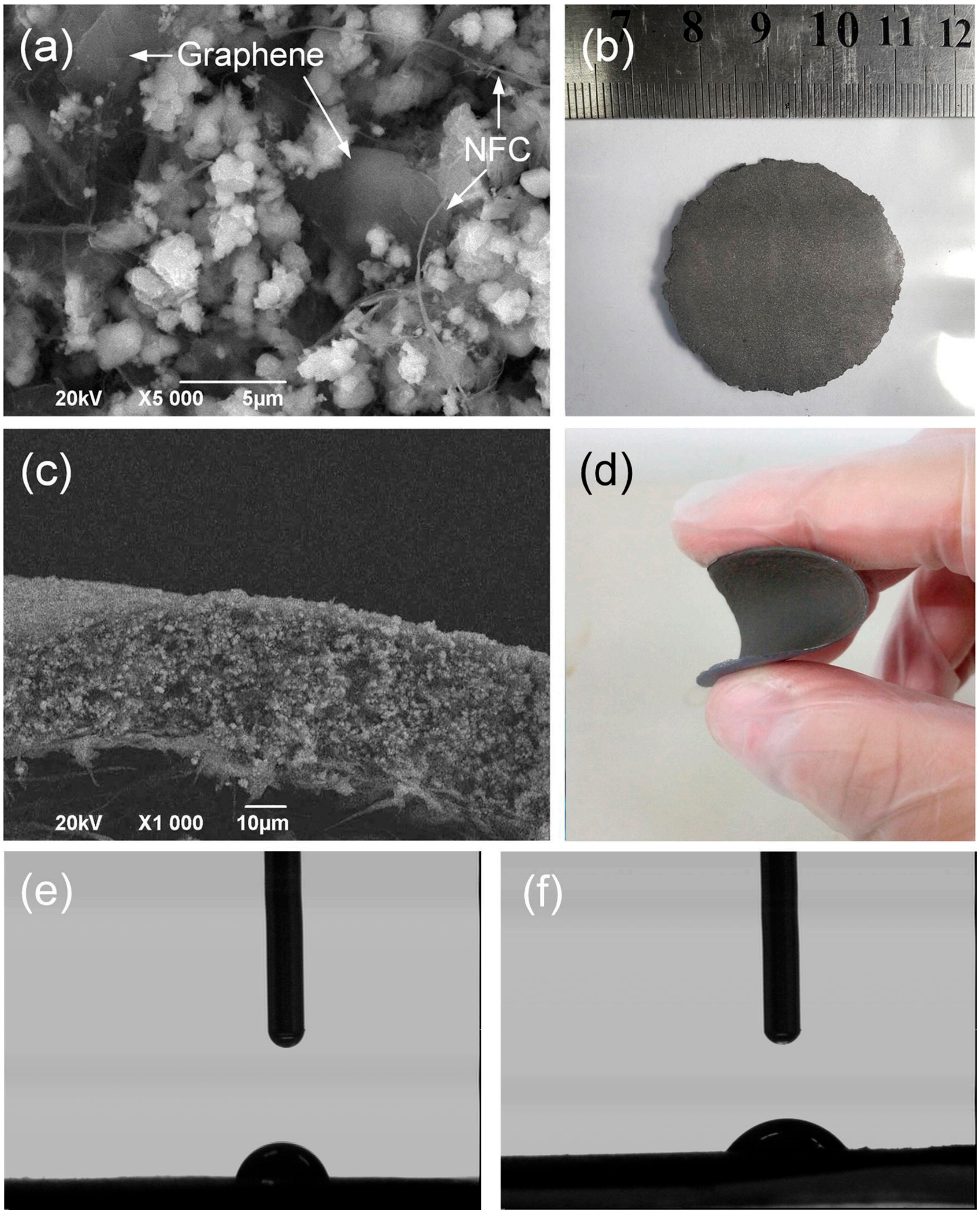

Figure 10. SEM images of (**a**) the surface and (**c**) the cross-section of a flexible LFP/G/NFC electrode; photos of a (**b**) flattened and (**d**) bent flexible LFP/G/NFC electrode; images of water contact angle on (**e**) pure $LiFePO_4$ and (**f**) flexible LFP/G/NFC electrode Reprint with permission from ref. [106]. Copyright 2018 Elsevier.

6. Challenges and Future Perspectives

While the incorporation of graphene into cellulose matrices has shown promising results in enhancing structural and functional properties, several challenges remain to be addressed. One of the main challenges is the uniform dispersion and prevention of agglomeration of graphene or its derivatives within the cellulose matrix. Agglomeration can lead to non-uniform property enhancements and potentially compromise the overall performance of the composite. Various strategies have been explored to address this challenge, including surface functionalization, solvent selection, and the use of dispersing agents or surfactants [109,110]. A study by Liu et al. investigated the effect of surface functionalization on the dispersion of graphene oxide (GO) in cellulose nanofiber (CNF) matrices. The researchers found that the introduction of carboxyl and hydroxyl groups on the GO surface improved its dispersion and interfacial interactions with the CNF matrix, resulting in enhanced mechanical and thermal properties of the composites [111]. Apart from graphene oxide, a large family of graphene derivatives bearing other functional groups—amines, thiols, sulfates, etc.—are known to date and are being employed, particularly for composite fabrication [112,113]. Another study by Li et al. explored the use of different solvents for the dispersion of graphene in microfibrillated cellulose (MFC) matrices. The researchers found that the use of N,N-dimethylformamide (DMF) as a solvent resulted in better dispersion and exfoliation of graphene, leading to improved mechanical and electrical properties of the composites compared to those prepared using water as the solvent [114].

Another challenge is the scalability and cost-effectiveness of the production processes. Many of the reported studies have been conducted on a laboratory scale, and the translation of these processes to industrial-scale production may require further optimization and cost considerations [115,116]. A study by Xie et al. explored the scalable production of graphene–cellulose composites using a continuous flow reactor [117]. The researchers developed a cost-effective and environmentally friendly method for the large-scale production of graphene–cellulose composites, which could potentially facilitate their commercialization and industrial applications [115,118]. Another study by Mendez et al. reported a cost-effective and scalable method for the production of graphene oxide (GO) using a modified Hummers method. The researchers demonstrated that the use of a more environmentally friendly oxidizing agent and the recycling of the oxidizing solution could significantly reduce the production cost of GO, making it more accessible for various applications, including the development of graphene–cellulose composites [119].

The long-term stability and durability of graphene–cellulose composites under various environmental conditions, such as temperature, humidity, and UV exposure, need to be thoroughly investigated to ensure their suitability for practical applications [120]. A study by Zhu et al. investigated the long-term stability and durability of graphene–cellulose composites under accelerated aging conditions. The researchers subjected the composites to elevated temperatures, humidity, and UV exposure to simulate long-term environmental conditions. The results showed that while the mechanical properties of the composites remained relatively stable, there was a gradual decrease in electrical conductivity over time, which was attributed to the degradation of the graphene network within the cellulose matrix [121]. To address the issue of long-term stability and durability, researchers have explored the use of protective coatings on graphene–cellulose composites [122]. A study by Xu et al. investigated the application of a polyurethane coating on graphene–cellulose composites for outdoor applications. The researchers found that the polyurethane coating effectively protected the composites from UV radiation, moisture, and other environmental factors, maintaining their mechanical and electrical properties over an extended period [123].

Despite the challenges, the field of graphene–cellulose composites holds immense potential for future research and development [35]. The exploration of novel processing techniques, such as 3D printing, electrospinning, and layer-by-layer assembly, could lead to the fabrication of composites with unique architectures and tailored properties for specific applications [124]. A study by Mohan et al. explored the 3D printing of graphene–cellulose composites for the fabrication of complex structures with tailored mechanical and electrical properties. The researchers developed a novel 3D printing technique that allowed for the precise control of the graphene distribution within the cellulose matrix, enabling the creation of composites with anisotropic properties and customized functionalities [125]. Another study by Jiang et al. investigated the electrospinning of graphene–cellulose nanofiber composites for tissue engineering applications. The researchers demonstrated that the incorporation of graphene into the electrospun cellulose nanofibers enhanced the mechanical properties, electrical conductivity, and biocompatibility of the resulting scaffolds, making them suitable for applications such as neural tissue regeneration and biosensing [126].

Furthermore, the incorporation of other functional materials, such as metal nanoparticles, carbon nanotubes, or conductive polymers, into graphene–cellulose composites could result in multifunctional materials with enhanced properties and expanded application potential [127]. A study by Zhu et al. explored the incorporation of silver nanoparticles into graphene–cellulose composites for antimicrobial applications. The researchers found that the synergistic effect of graphene and silver nanoparticles resulted in composites with enhanced mechanical properties, electrical conductivity, and antimicrobial activity, making them suitable for applications such as wound dressings and biomedical devices [128]. One notable study focused on the development of graphene oxide-functionalized bacterial cellulose–gelatin hydrogels for wound dressing applications. Ullah et al. (2023) created composite hydrogels by blending gelatin and graphene oxide–functionalized bacterial cellulose (GO-f-BC), using tetraethyl orthosilicate (TEOS) as a crosslinker. These hydrogels demonstrated impressive swelling capabilities in various media, with maximum swelling rates of 1902.83% in aqueous media, 1546.63% in PBS, and 1367.32% in electrolyte solutions. The composites also exhibited excellent hemocompatibility, with hemolysis rates below 0.5%, and showed antimicrobial activity against both Gram-positive and Gram-negative bacteria. Notably, cell viability and proliferation improved with increasing graphene oxide content, with optimal results observed in hydrogels containing 0.04 mg GO [129]. Another innovative approach was demonstrated by Xu et al. (2023), who developed a novel core–shell composite using pine needle-derived carbon fibers (PNCFs) and graphene for electromagnetic interference (EMI) shielding. Their study showcases the potential of combining biomass-derived carbon materials with graphene for advanced applications. The researchers created pore-rich PNCFs through KOH activation and integrated graphene using plasma-enhanced chemical vapor deposition (PECVD). The resulting composite achieved impressive electrical conductivity of 4.97 S cm^{-1} and excellent EMI shielding effectiveness, exceeding 70 dB over the X-band frequency range. Furthermore, the material showed promise for stealth applications due to its high absorption loss, which accounted for 90.8% of the total loss. This research underscores the potential of integrating biomass-derived carbon fibers with graphene to create high-performance EMI shielding materials [130].

7. Conclusions

The optimization of graphene content in cellulose matrices has proven to be a promising approach for enhancing the structural and functional performance of composite materials. Various optimization techniques, including RSM, PSO, and ANN, have been employed to identify the optimal graphene concentration and processing conditions. Quantitative analyses have demonstrated significant improvements in mechanical properties, thermal stability, electrical conductivity, barrier properties, and biocompatibility when the optimized graphene content is incorporated into cellulose matrices. These findings highlight the potential of graphene–cellulose composites for a wide range of applications, including structural materials, thermal insulation, conductive materials, tissue engineering, energy storage, and environmental remediation. However, challenges such as uniform dispersion, scalability, cost-effectiveness, and long-term stability need to be addressed to facilitate the practical implementation of these composites. Future research directions may include the exploration of novel processing techniques, the incorporation of additional functional materials, and the development of multifunctional composites with tailored properties for specific applications. Additionally, the investigation of long-term stability and durability under various environmental conditions, as well as the development of protective coatings or encapsulation strategies, could further enhance the practical applicability of these composites. Overall, the field of graphene–cellulose composites presents exciting opportunities for innovation and the creation of high-performance, sustainable, and multifunctional materials. Interdisciplinary collaborations among researchers, engineers, and industry partners will be crucial in overcoming the existing challenges and unlocking the full potential of these advanced composite materials.

Funding: This work and the project are realized with the financial support of the NSERC Discovery Grant RGPIN 1512/2021.

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

Abbreviations

ANN	artificial neural network
CNC	cellulose nanocrystal
CNF	cellulose nanofiber
CPF	chlorpyrifos
CVD	chemical vapor deposition
DIC	Digital Image Correlation
DSC	differential scanning calorimetry
ECC	engineered cementitious composites
FTIR	Fourier Transform Infrared spectroscopy
GO	graphene oxide
GOQD	graphene oxide quantum dots
GNP	graphene nanoplatelets
GQD	graphene quantum dots
MFC	microfibrillated cellulose
NFC	nanofibrillated cellulose
PHBV	Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)
PSO	particle swarm optimization
PVA	polyvinyl alcohol
RSM	response surface methodology
TGA	thermogravimetric analysis
UTS	ultimate tensile strength

References

1. Malnarič, I.; Krajnc, M.; Šebenik, U. Rheological study of hybrid aqueous suspension of TEMPO-oxidized cellulose nanofibrils and graphene oxide. *Cellulose* **2024**, *31*, 6105–6122. [CrossRef]
2. Ramezani, H.; Haji Ali Koohpayeh, M.; Tajedini, A.; Ramezani, G.; Mohseni, A. Nonlocal stability of curved carbon nanotubes conveying fluid based on Eringen's nonlocal elasticity theory in a thermomagnetic environment. *Acta Mech.* **2024**, *235*, 4273–4287. [CrossRef]
3. Luo, H.; Dong, J.; Yao, F.; Yang, Z.; Li, W.; Wang, J.; Xu, X.; Hu, J.; Wan, Y. Layer-by-Layer Assembled Bacterial Cellulose/Graphene Oxide Hydrogels with Extremely Enhanced Mechanical Properties. *Nano-Micro Lett.* **2018**, *10*, 42. [CrossRef] [PubMed]
4. Song, N.; Jiao, D.; Cui, S.; Hou, X.; Ding, P.; Shi, L. Highly Anisotropic Thermal Conductivity of Layer-by-Layer Assembled Nanofibrillated Cellulose/Graphene Nanosheets Hybrid Films for Thermal Management. *ACS Appl. Mater. Interfaces* **2017**, *9*, 2924–2932. [CrossRef] [PubMed]
5. Li, F.; Yu, H.-Y.; Wang, Y.-Y.; Zhou, Y.; Zhang, H.; Yao, J.-M.; Abdalkarim, S.Y.H.; Tam, K.C. Natural Biodegradable Poly(3-hydroxybutyrate-*co*-3-hydroxyvalerate) Nanocomposites with Multifunctional Cellulose Nanocrystals/Graphene Oxide Hybrids for High-Performance Food Packaging. *J. Agric. Food Chem.* **2019**, *67*, 10954–10967. [CrossRef] [PubMed]
6. Basha, I.K.; Abd El-Monaem, E.M.; Khalifa, R.E.; Omer, A.M.; Eltaweil, A.S. Sulfonated graphene oxide impregnated cellulose acetate floated beads for adsorption of methylene blue dye: Optimization using response surface methodology. *Sci. Rep.* **2022**, *12*, 9339. [CrossRef]
7. Khiam, G.K.; Karri, R.R.; Mubarak, N.M.; Khalid, M.; Walvekar, R.; Abdullah, E.C.; Rahman, M.E. Modelling and optimization for methylene blue adsorption using graphene oxide/chitosan composites via artificial neural network-particle swarm optimization. *Mater. Today Chem.* **2022**, *24*, 100946. [CrossRef]
8. Balasubramani, K.; Sivarajasekar, N.; Muthusaravanan, S.; Ram, K.; Naushad, M.; Ahamad, T.; Sharma, G. Efficient removal of antidepressant Flupentixol using graphene oxide/cellulose nanogel composite: Particle swarm algorithm based artificial neural network modelling and optimization. *J. Mol. Liq.* **2020**, *319*, 114371. [CrossRef]
9. Chen, Y.; Li, G.; Li, L.; Zhang, W.; Dong, K. Molecular dynamics simulation and experimental study on mechanical properties and microstructure of cement-based composites enhanced by graphene oxide and graphene. *Mol. Simul.* **2023**, *49*, 251–262. [CrossRef]
10. Tao, Y.; Du, J.; Cheng, Y.; Lu, J.; Min, D.; Wang, H. Advances in Application of Cellulose—MOF Composites in Aquatic Environmental Treatment: Remediation and Regeneration. *Int. J. Mol. Sci.* **2023**, *24*, 7744. [CrossRef]
11. Castellanos, H.G.; Aryanfar, Y.; Mohtaram, S.; Keçebaş, A.; Karaca-Dolgun, G.; Ahmad, S.; Asiri, A.N.M.; Islam, S. The efficacy of nano-cellulose-based composites in heavy metal removal from wastewater: A comprehensive review. *J. Chem. Technol. Biotechnol.* **2024**, jctb.7775. [CrossRef]

12. Mohamed, E.N.; Abd-Elhamid, A.I.; El-Bardan, A.A.; Soliman, H.M.A.; Mohy-Eldin, M.S. Development of carboxymethyl cellulose-graphene oxide biobased composite for the removal of methylene blue cationic dye model contaminate from wastewater. *Sci. Rep.* **2023**, *13*, 14265. [CrossRef] [PubMed]
13. Song, S.; Liu, Z.; Zhang, J.; Jiao, C.; Ding, L.; Yang, S. Synthesis and Adsorption Properties of Novel Bacterial Cellulose/Graphene Oxide/Attapulgite Materials for Cu and Pb Ions in Aqueous Solutions. *Materials* **2020**, *13*, 3703. [CrossRef] [PubMed]
14. D'Amora, U.; Dacrory, S.; Hasanin, M.S.; Longo, A.; Soriente, A.; Kamel, S.; Raucci, M.G.; Ambrosio, L.; Scialla, S. Advances in the Physico-Chemical, Antimicrobial and Angiogenic Properties of Graphene-Oxide/Cellulose Nanocomposites for Wound Healing. *Pharmaceutics* **2023**, *15*, 338. [CrossRef] [PubMed]
15. Qian, C.; Li, L.; Gao, M.; Yang, H.; Cai, Z.; Chen, B.; Xiang, Z.; Zhang, Z.; Song, Y. All-printed 3D hierarchically structured cellulose aerogel based triboelectric nanogenerator for multi-functional sensors. *Nano Energy* **2019**, *63*, 103885. [CrossRef]
16. Zhu, P.; Yu, Z.; Sun, H.; Zheng, D.; Zheng, Y.; Qian, Y.; Wei, Y.; Lee, J.; Srebnik, S.; Chen, W.; et al. 3D Printed Cellulose Nanofiber Aerogel Scaffold with Hierarchical Porous Structures for Fast Solar-Driven Atmospheric Water Harvesting. *Adv. Mater.* **2024**, *36*, 2306653. [CrossRef]
17. Santamaria, A.D.; Chandrasekaran, S.; Philbrick, O.; Worsley, M.A. 3D Printed Carbon Aerogels for Polymer-Electrolyte Fuel Cells. *ECS Trans.* **2022**, *108*, 153–163. [CrossRef]
18. Hashemi, S.A.; Ghaffarkhah, A.; Hosseini, H.; Arjmand, M. 3D-Printed Interfacially Jammed Emulsion Aerogels. *ACS Appl. Mater. Interfaces* **2024**, *16*, 46923–46936. [CrossRef]
19. Biswas, P.K.; Omole, O.; Peterson, G.; Cumbo, E.; Agarwal, M.; Dalir, H. Carbon and cellulose based nanofillers reinforcement to strengthen carbon fiber-epoxy composites: Processing, characterizations, and applications. *Front. Mater.* **2023**, *9*, 1089996. [CrossRef]
20. Sengupta, J.; Hussain, C.M. Advanced Graphene-Based Technologies for Antibiotic Removal from Wastewater: A Review (2016–2024). *C* **2024**, *10*, 92. [CrossRef]
21. Kasprzak, D.; Mayorga-Martinez, C.C.; Alduhaish, O.; Pumera, M. Wearable and Flexible All-Solid-State Supercapacitor Based on MXene and Chitin. *Energy Technol.* **2023**, *11*, 2201103. [CrossRef]
22. Cheng, C.; Li, S.; Thomas, A.; Kotov, N.A.; Haag, R. Functional Graphene Nanomaterials Based Architectures: Biointeractions, Fabrications, and Emerging Biological Applications. *Chem. Rev.* **2017**, *117*, 1826–1914. [CrossRef] [PubMed]
23. Tavares, L.; Sousa, L.R.; Silva, S.M.d.; Lima, P.S.; Oliveira, J.M. Effect of Incorporation of Graphene Nanoplatelets on Physicochemical, Thermal, Rheological, and Mechanical Properties of Biobased and Biodegradable Blends. *Polymers* **2023**, *15*, 3622. [CrossRef] [PubMed]
24. Hassanifard, S.; Behdinan, K. Impact of Rheology-Based Optimum Parameters on Enhancing the Mechanical Properties and Fatigue of Additively Manufactured Acrylonitrile-Butadiene-Styrene/Graphene Nanoplatelet Composites. *Polymers* **2024**, *16*, 1273. [CrossRef] [PubMed]
25. Narimani, A.; Kordnejad, F.; Kaur, P.; Bazgir, S.; Hemmati, M.; Duong, A. Rheological and thermal stability of interpenetrating polymer network hydrogel based on polyacrylamide/hydroxypropyl guar reinforced with graphene oxide for application in oil recovery. *J. Polym. Eng.* **2021**, *41*, 788–798. [CrossRef]
26. Kang, D.; Kim, S.H.; Shin, D.; Oh, J.T.; Kim, M.-G.; Lee, P.-C. Hygroscopic Behavior of Polypropylene Nanocomposites Filled with Graphene Functionalized by Alkylated Chains. *Nanomaterials* **2022**, *12*, 4130. [CrossRef]
27. Katti, P.; Verma, K.K.; Kumar, S.; Bose, S. Tuning the interface in epoxy-based composites and laminates through epoxy grafted graphene oxide enhances mechanical properties. *Nanoscale Adv.* **2021**, *3*, 6739–6749. [CrossRef]
28. Joshi, S.R.; Kumar, S.; Kim, S. Ecofriendly Polymer–Graphene-Based Conductive Ink for Multifunctional Printed Electronics. *Adv. Mater. Technol.* **2023**, *8*, 2201917. [CrossRef]
29. Islam, A.A. New wonder materials—Exciting technological horizon. *J. Bangladesh Acad. Sci.* **2024**, *48*, 1–25. [CrossRef]
30. He, F.; Yang, X.R.; Liu, Q.H.; Gong, X.Z. Preparation and Performance Evaluation of Graphene Heating Film. *Mater. Sci. Forum* **2022**, *1060*, 161–166. [CrossRef]
31. Jayatilaka, G.; Mohammadi, M.M.; Tehrani, M. Investigating Stress Transfer and Failure Mechanisms in Graphene Oxide-Cellulose Nanocrystals Films. In Proceedings of the American Society for Composites 2021, Online, 14–18 September 2021; Destech Publications, Inc.: Lancaster, PA, USA, 2021.
32. Naufal, M.I.; Wong, K.J.; Israr, H.A.; Nejad, A.F.; Rahimian Koloor, S.S.; Gan, K.W.; Faizi, M.K.; Siebert, G. Digital image correlation technique for failure and crack propagation of fibre-reinforced polymer composites—A review. *Compos. Adv. Mater.* **2024**, *33*, 26349833241253620. [CrossRef]
33. Vu, H.; Woodcock, J.W.; Krishnamurthy, A.; Obrzut, J.; Gilman, J.W.; Coughlin, E.B. Visualization of Polymer Dynamics in Cellulose Nanocrystal Matrices Using Fluorescence Lifetime Measurements. *ACS Appl. Mater. Interfaces* **2022**, *14*, 10793–10804. [CrossRef] [PubMed]
34. Ilyaei, S.; Sourki, R.; Akbari, Y.H.A. Capsule-based healing systems in composite materials: A review. *Crit. Rev. Solid State Mater. Sci.* **2021**, *46*, 491–531. [CrossRef]
35. Zhu, Z.; Tian, Z.; Liu, Y.; Yue, S.; Li, Y.; Wang, Z.L.; Yu, Z.; Yang, D. Human Nervous System Inspired Modified Graphene Nanoplatelets/Cellulose Nanofibers-Based Wearable Sensors with Superior Thermal Management and Electromagnetic Interference Shielding. *Adv. Funct. Mater.* **2024**, *34*, 2315851. [CrossRef]

36. Wang, N.; Li, Z.; Zhu, G.; Yu, Z.; Qin, R.; Zhou, J.; Lin, N. Chemical Modifications on Cellulose Nanocrystals for Composites: Surface Chemistry to Tailored Compatibility and Mechanical Enhancement. *Macromolecules* **2023**, *56*, 7505–7519. [CrossRef]
37. Cruz-Benítez, M.M.; Gónzalez-Morones, P.; Hernández-Hernández, E.; Villagómez-Ibarra, J.R.; Castro-Rosas, J.; Rangel-Vargas, E.; Fonseca-Florido, H.A.; Gómez-Aldapa, C.A. Covalent Functionalization of Graphene Oxide with Fructose, Starch, and Micro-Cellulose by Sonochemistry. *Polymers* **2021**, *13*, 490. [CrossRef]
38. Han, F.; Huang, H.; Liu, L. Dual bio-inspired strong and humidity-responsive composite cellulose nanofibril papers. *J. Mater. Sci.* **2022**, *57*, 8727–8738. [CrossRef]
39. Nekooei, A.; Miroliaei, M.R.; Shahabi-Nejad, M.; Sheibani, G.; Sheibani, H. Cellulose-wrapped graphene oxide as efficient adsorbents for pharmaceutical contaminants. *Inorg. Chem. Commun.* **2023**, *154*, 110997. [CrossRef]
40. Mohamed, A.; Ardyani, T.; Bakar, S.A.; Sagisaka, M.; Umetsu, Y.; Hussin, M.R.M.; Ahmad, M.K.; Mamat, M.H.; King, S.; Czajka, A.; et al. Preparation of conductive cellulose paper through electrochemical exfoliation of graphite: The role of anionic surfactant ionic liquids as exfoliating and stabilizing agents. *Carbohydr. Polym.* **2018**, *201*, 48–59. [CrossRef]
41. Dahiya, M.; Khanna, V.; Gupta, N. Review—Computational Studies of Graphene Reinforced Nanocomposites: Techniques, Parameters, and Future Perspectives. *ECS J. Solid State Sci. Technol.* **2024**, *13*, 061005. [CrossRef]
42. Ramezani, G.; Stiharu, I.; Van De Ven, T.G.M.; Nerguizian, V. Advancements in Hybrid Cellulose-Based Films: Innovations and Applications in 2D Nano-Delivery Systems. *J. Funct. Biomater.* **2024**, *15*, 93. [CrossRef]
43. Naddeo, F.; Baldino, L.; Cardea, S.; Naddeo, A.; Reverchon, E. Finite element multiscale modelling of elastic behavior of cellulose acetate—Graphene oxide nanocomposites, produced using a SC-CO2 assisted technique. *J. Supercrit. Fluids* **2018**, *140*, 248–257. [CrossRef]
44. Petry, R.; Silvestre, G.H.; Focassio, B.; Crasto de Lima, F.; Miwa, R.H.; Fazzio, A. Machine Learning of Microscopic Ingredients for Graphene Oxide/Cellulose Interaction. *Langmuir* **2022**, *38*, 1124–1130. [CrossRef]
45. Rahman, R.; Foster, J.T.; Haque, A. Molecular dynamics simulation and characterization of graphene-cellulose nanocomposites. *J. Phys. Chem. A* **2013**, *117*, 5344–5353. [CrossRef]
46. Alqus, R.; Eichhorn, S.J.; Bryce, R.A. Molecular Dynamics of Cellulose Amphiphilicity at the Graphene–Water Interface. *Biomacromolecules* **2015**, *16*, 1771–1783. [CrossRef]
47. Wang, S.; Wang, S.; Chen, S.; Li, A.; Ma, H.; Zhao, S.; Chen, Y.; Zhu, M.; Xie, M. Properties of Ag-GNP silver-graphene composites and finite element analysis of electrical contact coupling field. *J. Phys. Conf. Ser.* **2023**, *2587*, 012106. [CrossRef]
48. Wei, X.; Xia, Z.; Zhou, W.; Huang, P.; Fu, P. Simplified finite element method for resistance response of graphene composites considering size distribution and agglomeration. *J. Mater. Sci.* **2023**, *58*, 15696–15713. [CrossRef]
49. Nukala, S.G.; Kong, I.; Kakarla, A.B.; Patel, V.I.; Abuel-Naga, H. Simulation of Wood Polymer Composites with Finite Element Analysis. *Polymers* **2023**, *15*, 1977. [CrossRef] [PubMed]
50. Wu, C.-P.; Tan, T.-F.; Hsu, H.-T. A Size-Dependent Finite Element Method for the 3D Free Vibration Analysis of Functionally Graded Graphene Platelets-Reinforced Composite Cylindrical Microshells Based on the Consistent Couple Stress Theory. *Materials* **2023**, *16*, 2363. [CrossRef]
51. Tayebi, M.S.; Jedari Salami, S.; Tavakolian, M. Free vibration analysis of functionally graded composite rectangular plates reinforced with graphene nanoplatelets (GPLs) using full layerwise finite element method. *Proc. Inst. Mech. Eng. Part C J. Mech. Eng. Sci.* **2023**, *237*, 5865–5883. [CrossRef]
52. Senthilkumar, K.; Saba, N.; Chandrasekar, M.; Jawaid, M.; Rajini, N.; Alothman, O.Y.; Siengchin, S. Evaluation of mechanical and free vibration properties of the pineapple leaf fibre reinforced polyester composites. *Constr. Build. Mater.* **2019**, *195*, 423–431. [CrossRef]
53. Rafiee, R.; Eskandariyun, A. Predicting Young's modulus of agglomerated graphene/polymer using multi-scale modeling. *Compos. Struct.* **2020**, *245*, 112324. [CrossRef]
54. Zhu, C.; Monti, S.; Mathew, A.P. Cellulose Nanofiber-Graphene Oxide Biohybrids: Disclosing the Self-Assembly and Copper-Ion Adsorption Using Advanced Microscopy and ReaxFF Simulations. *ACS Nano* **2018**, *12*, 7028–7038. [CrossRef] [PubMed]
55. Hosseini, H.; Zirakjou, A.; McClements, D.J.; Goodarzi, V.; Chen, W.-H. Removal of methylene blue from wastewater using ternary nanocomposite aerogel systems: Carboxymethyl cellulose grafted by polyacrylic acid and decorated with graphene oxide. *J. Hazard. Mater.* **2022**, *421*, 126752. [CrossRef]
56. Méndez Martínez, K.M.; Nava Maldonado, F.M. DFT formalism studies on the structural and electronic properties of hexagonal graphene quantum dot with B, N and Si substitutional impurities. *Rev. Mex. Física* **2024**, *70*, 041601. [CrossRef]
57. Zhang, H.; Ding, L.; Zhang, Y.; Wu, T.; Li, Q. Adsorption of graphene oxide with cellulose acetate: Insights from DFT. *Mol. Phys.* **2022**, *120*, e2044083. [CrossRef]
58. Tachikawa, H. Hydrogen Storages Based on Graphene Nano-Flakes: Density Functional Theory Approach. *C* **2022**, *8*, 36. [CrossRef]
59. Dacrory, S. Antimicrobial Activity, DFT Calculations, and Molecular Docking of Dialdehyde Cellulose/Graphene Oxide Film Against COVID-19. *J. Polym. Environ.* **2021**, *29*, 2248–2260. [CrossRef]
60. Balasubramani, K.; Sivarajasekar, N.; Sarojini, G.; Naushad, M. Removal of Antidiabetic Pharmaceutical (Metformin) Using Graphene Oxide Microcrystalline Cellulose (GOMCC): Insights to Process Optimization, Equilibrium, Kinetics, and Machine Learning. *Ind. Eng. Chem. Res.* **2023**, *62*, 4713–4728. [CrossRef]

61. Champa-Bujaico, E.; Díez-Pascual, A.M.; Garcia-Diaz, P. Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) Bionanocomposites with Crystalline Nanocellulose and Graphene Oxide: Experimental Results and Support Vector Machine Modeling. *Polymers* **2023**, *15*, 3746. [CrossRef]
62. Soltani, H.; Karimi, A.; Falahatpisheh, S. The optimization of biodiesel production from transesterification of sesame oil via applying ultrasound-assisted techniques: Comparison of RSM and ANN–PSO hybrid model. *Chem. Prod. Process Model.* **2022**, *17*, 55–67. [CrossRef]
63. Ratnam, M.V.; Murugesan, M.P.; Komarabathina, S.; Samraj, S.; Abdulkadir, M.; Kalifa, M.A. Methylene Blue Adsorption BY UV-Treated Graphene Oxide Nanoparticles (UV/n-GO): Modeling and Optimization Using Response Surface Methodology and Artificial Neural Networks. *Int. J. Chem. Eng.* **2022**, *2022*, 1–13. [CrossRef]
64. Wang, W.; Wu, X.; Long, S. Optimizing the Methylene Blue Removalfrom Aqueous Solution Using Pomelo Peel BasedBiochar Assisted by RSM and ANN-PSO. *Pol. J. Environ. Stud.* **2021**, *31*, 329–346. [CrossRef]
65. Dolatabadi, M.; Naidu, H.; Ahmadzadeh, S. Adsorption characteristics in the removal of chlorpyrifos from groundwater using magnetic graphene oxide and carboxy methyl cellulose composite. *Sep. Purif. Technol.* **2022**, *300*, 121919. [CrossRef]
66. Bheel, N.; Mohammed, B.S.; Liew, M.S.; Zawawi, N.A.W.A. Effect of Graphene Oxide as a Nanomaterial on the Durability Behaviors of Engineered Cementitious Composites by Applying RSM Modelling and Optimization. *Buildings* **2023**, *13*, 2026. [CrossRef]
67. Yaw, C.T.; Koh, S.P.; Sandhya, M.; Ramasamy, D.; Kadirgama, K.; Benedict, F.; Ali, K.; Tiong, S.K.; Abdalla, A.N.; Chong, K.H. An Approach for the Optimization of Thermal Conductivity and Viscosity of Hybrid (Graphene Nanoplatelets, GNPs: Cellulose Nanocrystal, CNC) Nanofluids Using Response Surface Methodology (RSM). *Nanomaterials* **2023**, *13*, 1596. [CrossRef]
68. Huang, X.; Mo, Y.; Wu, W.; Ye, M.; Hu, C. Preparation and Properties of Waterborne Polyurethane/Carbon Nanotube/Graphene/Cellulose Nanofiber Composites. *Processes* **2024**, *12*, 1913. [CrossRef]
69. Perec, A.; Radomska-Zalas, A.; Fajdek-Bieda, A.; Pude, F. Process Optimization by Applying the Response Surface Methodology (Rsm) to the Abrasive Suspension Water Jet Cutting of Phenolic Composites. *Facta Univ. Ser. Mech. Eng.* **2023**, *21*, 575. [CrossRef]
70. Tan, H.; Xiao, D.; Navik, R.; Zhao, Y. Facile Fabrication of Polyaniline/Pristine Graphene–Bacterial Cellulose Composites as High-Performance Electrodes for Constructing Flexible All-Solid-State Supercapacitors. *ACS Omega* **2021**, *6*, 11427–11435. [CrossRef]
71. Kato, T.; Matsumoto, T.; Hongo, C.; Nishino, T. Mechanical and thermal properties of cellulose nanofiber composites with nanodiamond as nanocarbon filler. *Nanocomposites* **2018**, *4*, 127–136. [CrossRef]
72. Wang, S.; Ma, X.; Zheng, P. Sulfo-functional 3D porous cellulose/graphene oxide composites for highly efficient removal of methylene blue and tetracycline from water. *Int. J. Biol. Macromol.* **2019**, *140*, 119–128. [CrossRef]
73. Rahaman, H.; Islam, R.; Islam, R.; Alam, S.N.; Rahman, S.; Rahman, A.; Begum, B.A. Preparation, characterization, and adsorption kinetics of graphene oxide/chitosan/carboxymethyl cellulose composites for the removal of environmentally relevant toxic metals. *Int. J. Biol. Macromol.* **2024**, *257*, 128357. [CrossRef]
74. Cao, R.; Fan, M.; Hu, J.; Ruan, W.; Wu, X.; Wei, X. Artificial Intelligence Based Optimization for the Se(IV) Removal from Aqueous Solution by Reduced Graphene Oxide-Supported Nanoscale Zero-Valent Iron Composites. *Materials* **2018**, *11*, 428. [CrossRef]
75. Xiang, G.; Long, S.; Dang, A. Fabrication of the Ordered Mesoporous nZVI/Zr-Ce-SBA-15 Composites Used for Crystal Violet Removal and Their Optimization Using RSM and ANN–PSO. *Sustainability* **2022**, *14*, 6566. [CrossRef]
76. Cai, Z.; Remadevi, R.; Al Faruque, M.A.; Setty, M.; Fan, L.; Haque, A.N.M.A.; Naebe, M. Fabrication of a cost-effective lemongrass (*Cymbopogon citratus*) membrane with antibacterial activity for dye removal. *RSC Adv.* **2019**, *9*, 34076–34085. [CrossRef]
77. Bheel, N.; Mohammed, B.S.; Abdulkadir, I.; Liew, M.S.; Zawawi, N.A.W.A. Effects of Graphene Oxide on the Properties of Engineered Cementitious Composites: Multi-Objective Optimization Technique Using RSM. *Buildings* **2023**, *13*, 2018. [CrossRef]
78. Peng, Y.; Khaled, U.; Al-Rashed, A.A.A.A.; Meer, R.; Goodarzi, M.; Sarafraz, M.M. Potential application of Response Surface Methodology (RSM) for the prediction and optimization of thermal conductivity of aqueous CuO (II) nanofluid: A statistical approach and experimental validation. *Phys. A Stat. Mech. Its Appl.* **2020**, *554*, 124353. [CrossRef]
79. Solanki, D.; Sugandhi, S. A PSO-ANN Hybrid Model for Temporal Traffic Speed Forecasts in Urban Environments. *Int. J. Sci. Res. Eng. Manag.* **2024**, *8*, 1–6. [CrossRef]
80. Kim, H.C.; Panicker, P.S.; Kim, D.; Adil, S.; Kim, J. High-strength cellulose nanofiber/graphene oxide hybrid filament made by continuous processing and its humidity monitoring. *Sci. Rep.* **2021**, *11*, 13611. [CrossRef]
81. Sofyana; Muslim, A.; Supardan, M.D.; Ambarita, A.C.; Arahman, N. Combination of cellulose nanocrystal and graphene oxide as modifying agent for improving the performance of PVDF membranes. *Case Stud. Chem. Environ. Eng.* **2024**, *10*, 100873. [CrossRef]
82. Cicek Ozkan, B. Cellulose and chitosan biopolymer composites reinforced with graphene and their adsorption properties for basic blue 41. *Cellulose* **2022**, *29*, 9637–9655. [CrossRef]
83. Habibi, B.; Pashazadeh, S.; Pashazadeh, A.; Saghatforoush, L.A. An amplified electrochemical sensor employing one-step synthesized nickel–copper–zinc ferrite/carboxymethyl cellulose/graphene oxide nanosheets composite for sensitive analysis of omeprazole. *RSC Adv.* **2023**, *13*, 29931–29943. [CrossRef] [PubMed]
84. Gao, Z.; Li, Y.; Huang, P.; Zou, R.; Li, Y.; Fu, S. Graphene nanoplatelet/cellulose acetate film with enhanced antistatic, thermal dissipative and mechanical properties for packaging. *Cellulose* **2023**, *30*, 4499–4509. [CrossRef] [PubMed]

85. Zhang, D.; Huang, Y.; Xia, W.; Xu, L.; Wang, X. Dispersion characteristics and mechanical properties of epoxy nanocomposites reinforced with carboxymethyl cellulose functionalized nanodiamond, carbon nanotube, and graphene. *Polym. Compos.* **2024**, *45*, 398–412. [CrossRef]
86. Tabernero, A.; Baldino, L.; Cardea, S.; Martín Del Valle, E.; Reverchon, E. A Phenomenological Approach to Study Mechanical Properties of Polymeric Porous Structures Processed Using Supercritical CO_2. *Polymers* **2019**, *11*, 485. [CrossRef]
87. Zhang, H.; Zhai, D.; He, Y. Graphene oxide/polyacrylamide/carboxymethyl cellulose sodium nanocomposite hydrogel with enhanced mechanical strength: Preparation, characterization and the swelling behavior. *RSC Adv.* **2014**, *4*, 44600–44609. [CrossRef]
88. Phiri, J.; Johansson, L.-S.; Gane, P.; Maloney, T.C. Co-exfoliation and fabrication of graphene based microfibrillated cellulose composites—Mechanical and thermal stability and functional conductive properties. *Nanoscale* **2018**, *10*, 9569–9582. [CrossRef]
89. Shen, Y.; Zhang, X.; Su, J.; Lin, L.; Jiang, Z.; Qiu, L.; Wang, S.; Wu, B.; Pu, C.; Cai, X.; et al. Significantly Enhancing Mechanical and Thermal Properties of Cellulose-Based Composites by Adding Small Amounts of Lysozyme-Modified Graphene Nanoplatelets via Forming Strong Double-Cross-Linked Interface Interactions. *ACS Appl. Mater. Interfaces* **2023**, *15*, 43159–43168. [CrossRef]
90. Mustapha, N.I.; Mohamed, M.; Bakar, M.B.A.; Sobri, S.A. Thermal Properties of Unsaturated Polyester Reinforced Kenaf Core Fiber with Hybrid Nanofillers of Cellulose Nanocrystal and Graphene Nanoplatelet. *Borneo J. Sci. Technol.* **2023**, *5*, 75–83. [CrossRef]
91. Meng, X.; Wang, S.; Gao, W.; Han, W.; Lucia, L.A. Thermal pyrolysis characteristics and kinetic analysis of nanofibrillated cellulose/graphene oxide composites. *BioResources* **2020**, *15*, 4851–4865. [CrossRef]
92. Hekimoğlu, G.; Çakır, E.; Sarı, A.; Gencel, O.; Tyagi, V.V.; Sharma, R.K. Shape stabilized microcrystalline cellulose/methyl stearate/graphene nanoplatelet composite with enriched thermal conductivity and thermal energy storage/release performance. *Cellulose* **2023**, *30*, 10199–10214. [CrossRef]
93. Kashi, S.; Gupta, R.K.; Kao, N.; Hadigheh, S.A.; Bhattacharya, S.N. Influence of graphene nanoplatelet incorporation and dispersion state on thermal, mechanical and electrical properties of biodegradable matrices. *J. Mater. Sci. Technol.* **2018**, *34*, 1026–1034. [CrossRef]
94. Pottathara, Y. Graphene Based Composites with Cellulose Nanofibrils for Energy Storage Applications. 2017. Available online: https://www.semanticscholar.org/paper/Graphene-based-Composites-with-Cellulose-for-Energy-Pottathara/cd70dcdf88ae3abdc342aa4e85e726553377f6cf (accessed on 10 September 2024).
95. Piotr, K.; Jeszka, J.K.; Artur, M.; Leszek, S. Regenerated Cellulose/Graphene Composite Fibers with Electroconductive Properties. *Autex Res. J.* **2022**, *22*, 177–183. [CrossRef]
96. Santillo, C.; Wang, Y.; Buonocore, G.G.; Gentile, G.; Verdolotti, L.; Kaciulis, S.; Xia, H.; Lavorgna, M. Hybrid Graphenene Oxide/Cellulose Nanofillers to Enhance Mechanical and Barrier Properties of Chitosan-Based Composites. *Front. Chem.* **2022**, *10*, 926364. [CrossRef]
97. Qing, Y.; Sabo, R.; Cai, Z.; Wu, Y. Flexible Cellulose Nanofibril Composite Films with Reduced Hygroscopic Capacity. 2013. Available online: https://www.semanticscholar.org/paper/Flexible-cellulose-nanofibril-composite-films-with-Qing-Sabo/ab799ddf27e61a8f6f722b54a214362657c264c0 (accessed on 10 September 2024).
98. Oprea, M.; Voicu, S.I. Cellulose Composites with Graphene for Tissue Engineering Applications. *Materials* **2020**, *13*, 5347. [CrossRef]
99. Luo, Y.; Shen, H.; Fang, Y.; Cao, Y.; Huang, J.; Zhang, M.; Dai, J.; Shi, X.; Zhang, Z. Enhanced proliferation and osteogenic differentiation of mesenchymal stem cells on graphene oxide-incorporated electrospun poly(lactic-co-glycolic acid) nanofibrous mats. *ACS Appl. Mater. Interfaces* **2015**, *7*, 6331–6339. [CrossRef]
100. Zhu, Y.; Wang, T.; He, Z.; Liu, M.; Zhang, C.; Sun, G.; Wang, Q. Effect of Graphene Oxide in an Injectable Hydrogel on the Osteogenic Differentiation of Mesenchymal Stem Cells. 2024. Available online: https://pubmed.ncbi.nlm.nih.gov/39225005/ (accessed on 12 September 2024).
101. Sheikh, T.; Behdinan, K. Fused Deposition Modelling of Thermoplastic Polymer Nanocomposites: A Critical Review. *C* **2024**, *10*, 29. [CrossRef]
102. Cheng, Y. Advancements and challenges in nanomaterial-based medical implants. *Appl. Comput. Eng.* **2024**, *58*, 60–67. [CrossRef]
103. Karunanidhi, M.; Kanniyappan, H.; Zhan, E.; Mathew, R.; Sun, Y.; Wu, J.; Yan, Y.; Munirathinum, G.; Mathew, M.T. Abstract 7289: PancreaAlert: Intelligent nanoengineered biosensor for pancreatic cancer. *Cancer Res.* **2024**, *84*, 7289. [CrossRef]
104. Liu, Y.; Zhou, J.; Tang, J.; Tang, W. Three-Dimensional, Chemically Bonded Polypyrrole/Bacterial Cellulose/Graphene Composites for High-Performance Supercapacitors. *Chem. Mater.* **2015**, *27*, 7034–7041. [CrossRef]
105. Shen, Y.-D.; Xiao, Z.-C.; Miao, L.-X.; Kong, D.-B.; Zheng, X.-Y.; Chang, Y.-H.; Zhi, L.-J. Pyrolyzed bacterial cellulose/graphene oxide sandwich interlayer for lithium–sulfur batteries. *Rare Met.* **2017**, *36*, 418–424. [CrossRef]
106. Wang, Y.; He, Z.; Wang, Y.; Fan, C.; Liu, C.; Peng, Q.; Chen, J.; Feng, Z. Preparation and characterization of flexible lithium iron phosphate/graphene/cellulose electrode for lithium ion batteries. *J. Colloid Interface Sci.* **2018**, *512*, 398–403. [CrossRef]
107. Bacakova, L.; Pajorova, J.; Tomkova, M.; Matejka, R.; Broz, A.; Stepanovska, J.; Prazak, S.; Skogberg, A.; Siljander, S.; Kallio, P. Applications of Nanocellulose/Nanocarbon Composites: Focus on Biotechnology and Medicine. *Nanomaterials* **2020**, *10*, 196. [CrossRef]
108. Qian, Y.; Han, N.; Zhang, Z.; Cao, R.; Tan, L.; Li, W.; Zhang, X. Enhanced Thermal-to-Flexible Phase Change Materials Based on Cellulose/Modified Graphene Composites for Thermal Management of Solar Energy. *ACS Appl. Mater. Interfaces* **2019**, *11*, 45832–45843. [CrossRef]

109. Atif, R.; Inam, F. Reasons and remedies for the agglomeration of multilayered graphene and carbon nanotubes in polymers. *Beilstein J. Nanotechnol.* **2016**, *7*, 1174–1196. [CrossRef]
110. Gao, Y.-X.; Zou, J.-W.; Wang, X.-F.; Yang, J.; Li, Z.; Zhu, Y.-Y.; Wang, H.-M. An Approach to the Uniform Dispersion of Graphene Nanosheets in Powder Metallurgy Nickel-Based Superalloy. *Materials* **2019**, *12*, 974. [CrossRef] [PubMed]
111. Liu, Z.; Wang, Q.; Huang, X.; Qian, X. Surface Functionalization of Graphene Oxide with Hyperbranched Polyamide-Amine and Microcrystalline Cellulose for Efficient Adsorption of Heavy Metal Ions. *ACS Omega* **2022**, *7*, 10944–10954. [CrossRef]
112. Rabchinskii, M.K.; Sysoev, V.V.; Ryzhkov, S.A.; Eliseyev, I.A.; Stolyarova, D.Y.; Antonov, G.A.; Struchkov, N.S.; Brzhezinskaya, M.; Kirilenko, D.A.; Pavlov, S.I.; et al. A Blueprint for the Synthesis and Characterization of Thiolated Graphene. *Nanomaterials* **2021**, *12*, 45. [CrossRef]
113. Rabchinskii, M.K.; Besedina, N.A.; Brzhezinskaya, M.; Stolyarova, D.Y.; Ryzhkov, S.A.; Saveliev, S.D.; Antonov, G.A.; Baidakova, M.V.; Pavlov, S.I.; Kirilenko, D.A.; et al. Graphene Amination towards Its Grafting by Antibodies for Biosensing Applications. *Nanomaterials* **2023**, *13*, 1730. [CrossRef]
114. Li, M.; Xiao, M.; Wang, Q.; Zhang, J.; Xue, X.; Zhao, J.; Zhang, W.; Lu, C. Mechanically Strong and Electrically Conductive Polyethylene Oxide/Few-Layer Graphene/Cellulose Nanofibrils Nanocomposite Films. *Nanomaterials* **2022**, *12*, 4152. [CrossRef]
115. Sekwele, K.G.; Tichapondwa, S.M.; Mhike, W. Cellulose, graphene and graphene-cellulose composite aerogels and their application in water treatment: A review. *Discov. Mater.* **2024**, *4*, 23. [CrossRef]
116. Kuanyshbekov, T.; Guseinov, N.; Tolepov, Z.; Kurbanova, B.; Tulegenova, M.; Akatan, K.; Kantay, N.; Zhasasynov, E. Thermally Reduced Graphene Oxide Membranes From Local Kazakhstan Graphite "Ognevsky". *ChemistrySelect* **2023**, *8*, e202301746. [CrossRef]
117. Xie, Z.-T.; Asoh, T.-A.; Uetake, Y.; Sakurai, H.; Uyama, H. Dual roles of cellulose monolith in the continuous-flow generation and support of gold nanoparticles for green catalyst. *Carbohydr. Polym.* **2020**, *247*, 116723. [CrossRef] [PubMed]
118. Wan, Y.; Zhang, F.; Li, C.; Xiong, G.; Zhu, Y.; Luo, H. Facile and scalable production of three-dimensional spherical carbonized bacterial cellulose/graphene nanocomposites with a honeycomb-like surface pattern as potential superior absorbents. *J. Mater. Chem. A* **2015**, *3*, 24389–24396. [CrossRef]
119. Méndez-Lozano, N.; Pérez-Reynoso, F.; González-Gutiérrez, C. Eco-Friendly Approach for Graphene Oxide Synthesis by Modified Hummers Method. *Materials* **2022**, *15*, 7228. [CrossRef] [PubMed]
120. Chang, B.P.; Mohanty, A.K.; Misra, M. Studies on durability of sustainable biobased composites: A review. *RSC Adv.* **2020**, *10*, 17955–17999. [CrossRef]
121. Madhu, B.M.; Rashmi, T.V. Water Aging Effects on Graphene Nanoplatelets and Multi-walled Carbon Nanotube Reinforced Epoxy Glass Fiber Nanocomposites. *Indian J. Adv. Chem. Sci.* **2020**, *8*, 14–17.
122. Asghar, M.S.A.; Amir, M.; Hussain, U.; Sabri, M.M. Zinc and graphene oxide composites as new protective coatings for oil and gas pipes. *Polimery* **2023**, *68*, 378–385. [CrossRef]
123. Xu, W.; Lv, Y.; Kong, M.; Huang, Y.; Yang, Q.; Li, G. In-situ polymerization of eco-friendly waterborne polyurethane/polydopamine-coated graphene oxide composites towards enhanced mechanical properties and UV resistance. *J. Clean. Prod.* **2022**, *373*, 133942. [CrossRef]
124. Lyu, S.; Chen, Y.; Han, S.; Guo, L.; Chen, Z.; Lu, Y.; Chen, Y.; Yang, N.; Wang, S. Layer-by-layer assembled polyaniline/carbon nanomaterial-coated cellulosic aerogel electrodes for high-capacitance supercapacitor applications. *RSC Adv.* **2018**, *8*, 13191–13199. [CrossRef]
125. Mohan, D.; Sajab, M.S.; Kaco, H.; Bakarudin, S.B.; Mohamed Noor, A. 3D Printing of UV-Curable Polyurethane Incorporated with Surface-Grafted Nanocellulose. *Nanomaterials* **2019**, *9*, 1726. [CrossRef] [PubMed]
126. Ul-Islam, M.; Yasir, S.; Mombasawala, L.; Manan, S.; Wajid Ullah, M. Bacterial Cellulose: A Versatile Material for Fabrication of Conducting Nanomaterials. *Curr. Nanosci.* **2021**, *17*, 393–405. [CrossRef]
127. Rashid, A.B.; Haque, M.; Islam, S.M.M.; Uddin Labib, K.M.R. Nanotechnology-enhanced fiber-reinforced polymer composites: Recent advancements on processing techniques and applications. *Heliyon* **2024**, *10*, e24692. [CrossRef] [PubMed]
128. Yang, Y.; Zhou, R.; Yuan, M.; He, H. Nano Ag/PPy Biocomposites Based on Graphene Oxide Modified Bacterial Cellulose from the Juice of Xinhui Citrus and Its Antibacterial Activity. *Micromachines* **2023**, *14*, 1809. [CrossRef]
129. Khan, M.U.A.; Stojanović, G.M.; Hassan, R.; Anand, T.J.S.; Al-Ejji, M.; Hasan, A. Role of Graphene Oxide in Bacterial Cellulose−Gelatin Hydrogels for Wound Dressing Applications. *ACS Omega* **2023**, *8*, 15909–15919. [CrossRef]
130. Yang, Y.; Wan, C.; Huang, Q.; Hua, J. Pore-Rich Cellulose-Derived Carbon Fiber@Graphene Core-Shell Composites for Electromagnetic Interference Shielding. *Nanomaterials* **2022**, *13*, 174. [CrossRef]