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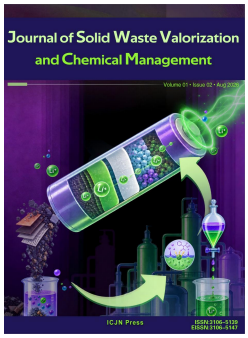

REVIEW

Preparation, Functionalization and Frontier Applications of Nanocellulose Hydrogels

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Abstract

Nanocellulose hydrogels are gaining traction as sustainable soft materials, thanks to the high surface area, abundant hydroxyl groups, mechanical strength, biocompatibility, and renewability of cellulose nanocrystals and nanofibers. Unlike conventional synthetic polymer hydrogels, those based on nanocellulose offer distinct advantages in building mechanically robust, multifunctional, and eco-friendly three-dimensional (3D) networks. This review highlights recent advances in the preparation, structural design, functionalization, and emerging applications of nanocellulose hydrogels. We begin by comparing major preparation routes — mechanical, chemical, chemical-mechanical and green solvent-based — in terms of efficiency, cost, environmental footprint, and scalability. Next, we discuss hydrogel fabrication strategies including physical and chemical crosslinking, double networks, interpenetrating polymer networks, and pore-structure control. Emphasis is placed on how interfacial interactions, energy dissipation, and network topology govern mechanical performance. Functionalization methods to introduce stimuli responsiveness, electrical conductivity, self-healing capability, and interfacial adhesion are then reviewed. Representative applications are covered in biomedical engineering, flexible electronics, smart devices, 3D printing, food packaging, and agricultural uses such as water retention and nutrient release. Finally, we critically examine key challenges: balancing mechanical strength with multifunctionality, dispersing nanofillers in multicomponent systems, limitations of current reinforcement theories, long-term stability, biosafety, and scalable green manufacturing. This review aims to guide the rational design, functional integration, and practical translation of nanocellulose hydrogels.

Keywords

Nanocellulose; Hydrogels; Double network structure; Functionalization; Flexible sensors; Green sustainable materials

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Highlights

- Nanocellulose hydrogels are reviewed from preparation to advanced applications.
- Structure-property relationships governing mechanics and function are clarified.
- NC, IPN, DN, and topological reinforcement strategies are critically compared.
- Dispersion, scalability, biocompatibility, and modeling remain key challenges.
- Biomimetic, green, and data-driven design define future research directions.

1. Introduction

1.1 Research Background and Significance

Hydrogels, as a class of hydrophilic polymeric materials featuring three-dimensional (3D) network structures, have long occupied a central position in soft matter science, biomedical engineering, and emerging intelligent devices owing to their high water content, soft texture, and tissue-like microenvironment[1,2]. Conventional hydrogels are predominantly constructed from synthetic polymers. Although their chemical structures can be precisely tailored, their intrinsic non-biodegradability imposes considerable environmental burdens. Moreover, the lack of biological activity presents fundamental limits

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to their advanced biomedical and sustainable applications. In addition, their performance drawbacks — including low toughness, poor stability, and insufficient mechanical robustness — cannot be ignored[3].

In the context of global efforts toward "carbon neutrality" and reducing dependence on fossil resources, the development of green functional materials derived from renewable biomass has become a frontier topic in materials science. Cellulose — the most abundant natural polymer on Earth with an annual biosynthesis capacity on the order of hundreds of billions of tons — is recognized as a promising candidate to replace petroleum-derived polymers[4]. Over the past decade, breakthroughs in nanotechnology have shifted cellulose utilization from the macroscopic toward the nanoscale. The resulting nanocellulose materials, mainly including cellulose nanocrystals (CNCs), cellulose nanofibrils (CNFs), and bacterial nanocellulose (BNC), not only fully retain the intrinsic biocompatibility, biodegradability, and facile chemical modifiability of cellulose, but also exhibit high aspect ratios, large specific surface areas, exceptional mechanical strength, and distinct optical properties due to nanoscale effects, enabling a qualitative leap in performance[5].

Nanocellulose can self-assemble into 3D networks via physical interactions and hydrogen bonding, aided by its inherent hydrophilicity and abundant functional groups. Owing to this, nanocellulose entities have emerged as ideal structural building blocks for hydrogel construction. Incorporating nanocellulose into hydrogel matrices is not a simple physical blending process; rather, nanocellulose can precisely serve as a rigid reinforcing skeleton or multifunctional crosslinking node. This strategy fundamentally addresses the long-standing challenges of conventional bio-based hydrogels, namely low mechanical strength and poor structural stability[1,6]. The resulting nanocellulose-based hydrogels integrate the reinforcement mechanisms of nanomaterials with the soft and hydrated characteristics of hydrogels, thereby demonstrating strategic potential as next-generation functional materials capable of responding to critical challenges in energy, environment, and healthcare. Particularly in biomedical applications, Curvello *et al.*[7] highlighted that the biomimetic design of nanocellulose-based hydrogels holds promise for reshaping clinical paradigms in soft tissue repair. Consequently, elucidating the structural design principles, functionalization strategies, and structure-performance relationships of nanocellulose-based hydrogels carries substantial academic significance and practical relevance for promoting the high-value utilization of biomass resources.

1.2 Current Research Progress

In recent years, driven by the convergence of advanced manufacturing technologies and macromolecular chemistry, research on nanocellulose-based hydrogels has experienced explosive growth, evolving from simple structural materials to multifunctional smart systems with sensing, actuation, and biomedical capabilities[8]. The research focus has progressively shifted from the early-stage fabrication of single-network systems toward the precise regulation of hierarchical porous architectures, multifunctional integration, and the exploration of intelligent responsiveness to complex environments.

With respect to fabrication methodologies and structural design, a variety of strategies have been developed, including physical crosslinking, chemical crosslinking, and interpenetrating double-network (DN) architectures. Physical crosslinking primarily relies on hydrogen bonding, electrostatic interactions, and coordination interactions, which collectively enable reversible and dynamic network structures in nanocellulose hydrogels[9]. Hydrogels constructed via this approach often exhibit pronounced shear-thinning behavior, which significantly facilitates their application as ink for 3D printing[10]. Sultan *et al.*[11] demonstrated that the rheological properties of cellulose nanofibril (CNF) hydrogels can be precisely tuned by adjusting CNF concentration and surface charge density, enabling high-fidelity shape retention during 3D printing. In contrast, chemical crosslinking introduces covalent network structures through the incorporation of small-molecule crosslinkers (e.g., glutaraldehyde, epichlorohydrin) or via photo-initiated radical polymerization, thereby imparting superior structural stability and solvent resistance to the hydrogels[12]. However, single-network hydrogels commonly suffer from the intrinsic trade-off between strength and toughness. To overcome this limitation, the DN strategy has been widely adopted, wherein a rigid nanocellulose network provides mechanical strength, while a soft polymeric network dissipates energy, resulting in a marked enhancement in fatigue resistance and mechanical robustness, as synergistic multi-network interactions significantly improve fracture energy and tensile strength[9,13].

In terms of functionalization and application exploration, surface chemical modification and composite formation are key methods for endowing nanocellulose hydrogels with specific functions, which are particularly prominent in the following aspects.

Mechanical Reinforcement and Biomimetic Materials: To meet the requirements of tissue engineering for extracellular matrix (ECM)-mimicking microenvironments and long-term mechanical support, nanocellulose hydrogels are frequently combined with materials such as alginates to develop high-performance bioinks that leverage their excellent shear-thinning behavior. This composite strategy not only significantly enhances the mechanical strength and shape fidelity of printed scaffolds, which means overcoming the limitations of pure nanocellulose systems in constructing complex organ-scale architectures, but also enables the precise fabrication of macroscopic structures such as human ear cartilage. These scaffolds provide homogeneous cell encapsulation environments and promote cellular proliferation and functional maturation, achieving dual biomimicry in both mechanical performance and biological function[7,14].

Flexible Sensing and Electronic Skin: The inherently poor electrical conductivity of conventional hydrogels restricts their direct application in electronic devices. To address this challenge, conductive hydrogels have been developed that endow hydrogels with electrical conductivity while preserving their intrinsic advantages[15-17]. This is typically achieved by integrating traditional conductive components — including metallic nanoparticles, conjugated polymers, and carbon nanotubes — into hydrogel matrices[18,19]. Nanocellulose-based conductive hydrogels, as advanced multifunctional materials, not only exhibit excellent electrical conductivity but also achieve multiple desirable features such as self-healing, anti-freezing, and self-adhesive properties through rational material design. These multifunctional characteristics enable broad application prospects in electronics, wearable devices, biomedical engineering, and environmental monitoring[19,20]. For instance, Liu *et al.*[21] incorporated nanocellulose into gelatin and polyacrylamide to construct a brittle-ductile double-network hydrogel. By replacing water molecules within the hydrogel using a mixed solution of zwitterionic betaine and ammonium sulfate through a simple solvent exchange process, a tough, highly conductive, and anti-freezing hydrogel was obtained. The synergistic effects of nanocellulose reinforcement and the "salting-out" effect induced by

gelatin and ammonium sulfate endowed the hydrogel with outstanding mechanical performance, while the zwitterionic betaine and ammonium sulfate conferred remarkable anti-freezing capability (down to -40 ℃) and high electrical conductivity (1.5 $S \cdot m^{-1}$).

Drug Delivery and Stimuli-Responsive Systems: Hydrogels share several physical characteristics with living tissues, including high water content, dense structures, and low interfacial tension with aqueous media, rendering them particularly attractive for biomedical applications, especially in drug delivery. Through grafting modifications of hydroxyl groups on nanocellulose surfaces, smart hydrogels responsive to pH, temperature, or light stimuli can be fabricated to achieve spatiotemporally controlled drug release[22,23]. For example, Liu *et al*.[24] reported a pH- and near-infrared (NIR)-responsive hydrogel for drug delivery and wound healing. In this system, polydopamine (PDA) was incorporated into a TEMPO-oxidized cellulose nanofibril (TOCNF) network via ionic crosslinking with calcium ions to form a PDA/TOCNF hydrogel. Under NIR irradiation or acidic conditions, tetracycline hydrochloride loaded onto PDA could be released on demand from the hydrogel matrix.

Despite these remarkable advances, several critical challenges remain. First, trade-off between high performance and multifunctionality: achieving the simultaneous integration of high mechanical performance and multifunctional properties remains intrinsically challenging. High strength and modulus typically originate from the high crystallinity and dense crosslinking of nanocellulose networks, whereas flexibility, stretchability, self-healing, conductivity, and stimulus responsiveness rely on low crosslink densities or dynamic reversible interactions. These requirements are often structurally competing, making it difficult to achieve compatibility. Although strategies such as nanocomposite architectures, double-network systems, and topological networks have partially alleviated this issue, most studies remain empirical in nature. A systematic understanding of nanofiller dispersion, interfacial interactions, and multicomponent coupling effects on macroscopic mechanical and functional responses is still lacking. Realizing the synergistic integration of "high performance-multifunctionality" through rational structural design thus remains a core scientific challenge.

Second, barriers to translation from laboratory-scale preparation to industrial production: the fabrication of nanocellulose-based hydrogels is still largely confined to laboratory-scale processes, with slow industrial progress. Nanocellulose production remains costly and energy-intensive: physical methods require high energy input, chemical methods pose risks of environmental pollution and equipment corrosion, while enzymatic approaches — although environmentally benign — suffer from high enzyme costs and long reaction times, limiting scalability. Furthermore, the crosslinkers, functional additives, and polymerization conditions required for hydrogel network construction are often complex and difficult to control at scale, potentially introducing biotoxicity or environmental burdens. Particularly for biomedical and flexible electronic applications, long-term stability, biosafety, and biodegradability require extensive validation. Consequently, the development of low-cost, green, sustainable, and scalable fabrication strategies is essential for advancing nanocellulose hydrogels toward industrialization[25].

Third, insufficient mechanistic understanding of structure-property relationships in complex systems: current understanding of the "structure-property-function" relationships in multilevel composite nanocellulose hydrogel systems remains limited. Although existing theoretical models — such as percolation theory, double-network theory, and colloidal reinforcement models — provide valuable guidance, they are typically based on idealized systems and fail to accurately capture the spatial distribution of nanofillers, interfacial chemical interactions, and dynamic network evolution. In real systems, multiscale coupling, hydrogen-bond rearrangement, and stress-transfer mechanisms between nanocellulose and polymer matrices are highly complex, leading to pronounced nonlinearity and time-dependent macroscopic behavior. The lack of quantitative descriptions of these relationships hinders predictive material design and limits engineering applications in high-end functional fields.

Fourth, environmental stability issues: most bio-based hydrogels are prone to dehydration or freezing under extreme conditions (e.g., ultra-low temperatures, high temperatures, or high salinity), resulting in functional degradation and severely restricting their use in outdoor or harsh environments.

In summary, nanocellulose-based hydrogels, owing to their unique multiscale structural features, tunable network architectures, and outstanding mechanical and functional potential, have emerged as a highly promising research direction in the field of biomass-derived functional materials. From a broader perspective, the field is undergoing a pronounced transition from a "materials preparation-oriented" paradigm to a "structure-function synergistic design-oriented" paradigm. Early studies primarily focused on feasibility verification and mechanical reinforcement effects, whereas recent research emphasizes network topology regulation, multiscale structure-property relationship elucidation, and systematic multifunctional integration. The research paradigm is progressively shifting from empirical tuning toward mechanism-driven design, highlighting the precise control of nanocellulose dispersion states, interfacial interactions, and crosslinking modes to achieve synergistic optimization of mechanical performance, environmental adaptability, and intelligent responsiveness. While these advances have promoted deeper applications in biomedical engineering and flexible electronics, critical challenges remain in understanding performance synergy mechanisms, decoding structure-property relationships, and realizing scalable and sustainable fabrication[2,15,26]. Accordingly, this review systematically summarizes the preparation methods of nanocellulose and its construction strategies within hydrogel systems, with a particular focus on network topology design, functionalization mechanisms, and diversified applications. Current challenges and theoretical controversies are further discussed, followed by perspectives on future development, aiming to provide insights and guidance for the rational design and high-value utilization of nanocellulose-based hydrogels.

Unlike previous reviews that have largely focused on fibrillated cellulose as sustainable technological materials, on cellulose-based therapeutics, or on cellulose nanocomposites from a supramolecular chemistry perspective, the present review centers specifically on nanocellulose hydrogels. We emphasize the full chain that links nanocellulose preparation, hydrogel network construction, functionalization mechanisms, and emerging applications. Going beyond a mere listing of individual preparation methods or application examples, we highlight how nanocellulose dispersion, interfacial interactions, crosslinking modes, and network topology work together to determine mechanical reinforcement, stimuli responsiveness, conductivity, self-healing ability, and overall application performance. Furthermore, we critically compare the advantages an d limitations of various structural design strategies — including nanocomposite networks, interpenetrating polymer networks, double networks, and topological hydrogels — with an eye toward scalability, stability, and practical translation.

2. Topological Design of Nanocellulose Hydrogels

2.1 Preparation of Nanocellulose

NC mainly comprises three representative types: CNCs, CNFs, and BNC[27]. For nanocellulose-based hydrogels, the fibrous morphology, aspect ratio, and dispersion state of nanocellulose play decisive roles in determining the reinforcement efficiency and overall performance of the hydrogel network[28]. To provide a visual overview of the overall preparation logic, **Figure. 1** illustrates the typical process from nanocellulose extraction to hydrogel network formation.

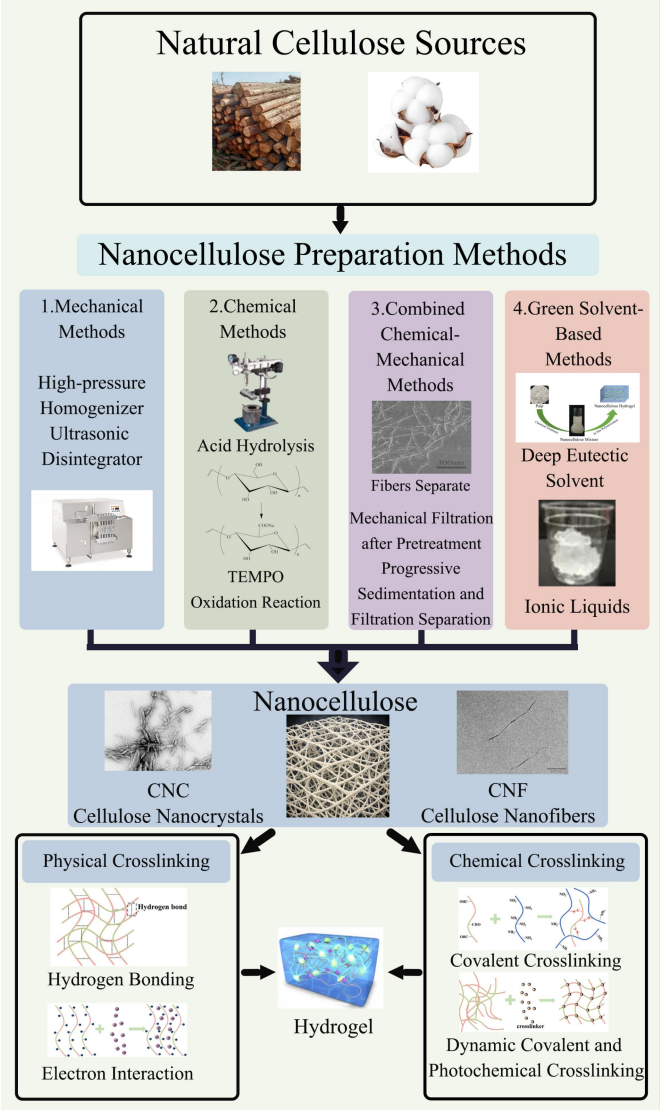

Figure. 1 Preparation process of nanocellulose hydrogels. Produced by Adobe Photoshop 2024 version 25.9.1.626 and ChemDraw 16 version 16.0.0.82. Reproduced with permission[29]. Copyright 2024, Elsevier. **NOTE:** CNC: cellulose nanocrystal; CNF: cellulose nanofibril.

2.1.1 Mechanical Methods

Mechanical methods primarily include high-pressure homogenization, grinding, and ultrasonic disintegration. These approaches rely on intense physical forces to delaminate and liberate cellulose microfibrils or crystalline domains from natural cellulose sources. Mechanical methods do not require chemical reagents, are environmentally benign, and preserve the native chemical structure of cellulose. However, they are generally associated with high energy consumption, low processing efficiency, and a tendency to induce fiber shortening or breakage during prolonged treatment.

2.1.2 Chemical Methods

Chemical methods involve the selective disruption of amorphous regions in cellulose via acid hydrolysis, TEMPO-mediated oxidation, hydrogen peroxide oxidation, or sulfuric/hydrochloric acid treatment, thereby releasing nanoscale cellulose entities. These methods offer advantages such as controllable fiber dimensions, high crystallinity, and rapid reaction kinetics. Nevertheless, their drawbacks include the generation of large volumes of chemical waste, harsh reaction conditions, and considerable environmental burdens.

Notably, Li *et al.*[30] emphasized that CNFs prepared via TEMPO oxidation possess abundant surface carboxyl groups, which significantly enhance ionic crosslinking interactions with sodium alginate (SA), leading to improved mechanical properties of the resulting hydrogels.

Figure. 2 presents the preparation and morphological structures of cellulose nanofibrils and cellulose nanocrystals, providing a visual comparison that clarifies how these distinct forms arise from different treatment routes.

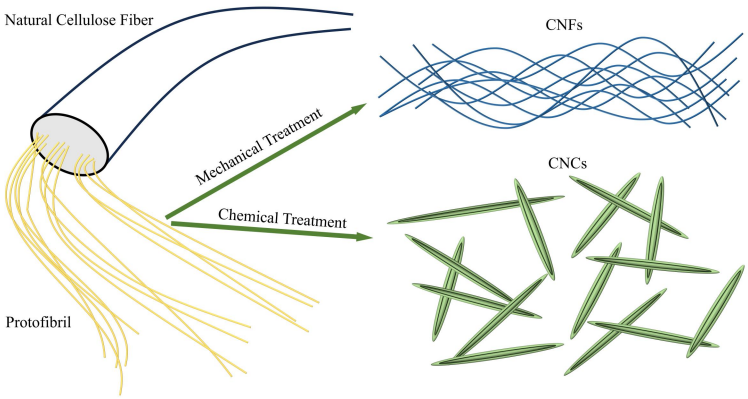

Figure. 2 Preparation and morphological structure of cellulose nanofibrils, cellulose nanocrystals. CNFs are obtained by mechanical treatment of cellulose and have a fiber entangled network structure. CNCs are obtained by chemical treatment of cellulose, which is in the form of short whiskers. Produced by Microsoft PowerPoint version 16.0.20026.20182. **NOTE:** CNFs: cellulose nanofibrils; CNCs: cellulose nanocrystals.

2.1.3 Combined Chemical-Mechanical Methods

In combined chemical-mechanical approaches, cellulose is first subjected to chemical pretreatment — such as sodium chlorite acidification or TEMPO oxidation — to weaken interfibrillar hydrogen bonding, followed by mechanical disintegration to obtain nanofibers. Compared with purely mechanical or chemical methods, this strategy substantially reduces energy consumption and preserves fiber integrity. However, the overall process is more complex, and the chemical pretreatment stage still poses environmental concerns. At present, this approach is regarded as one of the most widely adopted technical routes in laboratory-scale and pilot-scale studies.

2.1.4 Green Solvent-Based Methods

Green solvent-based methods employ deep eutectic solvents (DESs) or ionic liquids (ILs) as alternatives to conventional chemical solvents for cellulose processing. Li *et al.*[30] successfully extracted CNFs from okara using DES combined with high-shear processing, achieving high-purity nanocellulose with low energy input. These solvents are generally biodegradable, low-toxicity, and recyclable, rendering the overall process environmentally friendly. However, their high cost and the lack of mature industrial-scale systems currently limit large-scale implementation.

Based on the preparation principles and morphological characteristics discussed above, **Table 1** summarizes the main preparation methods and compares their advantages and limitations.

Table 1. Comparison of main preparation methods for nanocellulose hydrogels

Method	Representative Process	Advantages	Disadvantages	References
Mechanical Methods	High-pressure homogenization, grinding	Environmentally friendly, retain the native fiber structure	High energy consumption risk of fiber fracture	[28]
Chemical Methods	Acidolysis, TEMPO oxidation	Controllable size, rapid reaction	Chemical waste generation; environmental burden	[30]
Combined Chemical-Mechanical Methods	TEMPO and high shear	Low energy consumption, excellent performance	Complex process	[27]
Green Solvent-Based Methods	DESs, ILs	Environmentally friendly, renewable	High cost, difficult for large-scale application	[30]

NOTE: DESs: deep eutectic solvents; ILs: ionic liquids.

2.2 Construction Strategies of Nanocellulose-Based Hydrogels

2.2.1 Physical Crosslinking

Physical crosslinking refers to the construction of nanocellulose-based hydrogels through reversible physical interactions, including physical entanglement of nanocellulose chains, van der Waals forces, hydrogen bonding, and ionic interactions. As no chemical crosslinkers are required, hydrogels prepared via physical methods generally exhibit good biosafety and biocompatibility. In addition, these methods are simple, non-toxic, and reversible. However, the resulting hydrogels often suffer from limited mechanical strength and poor long-term structural stability.

2.2.1.1 Hydrogen-Bond Crosslinking

Hydrogen bonding is the most prevalent physical crosslinking mechanism in nanocellulose-based hydrogels. Abundant hydroxyl groups on the surfaces of CNFs or CNCs form multiple hydrogen bonds with polymeric matrices such as sodium alginate (SA), polyvinyl alcohol (PVA), and gelatin, thereby stabilizing the hydrogel network. Li *et al.*[30] fabricated CNF/SA composite hydrogels using a deep eutectic solvent (DES) approach and demonstrated that a continuous nanoscale network was formed through extensive hydrogen bonding. This network significantly enhanced the compressive modulus and swelling stability of the hydrogel. Scanning electron microscopy (SEM) observations revealed that increasing CNF content led to reduced pore size and improved structural uniformity, thereby optimizing network compactness and crystallinity.

2.2.1.2 Ionic Coordination Crosslinking

Ionic coordination crosslinking typically occurs between multicarboxyl polymers, such as SA, and multivalent cations (e.g., Ca^{2+}, Zn^{2+}, Fe^{3+}), forming the characteristic “egg-box” structure. This type of crosslinking is reversible and enables controlled

dissociation and reassembly in response to external stimuli such as pH or ionic strength variations.

For instance, in Ca^{2+}–SA systems, Ca^{2+} ions coordinate with carboxyl groups in the G-block regions to generate a three-dimensional network. The incorporation of CNFs enhances hydrogen-bond density and increases the distribution of interfacial coordination sites, thereby improving both the stability and flexibility of the hydrogel network[30].

2.2.2 Chemical Crosslinking

Chemical crosslinking involves the formation of covalent bonds either between nanocellulose chains or between nanocellulose and other polymeric components to construct three-dimensional networks, typically through photo-initiated polymerization or reactions with crosslinking agents such as glutaraldehyde (GA) or ethylene glycol diglycidyl ether (EGDE). Hydrogels prepared via chemical crosslinking generally exhibit superior mechanical strength and structural stability. However, some chemical crosslinkers may introduce biotoxicity, and the resulting networks are usually irreversible.

2.2.2.1 Covalent Bond Crosslinking and Network Stability

Kalu *et al.*[31] employed $DMol^3$ calculations combined with molecular dynamics (MD) simulations to elucidate the chemical crosslinking mechanisms in PVA/cellulose composite hydrogels. Their results revealed that the binding energy between PVA-OH and cellulose-OH groups strongly depends on water content. At a water content of 40%, the cohesive energy density (CED) reached its maximum, indicating the formation of a dense and energetically stable covalent network. Moreover, the study demonstrated that elevated temperatures or excessive solvation increased the intermolecular spacing between covalent bonds and reduced electron density, thereby weakening the overall rigidity of the network.

2.2.2.2 Dynamic Covalent and Photochemical Crosslinking

In recent years, dynamic covalent bonds (e.g., Schiff base bonds, –C=N–) and photochemical crosslinking have emerged as effective strategies for achieving controllable chemical networks. By introducing reversible covalent reactions (such as aldehyde-amine condensation or borate ester exchange) or photosensitive moieties (e.g., methacrylate (MA), acrylamide (AAm)), hydrogels can maintain high mechanical strength while enabling localized self-healing and photo-responsive regulation. This "chemical-physical synergy" has become a key trend in the development of high-performance intelligent hydrogels.

2.3 Network Architecture Design

Conventional hydrogels exhibit limited mechanical properties, which often fail to meet the requirements of practical applications. Therefore, designing diverse network architectures to significantly enhance strength, toughness, self-healing capability, and stability has become a critical strategy. DN hydrogels, composed of a rigid primary network and a flexible secondary network, employ a "soft-encases-hard" structure to preferentially dissipate energy under stress, thereby improving fracture elongation and fatigue resistance. For instance, the hydrogel exhibits a compressive strength of up to 363 kPa upon incorporation of microfibrillated cellulose (MFC)[28]. Interpenetrating polymer networks (IPN/SIPN) achieve synergistic reinforcement through the interpenetration of distinct polymer networks, while maintaining flexibility and imparting self-healing capability. Such structures have been widely applied in self-healing materials, drug hydrogels, and its nanoscale interpenetrating structure promotes uniform water diffusion and mitigates phase separation[32]. Nanocomposite networks, formed by incorporating micro- or nanofibers (e.g., MFC, or CNF) to create high-aspect-ratio frameworks, leverage hydrogen bonding and physical entanglements to reinforce local and overall structural stability, improving creep resistance and durability. Mechanical properties can further be optimized by regulating the proportion of nanofillers; for example, in CNF/SA systems, increasing Ca^{2+} concentration reduces pore size from approximately 200 μm to 60 μm while tripling the compressive modulus[30]. Overall, these three network types offer distinct advantages: DN networks emphasize macroscopic toughness and energy dissipation, IPN/SIPN networks balance flexibility and self-healing, and nanocomposite networks enhance microstructural stability and durability. However, each has limitations: DN hydrogels exhibit limited self-healing, IPNs are complex to fabricate, and nanocomposite networks require homogeneous dispersion, with excessive filler potentially reducing toughness. Rational combination and design of these strategies can enable synergistic optimization of hydrogel mechanical performance and functionality. To better illustrate how polymer networks are interwoven in such systems, **Figure. 3** presents a representative semi-interpenetrating polymer network structure.

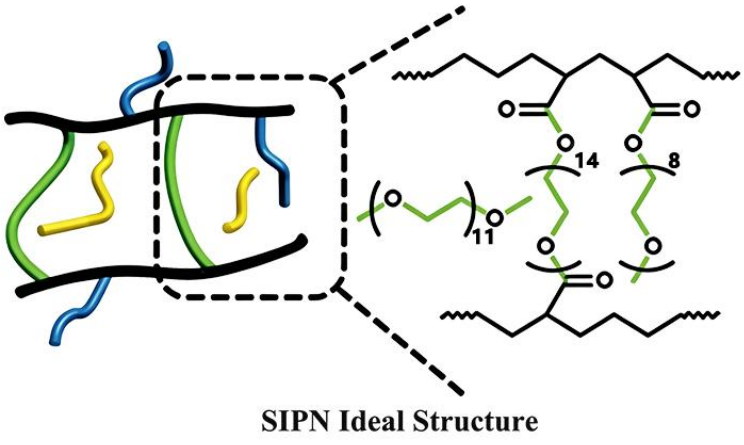

Figure. 3 Functional semi-interpenetrating polymer networks. Reproduced with permission. Copyright 2026, John Wiley and Sons[32]. **NOTE**: SIPN: semi-interpenetrating polymer networks; Black lines: cross-linked polyethylene oxide (PEO) backbone; Green lines: poly (ethylene glycol) diacrylate (PEGDA); Blue lines: poly (ethylene glycol) methyl ether acrylate (PEGMEA); Yellow lines: poly (ethylene glycol) dimethyl ether (PEGDME).

2.4 Functionalization and Performance Optimization

In recent years, significant breakthroughs have been achieved in nanocellulose-based hydrogels through molecular structure regulation and interfacial chemical modification, enabling multidimensional performance enhancement, including mechanical reinforcement, stimuli responsiveness, electrical conductivity, sensing capability, self-healing, and adhesion. This section systematically discusses various functionalization strategies, their underlying performance optimization mechanisms, and the synergies and trade-offs among different functional designs. The main functionalization pathways are schematically summarized in the **Figure. 4**.

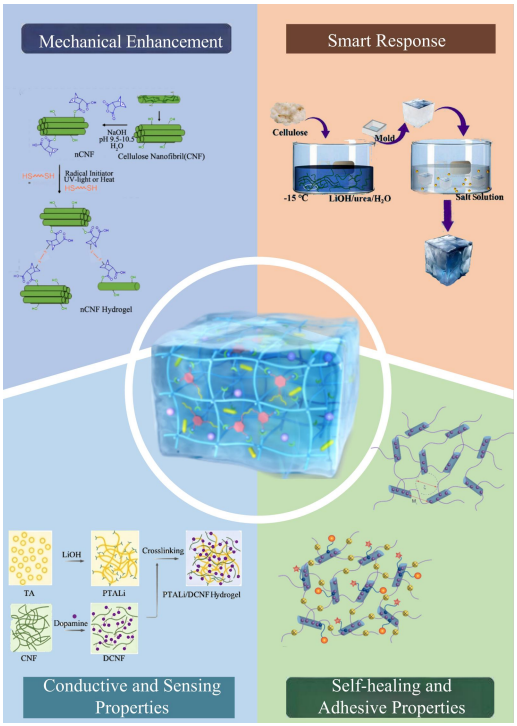

Figure. 4 Functionalization mechanism of nanocellulose hydrogels. Produced by Adobe Photoshop 2024 version 25.9.1.626. Reproduced with permission[2,7,33,34,35]. Copyright 2025, Elsevier[2]. Copyright 2019, Elsevier[7]. Copyright 2024, Elsevier[33]. Copyright 2025, Elsevier[34]. Copyright 2021, Springer Nature[35]. **NOTE:** TA: thioctic acid; PTALi: lithiated poly (thioctic acid); DCNF: dopamine-grafted cellulose nanofibers.

2.4.1 Mechanisms of Mechanical Enhancement

Dual-crosslinked networks, such as ionic-covalent entanglements (ICE), combine a rigid nanocellulose scaffold with a soft polymer matrix (e.g., GelMA/SA) to distribute stress effectively and suppress crack propagation. Incorporation of 0.70wt% MFC enhanced Young's modulus by about 72%, highlighting efficient macroscopic and microscopic reinforcement pathways. Dynamic hydrogen bonds between cellulose and polymer chains continuously rupture and reform under stress, dissipating energy and improving deformation resistance. This mechanism is pivotal for high toughness and fatigue resistance[28].

2.4.2 Stimuli-Responsive Properties

Hydrogels functionalized via chemical modifications or polymer composites respond to pH, temperature, light, and ionic conditions, enabling controlled drug release, adaptive swelling, or ion-responsive performance. For example, aminated CNF hydrogels regulate nutrient release under varying soil pH, while photo-crosslinked GelMA/CNF systems allow precise network density tuning[31,36].

2.4.3 Electrical Conductivity and Sensing Performance

Incorporating graphene (GN), carbon nanotubes (CNTs), or conductive polymers (e.g., polyaniline (PANI)) into cellulose hydrogels is a primary strategy for achieving high electrical conductivity. Zheng *et al.*[37] prepared a TOCNF-supported graphene hydrogel exhibiting high stretchability (850%), electrical conductivity (2.5 $S \cdot m^{-1}$), and self-healing efficiency (96.70%). The surface carboxyl groups of TOCNFs ensured uniform dispersion and strong interfacial interactions, preserving the continuity of graphene's electronic pathways. Jiao *et al.*[38] further introduced PANI into a TOCNF/polyacrylic acid (PAA) system to fabricate a conductive hydrogel sensor with a conductivity of 3.95 $S \cdot m^{-1}$ and a gauge factor (GF) of 8.0. The high sensitivity originates from synergistic regulation of electrical and mechanical pathways within the multiscale network: TOCNFs provide continuous conductive frameworks, PANI facilitates electron transport, and dynamic hydrogen bonding and metal coordination impart structural adaptability.

2.4.4 Self-Healing and Adhesive Properties

The self-healing capability of hydrogels primarily relies on reversible interactions such as hydrogen bonding, metal-ion coordination, and dynamic covalent bonding. Studies have shown that Fe^{3+}-carboxyl coordination enables up to 98% recovery of mechanical and electrical properties at room temperature[39]. The abundant carboxyl groups on TOCNFs act as multivalent coordination sites, promoting rapid network reconstruction after damage and imparting excellent durability and repeatability. The polar hydroxyl groups and intrinsic surface roughness of cellulose enhance interfacial adhesion between hydrogels and biological tissues or inorganic substrates. Li *et al.*[30] reported that CNF/SA composite hydrogels exhibited favorable bioadhesion and tissue compatibility due to the synergistic effects of hydrogen bonding

and Ca^{2+}-mediated ionic crosslinking.

2.5 Diversified Application Exploration

Owing to their outstanding mechanical properties, biocompatibility, reparability, and environmental sustainability, nanocellulose-based hydrogels have become focal materials in multidisciplinary research. Their application scope has expanded from conventional pharmaceutical carriers to flexible electronics, energy devices, and green manufacturing, forming a multifunctional development landscape[28,40].

2.5.1 Biomedical Engineering Applications

2.5.1.1 Drug Delivery and Controlled Release Systems

Nanocellulose-based hydrogels exhibit excellent biocompatibility and biodegradability, while surface hydroxyl groups can be functionalized via carboxylation or amination to enable selective adsorption and sustained release of drugs or bioactive molecules[27]. By regulating crosslinking density and surface functional groups, drug release rates can be precisely controlled under different pH environments, enabling targeted and sustained delivery.

2.5.1.2 Tissue Engineering Scaffolds

Composite hydrogels based on nanocellulose, sodium alginate, and gelatin have demonstrated excellent cell proliferation and biocompatibility in cartilage and skin tissue engineering. The MFC-reinforced ICE hydrogels developed by Li *et al.*[28] exhibit high strength and extensibility, with a Young's modulus of 363.4 kPa, well-matched to the mechanical properties of soft tissues. The porous structure and high specific surface area of nanocellulose provide an ideal microenvironment for cell adhesion, migration, and nutrient transport[40].

2.5.1.3 Wound Dressings and Antibacterial Materials

Nanocellulose-based hydrogels possess high water absorption and breathability, maintaining a moist environment conducive to wound healing. They effectively absorb exudates, reduce inflammatory responses, and can be functionalized with silver ions or antibiotics to impart antibacterial activity. CNF/SA composite hydrogels, enabled by Ca^{2+}-mediated crosslinking, exhibit excellent tissue adhesion and plasticity, making them promising candidates for wound dressing applications[30].

Nanocellulose hydrogels for biomedical applications have evolved from simple drug-delivery carriers into multifunctional platforms that integrate mechanical support, bioactive molecule delivery, antibacterial activity, and tissue-compatible microenvironments. Nevertheless, their clinical translation remains limited by several unresolved issues, including batch-to-batch reproducibility, sterilization stability, degradation behavior, immune responses, and regulatory requirements. Future studies should move beyond short-term cytocompatibility tests and release profiles, and should place greater emphasis on standardized biosafety evaluation, in vivo degradation mechanisms, and long-term performance under clinically relevant conditions.

2.5.2 Flexible Electronics and Intelligent Devices

2.5.2.1 Wearable Sensors and Electronic Skin

Conductive nanocellulose hydrogels form flexible electrical networks through integration with graphene, CNTs, or conductive polymers such as PANI, combining conductivity, stretchability, and self-healing. These integrated properties enable nanocellulose hydrogels to serve in diverse application scenarios. A summary of the major application fields is provided in the **Figure. 5**.

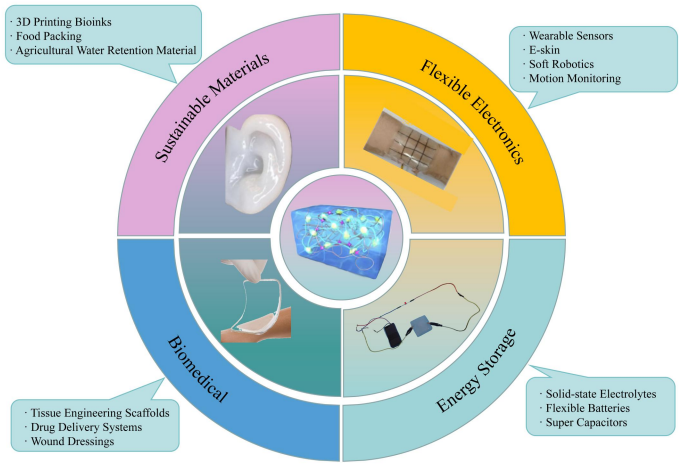

Figure. 5 Application fields of nanocellulose hydrogels. Produced by Adobe Photoshop 2024 version 25.9.1.626. Reproduced with permission[7,41,42]. Copyright 2019, Elsevier[7]. Copyright 2024, Elsevier[41]. Copyright 2021, Elsevier[42].

Hydrogels based on PAA/TOCNF/CNTs exhibit high viscoelasticity and strain responsiveness while maintaining stable electrical performance under repeated deformation, making them suitable for soft robotic actuators and flexible actuation units[39]. The TOCNF–CNTs/PAA-0.7 composite hydrogel achieved an elongation at break of 850% and a self-healing efficiency of 99%, enabling repeated monitoring of complex joint motions. Recently, CNF/Fe^{3+}-mediated dynamic supramolecular hydrogels have achieved the unification of strong adhesion and light-triggered detachability, and can be used for self-powered electronic skin and wireless human motion monitoring[43]. Furthermore, PVA/CNF supramolecular networks can exhibit orientation response during stretching, thereby achieving ultra-high stretchability, fatigue resistance, and crack propagation resistance, providing a structural foundation for long-term wearable sensing[44].

2.5.2.2 Energy Storage and Electrolyte Materials

In energy storage applications, the development of safe and high-performance solid or quasi-solid electrolytes is critical for flexible electronics and next-generation energy devices. Through

molecular modification and composite strategies, multidimensional regulation of ion transport, mechanical integrity, and interfacial stability can be achieved. Yuan *et al*.[45] demonstrated selective separation of Li^+ and Fe^{3+} ions via amino acid-functionalized systems. Li *et al*.[16] significantly enhanced ionic mobility and water retention in PVA-based hydrogel electrolytes reinforced with carboxyl modified nanocellulose (C-CNF), achieving an optimized ionic conductivity of 3.12×10^{-1} S·cm^{-1}. As rigid nanoframeworks, nanocellulose structures markedly improve mechanical strength, ensuring stable operation of flexible devices under deformation[46]. Moreover, Xu *et al*.[47] reduced free water content via hydrogen-bond regulation to fabricate PVA/nanocellulose–H_2SO_4 hydrogel electrolytes with an ultralow freezing point of -77.35 ℃, greatly extending the low-temperature operational limit of energy storage devices.

Flexible electronics require high conductivity, stretchability, self-healing ability, and environmental stability — all under repeated bending, stretching, or deformation. Adding conductive fillers such as graphene, CNTs, MXene, or conductive polymers can substantially improve electrical performance. But there are trade-offs: higher cost, possible biocompatibility concerns, or compromised long-term stability. One practical way forward is to use nanocellulose to guide interfacial assembly, building stable conductive pathways. At the same time, efforts are needed to reduce filler aggregation and improve resistance to dehydration, freezing, and fatigue over months of use.

2.5.3 Sustainability and Green Applications

2.5.3.1 3D Printing and Processable Bioinks

Nanocellulose hydrogels exhibit shear-thinning behavior and high viscoelasticity, making them ideal matrices for 3D-printable bioinks. Printed structures rapidly achieve self-supporting integrity, enabling personalized tissue fabrication and biodegradable template printing. Auvinen *et al*.[48] employed 3D printing to precisely regulate drug release by fabricating inert polylactic acid (PLA) capsules subsequently filled with drug-loaded CNF hydrogels. By varying internal geometry and CNF content, drug release profiles were successfully tuned.

2.5.3.2 Food Packaging and Preservation Materials

Yu *et al*.[49] reinforced PVA/SA composite hydrogels with soybean hull-derived nanocellulose for refrigerated pork packaging, extending shelf life to 10 days. The enhancement mechanism involves improved fatigue resistance and gas barrier properties imparted by CNFs, which suppress moisture loss and oxidative reactions while preserving meat color and freshness.

2.5.3.3 Agricultural Water Retention and Nutrient Release Systems

The pH-responsive aminated nanocellulose hydrogel (A-CNF/PAM-PAEM) developed by Shaghaleh *et al*.[36] significantly increased soil water retention (+15.70%) and nitrogen use efficiency (+38.60%) under neutral irrigation conditions. Its biodegradability renders it a promising carrier for future water-saving fertilizers and environmentally friendly soil conditioners.

To provide a clearer overview of practical implementations, **Table 2** summarizes several representative nanocellulose hydrogel systems, highlighting their preparation methods, functionalization strategies, core performance parameters, and application fields.

Table 2. Representative nanocellulose-based hydrogel systems and their key properties

Representative Systems	Preparation/ Functionalization Method	Core Performance Parameters	Main Applications Key Advantages/ Limitations
Nanocellulose-Based Conductive Hydrogels[2]	Crosslinking and composite of CNF/CNC/BC with conductive fillers; Surface carboxyl/hydroxyl group modification, forming a conductive network	Elongation at break: 100%–600% Conductivity: 10^{-3}–10^{-1} S·cm^{-1}	Flexible sensors and bioelectronics; High conductivity and self-healing; Limited by cost and stability
PDA/TOCNF Physically Crosslinked Hydrogels[24]	TEMPO-oxidized CNF combined with PDA is crosslinked via Ca^{2+} ions to form a physical network; PDA is endowed with pH/NIR response and drug carrier functions	Compressive modulus: ≈ 40 kPa Good flexibility; NIR light-responsive drug release	Drug controlled release and wound healing; High biocompatibility and simple preparation; Relatively low mechanical strength
TOCN/PNIPAM-PAM Bilayer Hydrogel Actuator[50]	One-step preparation of TOCN/PNIPAM and TOCN/PAM bilayer structures; Fe^{3+} ions endow shape memory and conductivity	Compressive strength: ≈ 89 kPa Elongation at break: ≈ 171% Shape memory response	Soft robotics and flexible sensors; Excellent mechanical properties and fast response; Sensitive to humidity and complex preparation
Nanocellulose Biomedical Hydrogels[51]	TEMPO oxidation or acid hydrolysis for CNC/CNF production; Grafting peptide, protein, or drug molecules to achieve bio-recognition	Storage modulus: 50 kPa–500 kPa Water content: > 95%	Tissue engineering and drug delivery; Controllable pore structure and high biosafety; Difficulty in precisely controlling the degradation rate
TOCNF/TA@MXene Composite Hydrogel[52]	TA is coated on MXene and TOCNF is introduced as a support to form a 3D network; The catechol groups of TA form hydrogen bonds with MXene, enhancing antioxidant properties	Elongation: > 800% Output voltage: 106 V Current: ≈ 2 μA	Self-powered wearable TENG and health monitoring; Ultra-high stretchability and electrical output; MXene oxidation resistance remains challenging
Nanocellulose Biomedical Hydrogels[51]	TEMPO oxidation or acid hydrolysis for CNC/CNF production; Grafting peptide, protein, or drug molecules to achieve bio-recognition	Storage modulus: 50 kPa–500 kPa Water content: > 95%	Tissue engineering and drug delivery; Controllable pore structure and high biosafety; Difficulty in precisely controlling the degradation rate

Table 2. Representative nanocellulose-based hydrogel systems and their key properties (continued)

Representative Systems	Preparation/ Functionalization Method	Core Performance Parameters	Main Applications Key Advantages/ Limitations
TOCNF/TA@MXene Composite Hydrogel[52]	TA is coated on MXene and TOCNF is introduced as a support to form a 3D network; The catechol groups of TA form hydrogen bonds with MXene, enhancing antioxidant properties	Elongation: > 800% Output voltage: 106 V Current: ≈ 2 μA	Self-powered wearable TENG and health monitoring; Ultra-high stretchability and electrical output; MXene oxidation resistance remains challenging
BP@PDA-TOCNs/PAM Conductive Hydrogel[53]	Free radical polymerization to introduce BP@PDA and TOCNs in one step; PDA prevents BP oxidation; BP provides photoresponse; TOCN provides reinforcement	GF = 6.0; S (pressure sensing capability) = 0.13 kPa^{-1}; Excellent photothermal response	Photo-responsive sensors and smart wearable devices; Excellent photothermal/ conductive/mechanical performance; BP long-term stability insufficient
Nanocellulose-Based Drug Delivery Hydrogels[12]	CNC, CNF, and BC form 3D networks through physical/chemical cross-linking; Surface modification (amination or carboxylation)	Release time from minutes to months; Tunable mechanical modulus and porosity	Drug delivery; Sustainable release and high biocompatibility; Drug loading amount and degradation rate need optimization
BNC/Cationic Starch Composite Hydrogel[54]	BNC is mixed with cationic starch to form a physically cross-linked network, Starch ratio controls rheology and transparency	High transparency; Good strength; Low hygroscopicity; 3D printable	Sustainable materials and biodegradable packaging films; Renewable, eco-friendly and suitable for 3D printing; Relatively weak waterproof performance
MWCNTs/CNWs/PAM/SA Dual-Network Conductive Hydrogel[55]	Constructing a PAM/SA double network via free radical polymerization and reinforced by MWCNTs and CNWs; CNWs form hydrogen bond network for mechanical enhancement; MWCNTs improve conductivity	GF = 5.65; Strain > 100%; Voltage output: 28 V	Flexible tactile sensors and self-powered generators; High conductivity and sensitivity; Affected by humidity

NOTE: CNF: cellulose nanofibril; CNC: cellulose nanocrystal; NIR: near-infrared; BNC: bacterial nanocellulose; PAM: polydopamine; SA: sodium alginate; GF: gauge factor; TA: thioctic acid; PDA: polydopamine; BP: black phosphorus; TOCNF: TEMPO-oxidized cellulose nanofibrils; MWCNTs: multi-walled carbon nanotubes; BC: bacterial cellulose; CNWs: cellulose nanowhiskers; TENG: triboelectric nanogenerator; TOCN: TEMPO-oxidized cellulose nanofibers; PNIPAM: poly (N-isopropylacrylamide).

It should be noted that, although **Table 2** summarizes several representative nanocellulose hydrogel systems reported in different studies, these investigations were conducted independently and under diverse experimental conditions. Moreover, the field of advanced nanocellulose hydrogel research still lacks universally accepted international standards for characterization and performance evaluation. Consequently, the key performance parameters listed for different hydrogel systems do not possess strict cross-comparability. Instead, these parameters should be interpreted primarily as indicators of the advantages of a given hydrogel system relative to conventional materials employed in the same application domain. For example, the TOCN/PNIPAM–PAM bilayer hydrogel actuator is a hydrogel material designed for flexible sensing applications, with particular emphasis placed on its mechanical responsiveness. Accordingly, its key performance metrics highlight superior compressive strength and elongation at break compared with conventional rubber-based polymer materials commonly used in similar applications, thereby demonstrating its excellent flexibility. However, these mechanical parameters cannot be directly compared with the compressive modulus reported for the PDA/TOCNF physically crosslinked hydrogel system. Although the latter parameter likewise reflects the flexibility-related characteristics of the hydrogel, the PDA/TOCNF physically crosslinked hydrogel is primarily intended for biomedical applications such as controlled drug delivery and wound healing. Significant differences in raw material selection, fabrication strategies, and characterization objectives between these two hydrogel systems inevitably lead to distortions in any direct horizontal comparison of their performance metrics. In contrast, the key performance data of BP@PDA–TOCNs/PAM conductive hydrogel and MWCNTs/CNWs/PAM/SA double-network conductive hydrogel exhibit a relatively high degree of comparability. Both systems are prepared via free-radical polymerization, both prioritize enhanced electrical conductivity and high gauge factor values, and both face challenges associated with maintaining conductivity stability under complex environmental conditions. These similarities reflect common limitations currently constraining the development of conductive hydrogel systems. Overall, representative nanocellulose hydrogel systems reported in recent years can generally be categorized into several major classes, including biomedical hydrogels, conductive hydrogels for sensing and electronic device applications, and structural hydrogels for packaging and coating applications. A shared characteristic among these systems is that they offer significant functional advantages over conventional materials within their respective application fields. Nevertheless, insufficient stability remains a widespread challenge, and most systems exhibit pronounced environmental sensitivity. Therefore, further research efforts are urgently required to optimize their long-term stability and practical applicability.

In sustainability-oriented applications, nanocellulose hydrogels offer intrinsic advantages, including renewability, biodegradability,

and tunable water-retention capacity. However, their practical use in food packaging, agriculture, and environmental remediation requires a systematic assessment of production cost, degradation products, scalability, and life-cycle environmental impacts. Future research should therefore combine material performance optimization with life-cycle assessment and techno-economic analysis to determine whether nanocellulose hydrogels provide genuine sustainability advantages over conventional petroleum-derived materials.

3. Challenges and Controversies

3.1 Compatibility Balance between Mechanical Properties and Functional Characteristics

In the material design of cellulose-based hydrogels, achieving a synergistic optimization between mechanical performance and specific functional properties represents a central challenge for their advanced applications. This issue of compatibility balance fundamentally originates from the potential synergy or competition among the structural bases underlying different performance attributes. In general, the favorable mechanical properties of cellulose hydrogels are mainly governed by the intrinsic characteristics of cellulose — such as high crystallinity, high Young's modulus, and large aspect ratio — as well as by strong physical interactions (e.g., hydrogen bonding and van der Waals forces) and chemical interactions (e.g., covalent bonding between polymer chains) within the hydrogel network. Moreover, in emerging cellulose hydrogel designs, optimization of the overall polymer network architecture also plays a critical role.

In contrast, the origins of functional properties in cellulose hydrogels vary depending on their specific types. The first category comprises mechanical performance extensions, such as high stretchability, high flexibility, and shape memory behavior. These properties typically arise from the unique polymer network structures of cellulose hydrogels. The second category includes "specialized functionalities" that enable applications in diverse fields, such as electrical conductivity, adsorption capacity, and thermal conductivity. These functionalities are generally derived from specific doping strategies, dopant components, or chemical modification routes, and are less directly related to the intrinsic network structure of the cellulose hydrogel itself. The third category involves environmentally friendly characteristics, such as biocompatibility and biodegradability, which primarily originate from cellulose itself as a bio-based material with inherent advantages.

Based on the above considerations, it can be concluded that when the mechanical properties and functional characteristics of cellulose hydrogels share similar structural origins, a favorable compatibility balance can often be achieved. Conversely, when their underlying mechanisms are contradictory, trade-offs between mechanical performance and functionality become unavoidable. Accordingly, representative research progress corresponding to these three categories of functional characteristics is summarized below.

First category (mechanical-performance-derived functionalities): Rafi Ullah, Luqman Ali Shah, and co-workers[56] incorporated CNCs, rich in active hydroxyl groups, into hydrogel networks. The hydroxyl groups of CNCs formed extensive hydrogen bonding interactions with other monomers, thereby enhancing multiple mechanical properties of the hydrogel, including elasticity and toughness, and realizing effective performance synergy.

Second category (specialized functional properties): Jingquan Han, Ding, *et al*.[57] combined the high electrical conductivity of polypyrrole (PPy) with the excellent mechanical performance and high modifiability of CNFs. By dispersing pyrrole monomers onto flexible CNF chains, they obtained nanocomposites that simultaneously exhibited mechanical robustness and electrical conductivity. Subsequent incorporation of these nanocomposites into hydrogel matrices enabled compatibility between mechanical strength and conductive functionality in cellulose-based hydrogels.

Third category (environmentally friendly functionalities): Yang *et al*.[40,58] fabricated a novel pH-responsive hydrogel (RPC/PB) for the treatment of bacterial-infected wounds using a PVA–borax system and resveratrol-grafted CNFs (RSV-CNF), a natural antibiotic. The resulting hydrogel exhibited excellent mechanical strength (fracture strength of 149.60 kPa), strong adhesion (tissue shear stress of 54.20 kPa), and pH-responsive drug release behavior, along with outstanding antibacterial activity, skin tissue regeneration, and wound closure performance. The synergy between biocompatibility and mechanical performance was attributed to the natural origin of the components and their ability to reinforce the existing hydrogel network.

3.2 Comparison of Preparation Methods in Terms of Cost, Environmental Impact, and Scalability

The preparation of cellulose-based hydrogels can generally be divided into three major stages[26,59,60]. The first stage involves the production of cellulose nanofibers, including chemical, mechanical, and enzymatic methods, which primarily determine the aspect ratio and surface activity of the resulting cellulose materials. The second stage focuses on the modification of cellulose nanofibers to generate various derivatives that satisfy the functional requirements of cellulose hydrogels. The third stage is the construction of the three-dimensional hydrogel network, including chemical crosslinking, physical crosslinking, and polymerization-induced crosslinking. The methods employed in this stage influence the interactions within the hydrogel network and ultimately determine hydrogel performance. Both the first and third stages can be regarded as key components of cellulose hydrogel preparation strategies.

Regarding cellulose nanofiber preparation techniques, chemical methods often rely on costly chemical reagents, such as oxidants in TEMPO-mediated oxidation or strong acids in acid hydrolysis, resulting in high equipment costs and environmental risks. Nevertheless, due to their long development history, these methods remain the most mature and widely used technologies. Physical methods avoid significant environmental hazards and require relatively simple equipment and processes; however, they are energy-intensive and typically yield cellulose materials with limited performance advantages. Consequently, physical methods are often combined with chemical treatments to enhance cellulose properties. Enzymatic methods present the lowest environmental risk but suffer from extremely high enzyme costs and immature industrial processes, with scalability still under investigation.

With respect to crosslinking technologies, physical crosslinking methods — such as freeze-thaw-induced, photo-induced, and radiation-induced crosslinking — mainly rely on non-covalent interactions (e.g., hydrogen bonding and van der Waals forces) to maintain hydrogel structures. These approaches avoid complex crosslinkers and sophisticated instrumentation, offering advantages in terms of cost and environmental friendliness. However, the resulting hydrogels generally exhibit limited mechanical strength and stability and are more sensitive to environmental changes.

Chemical crosslinking employs crosslinking agents (e.g., citric acid, epichlorohydrin, glutaraldehyde) to induce three-dimensional covalent networks, producing hydrogels with superior mechanical properties and functional performance. Nonetheless, residual crosslinkers may compromise biocompatibility and pose environmental risks. Polymerization-induced crosslinking, achieved via bulk polymerization, solution copolymerization, or radiation polymerization, yields hydrogels with covalently polymerized networks and often complex structures, providing outstanding mechanical and functional performance at the expense of higher costs and potential environmental risks associated with toxic monomers.

3.3 Achieving Uniform Dispersion of Nanofillers in Multicomponent Composite Systems

In current research, dispersing cellulose nanofibers, nanoparticles, or their modified derivatives into hydrogel polymer matrices has become a common strategy for fabricating high-performance cellulose-based hydrogels. This approach requires nanofillers to be uniformly dispersed and stably maintained within the matrix to ensure homogeneous and stable macroscopic properties. A widely adopted design strategy involves exploiting the surface activity of cellulose nanoparticles to enable physical adsorption or chemical grafting of polymer chains onto particle surfaces, thereby forming three-dimensional physical crosslinking networks. This strategy necessitates sufficient surface activity of cellulose nanofillers, effective dispersion techniques, and feasible methods to promote intercomponent interactions for network formation.

Zhou *et al.*[61,62] prepared cellulose nanocrystal (CNC)/polyacrylamide composite gels and demonstrated that CNCs not only served as reinforcing agents but also acted as physical crosslinking points, accelerating gelation. At a CNC concentration of 6.70%, the gel exhibited optimal compressive performance, with compressive strength at 85.20% strain exceeding that of pure polyacrylamide by more than 2.5 times.

Shao[63] anchored tannic acid onto nanocellulose surfaces via surface deposition, using it as a dynamic bridging agent. Through combined covalent and ionic crosslinking, tannic-acid-coated nanocellulose (TA@CNC) ionic gels with excellent mechanical performance, self-healing capability, and self-adhesion were obtained. This approach was further extended to fabricate various tannic-acid-modified nanocellulose-reinforced hydrogels, achieving effective dispersion and interfacial interactions of nanofillers.

Zheng *et al.*[64] dispersed TEMPO-oxidized nanocellulose-graphene conductive nanocomposites into acrylic acid-acrylamide hydrogel matrices, producing cellulose hydrogels with both excellent mechanical and electrical performance. The dispersion and integration of the nanoreinforcement relied on abundant hydrogen bonding interactions as an initial driving force, followed by the formation of ionic coordination bonds via active functional groups to construct dense crosslinked networks.

Wu *et al.*[65] developed cellulose-based hydrogels incorporating liquid metal (LM) as a multifunctional filler. To stabilize and uniformly disperse LM droplets within the hydrogel matrix, cationic nanocellulose rich in quaternary ammonium groups was employed to encapsulate the droplets. This design enabled strong intercomponent interactions, resulting in a double-network structure with combined electrical conductivity and self-healing capability.

Overall, strategies for promoting initial nanofiller dispersion include physical methods (e.g., ultrasonication and shear mixing) and solvent-based approaches (e.g., controlled solvent evaporation and solvent system optimization). Surface activation methods, such as surface grafting and chemical oxidation, enhance nanofiller reactivity, while physical and chemical crosslinking techniques facilitate network formation through intercomponent interactions. Together, these approaches constitute essential tools for constructing multicomponent composite hydrogel systems.

3.4 Enhancement of Mechanical Properties

Among the various strategies proposed for improving the mechanical performance of nanocellulose-based hydrogels — such as nanocomposite reinforcement, IPNs, DN hydrogels, and topological hydrogel design — which approaches exhibit the greatest potential for industrial implementation? What are the fundamental scientific bottlenecks limiting their further development? Addressing these questions requires discussion supported by percolation theory, colloidal particle reinforcement theory, and related mechanistic frameworks.

At present, several mainstream strategies have been explored to enhance the mechanical performance of nanocellulose hydrogels, including the construction of NC systems, IPNs, DN hydrogels, and hydrogels with specially designed topological structures. However, each of these approaches faces distinct challenges in terms of scalability and further development toward industrial applications[57,62,66-68].

Nanocomposite hydrogels enhance mechanical properties by uniformly dispersing nanocellulose colloidal particles as fillers within a hydrogel matrix, allowing strong surface interactions between the filler and the matrix to form a three-dimensional reinforcing network. The key challenges of this strategy lie in identifying compatible nanocellulose fillers and hydrogel matrices, optimizing component ratios, and developing effective dispersion methods. According to percolation theory, effective mechanical reinforcement occurs only when the filler content reaches a percolation threshold, at which point a continuous filler-matrix interaction network is formed throughout the composite. However, in practical applications, excessively high filler loading often leads to reduced toughness and poor processability, ultimately counteracting the intended mechanical enhancement. Constrained by the limitations of current theoretical models, nanocomposite nanocellulose hydrogels remain distant from large-scale industrial implementation.

Interpenetrating polymer network hydrogels are composed of two or more independent hydrogel networks that are physically interwoven without covalent bonding between them. Because the individual networks remain chemically independent, each can retain its intrinsic properties, resulting in enhanced overall mechanical performance. Additional functional characteristics can be introduced by incorporating functional groups with specific structural roles. The major challenges associated with IPN hydrogels lie in the synchronous construction of multiple networks and the maintenance of long-term structural stability. Since the networks are held together primarily by physical entanglement rather than chemical bonding, they are prone to slippage and disentanglement under prolonged cyclic loading, leading to mechanical degradation. Moreover, due to difficulties in precisely controlling IPN formation, the underlying energy dissipation mechanisms remain insufficiently understood. As a result, IPN hydrogels are currently confined to laboratory-scale research, with further progress relying heavily on advances in theoretical understanding.

Double-network hydrogels represent a specialized class of IPN hydrogels characterized by highly asymmetric network structures.

Unlike conventional IPNs, DN hydrogels impose strict requirements on polymer network composition, monomer type, and crosslinking density. Typically, the first network is a highly crosslinked polyelectrolyte network, while the second network consists of a loosely crosslinked or non-crosslinked neutral polymer with high molecular weight, with the concentration ratio of the two networks generally below 5%. According to prevailing theories, the first network serves as a rigid scaffold that provides mechanical support, while the second network undergoes deformation to dissipate energy. Upon overloading and partial fracture of the first network, substantial energy is absorbed, and newly generated entanglement points reinforce the second network, thereby enhancing overall mechanical performance. Consequently, DN hydrogels often exhibit superior strength and toughness. However, the preparation of DN hydrogels is hindered by complex, multistep fabrication processes and limited controllability. Common two-step synthesis methods also suffer from potential interfacial inhomogeneity between the two networks, posing challenges for reproducibility and scalability.

Topological hydrogel design aims to enhance mechanical properties by regulating the topology of polymer chains within the hydrogel network, including entanglement, branching, and ring formation. By tailoring chain topology, specific mechanical properties can be selectively enhanced. The primary challenges of this approach include the lack of quantitative understanding of how specific topological features influence mechanical behavior, as well as the substantial difficulty in precisely controlling polymer chain topology. In addition, issues related to raw material cost and long-term structural stability of the final products remain unresolved.

In summary, although several viable strategies have been proposed to enhance the mechanical performance of nanocellulose hydrogels, their industrial translation remains constrained by limitations in existing reinforcement theories — such as percolation theory and colloidal particle/polymer gel reinforcement models. Continued theoretical refinement and experimental validation are required to bridge the gap between laboratory research and industrial application. At present, nanocomposite systems represent the most mature and industrially promising approach, while IPN and DN hydrogels may be suitable for small-scale applications. Topological hydrogel design, although conceptually appealing, remains highly dependent on further theoretical and methodological advances. To compare these reinforcement strategies more clearly, **Table 3** summarizes their representative systems, dominant mechanisms, advantages, and major limitations.

Table 3. Reported reinforcement strategies, mechanisms, advantages and limitations of nanocellulose hydrogels

Structural Strategy/ Representative System	Reported Reinforcement Mechanism	Reported Advantages	Reported Limitations / Challenges
Nanocomposite Hydrogel[61,62]; CNC/Polyacrylamide Composite Gel	CNCs act as reinforcing fillers and physical crosslinking points; Reinforcement depends on filler dispersion and filler-matrix interactions	Improved compressive performance; CNCs can accelerate gelation and enhance mechanical strength	Excessive filler loading may cause aggregation and reduce processability; Reinforcement depends on dispersion quality
Nanocomposite Conductive Hydrogel[15,57]; CNF/PPy or Nanocellulose-Reinforced Conductive Hydrogel	CNFs provide mechanical support and modifiable surfaces; Conductive components provide electrical pathways	Simultaneous mechanical enhancement and conductivity	Conductive fillers may cause dispersion problems and raise cost, stability, or biocompatibility concerns
IPN Hydrogel[67]; Alginate/Sericin/CNC Interpenetrating Network Composite Hydrogel	Interpenetrating networks improve physicochemical properties through physical entanglement and network coupling	Enhanced physicochemical properties and biological activity	Precise control of multiple networks and long-term stability remain challenging
Double-Network Hydrogel[21]; Tough Conductive Double-Network Zwitterionic Hydrogel Containing Nanocellulose	Rigid/soft network synergy and sacrificial energy dissipation improve toughness and fatigue resistance	Tough, conductive and anti-freezing performance for flexible sensors	Multicomponent formulation and preparation complexity may affect reproducibility
Cellulose-Composited Hydrogel[28]; Cellulose-Composited Hydrogels with Multiscale Mechanical Enhancement	Nanocellulose contributes to multiscale reinforcement and stress transfer within hydrogel networks	Improved multiscale mechanical properties	Structure-property relationships require further clarification
Topological/Anisotropic Design[75,76]; Bio-Inspired Anisotropic Nanocellulose Scaffolds	Directional alignment and anisotropic architecture guide mechanical and biological performance	Tunable anisotropic structures; Potential in tissue engineering	Requires precise processing control; Large-scale reproducibility remains to be validated

NOTE: PPy: polypyrrole.

Mechanical reinforcement in nanocellulose hydrogels is governed by the coupled effects of filler dispersion, interfacial bonding, network topology, and energy-dissipation mechanisms. Uniformly dispersed nanocellulose can form continuous stress-transfer pathways within the hydrogel matrix, while hydrogen bonding, ionic coordination, and covalent interactions determine the efficiency of interfacial load transfer. In DN and IPN systems, deformation-induced energy dissipation mainly arises from sacrificial bond rupture, network disentanglement, and reversible crosslinking. Future reinforcement strategies should therefore focus not only on increasing nanocellulose content or intrinsic filler strength, but also on controlling spatial distribution, interfacial compatibility, and dynamic reversibility within the hydrogel network.

4. Future Perspectives

Based on the above analysis, several promising research directions are proposed to guide the future development of nanocellulose-based hydrogels.

4.1 Research Gaps and Limitations

Despite extensive progress in cellulose hydrogel research — from raw material acquisition and synthesis to structural characterization, theoretical modeling, performance evaluation, and application optimization — significant gaps and limitations persist.

Systematic analyses of recent reviews and frontier studies indicate that, although nanocellulose hydrogels have achieved notable advances in structural design and functional exploration, most studies remain at the laboratory proof-of-concept stage, lacking comprehensive evaluation under long-term service conditions and realistic application scenarios[12].

First, the vast majority of cellulose hydrogel products reported to date are confined to laboratory-scale preparation, with substantial challenges remaining before large-scale production can be realized. In particular, for biomedical applications, critical issues such as raw material safety and non-toxicity, process scalability and practicality, and long-term stability and biocompatibility require extensive and authoritative validation. Current studies are predominantly limited to short-term experiments and animal models[69].

Second, existing theoretical models are still insufficient for accurately describing the structural complexity of cellulose hydrogels and elucidating the origins of their functional properties. Percolation theory[70-72], which employs a percolation threshold to predict the formation of three-dimensional interconnected networks and associated performance transitions, provides valuable guidance for nanocomposite hydrogel design. However, this theory oversimplifies cellulose hydrogel systems by neglecting key factors such as filler dispersion homogeneity and complex filler-matrix interactions, leading to inaccuracies in identifying performance transition points. Furthermore, percolation theory primarily addresses connectivity at the nano- and microscale, without accounting for larger-scale fiber entanglement, hierarchical network structures, or the dynamic behavior of hydrogels during service, limiting its applicability in designing complex multilevel structures.

Double-network theory[73,74], which explains high mechanical strength and energy dissipation through sacrificial network mechanisms, offers improved guidance for designing hydrogels with complex three-dimensional network topologies. Nevertheless, current experimental techniques are inadequate for precisely characterizing the coupled multilevel networks within cellulose hydrogels, making theoretical predictions challenging. Additionally, the construction of ideal DN structures often requires specialized crosslinkers and complex processing steps, which may compromise the inherent biocompatibility and environmental friendliness of cellulose-based hydrogels.

Third, although cellulose is abundantly available as a bio-based resource, fully leveraging this advantage requires the low-cost and high-efficiency extraction of high-quality nanocellulose from non-food biomass. This poses stringent requirements on extraction technologies. Existing physical, chemical, and enzymatic methods all suffer from inherent drawbacks, including broad aspect ratio distributions, high environmental burdens, low efficiency, and high costs, rendering large-scale production impractical. Consequently, the future development and application of cellulose hydrogels critically depend on continued advances in nanocellulose extraction technologies from non-food biomass.

4.2 Biomimetic Design and Intelligent Integration

Future research should focus on the development of high-performance hydrogels that mimic the hierarchical structures and anisotropic properties of biological tissues such as cartilage and skin. By adopting strategies such as directional freezing, mechanical force-induced alignment, or stress-assisted assembly, it is possible to engineer anisotropic architectures with tailored mechanical strength and mass transport behavior. Furthermore, integrating multiple functionalities — including sensing, actuation, drug delivery, and self-healing — into a single hydrogel system represents a key step toward the construction of intelligent biomedical platforms capable of simultaneous diagnostic sensing and therapeutic intervention[75]. For example, Jiao *et al.*[69,76] developed a facile swelling-induced nanofiber alignment strategy combined with an in situ biomineralization reinforcement approach to fabricate nanocellulose-based scaffolds with tunable anisotropic structures and mechanical properties. These scaffolds demonstrated significant potential in guiding the differentiation of bone marrow-derived mesenchymal stem cells, as well as in supporting cell growth and proliferation, highlighting their promise for tissue engineering applications.

4.3 Green Manufacturing and Life Cycle Assessment

Another critical direction lies in the development of environmentally benign, low-cost, and scalable processes for nanocellulose extraction and hydrogel fabrication. Although substantial progress has been achieved in material design and application exploration, translating nanocellulose hydrogels from laboratory-scale demonstrations to industrial-scale production remains a major challenge. Future research must therefore shift from an exclusive focus on enhancing material performance toward establishing a comprehensive and quantitative sustainability framework. This requires not only emphasizing the intrinsic "green" attributes of cellulose-based materials, but also applying systematic scientific methodologies — such as life cycle assessment (LCA) — to ensure true sustainability throughout the entire material life cycle. Such efforts are essential for supporting long-term environmental protection and sustainable agricultural development.

4.4 Multiscale Simulation and Data-Driven Design

The integration of computational modeling techniques (e.g., molecular dynamics simulations) with data-driven approaches such as machine learning offers powerful tools for advancing the rational design of nanocellulose hydrogels. These methods can be employed to elucidate hydrogen-bonding interactions between nanocellulose and polymer matrices, establish multiscale correlations linking molecular structure, network topology, and macroscopic properties, and predict the influence of parameters such as crosslinking density on bulk mechanical behavior. For instance, Maharjan *et al.*[77] utilized advanced characterization techniques to elucidate the three-dimensional spatial structure of regenerated cellulose-chitosan composite hydrogels and applied mathematical models to correlate their porous morphology with observed performance characteristics. Similarly, Costa *et al.*[78] characterized the microstructure of bacterial nanocellulose using scanning electron microscopy and finite element modeling, revealing pronounced three-dimensional anisotropy at the microscale. They further proposed a vertical expansion-induced stretching mechanism to explain the formation of anisotropic structures, providing valuable insights for the design and analysis of anisotropic cellulose-based hydrogels.

Future development of nanocellulose hydrogels should shift from proof-of-concept demonstrations toward mechanism-guided design. Among current reinforcement strategies, nanocomposite reinforcement appears to be the most feasible route for scale-up

because of its relatively simple formulation and compatibility with existing processing technologies. DN and IPN hydrogels can provide superior mechanical performance, but their multistep preparation and limited reproducibility remain major barriers to large-scale production. Topological design represents a promising long-term direction, although it still requires deeper theoretical understanding and more precise synthetic control. Bridging laboratory prototypes with practical applications will require the integration of scalable nanocellulose extraction, controllable interfacial assembly, standardized performance evaluation, and data-driven formulation optimization.

5. Conclusions

Nanocellulose hydrogels are sustainable soft materials that combine the renewability, excellent mechanical properties, rich surface chemistry, and biocompatibility of nanocellulose with the porous, water-retaining, and tunable three-dimensional network structure of hydrogels. This review outlines the preparation of nanocellulose, the construction and functionalization of hydrogels, and their advanced applications. Various preparation and crosslinking strategies enable the regulation of morphology, mechanics, and functionality. Key factors, including dispersion, interfacial interaction, crosslinking density, energy dissipation, and network structure, determine the final performance, endowing hydrogels with high strength, self-healing, stimuli responsiveness, conductivity, and stability.

Nanocellulose hydrogels show broad application prospects in biomedicine, flexible electronics, smart devices, 3D printing, food packaging, and agricultural water retention. However, challenges still exist in balancing mechanical strength and multifunctionality, filler dispersion, interfacial compatibility, theoretical description, long-term stability, biosafety, and scalable green manufacturing. Future research should focus on mechanism-based design, precise structure control, standardized evaluation, and industrial translation. With continuous development, nanocellulose hydrogels will become important sustainable soft materials for multidisciplinary applications.

Author Contributions

Yanhua Liu conceived, supervised, and revised the manuscript. Zheng Xu collected the literature, drafted the paper, and prepared all figures and tables. Pengjun Zhao analyzed the literature and revised the manuscript. Zhengjie Lin organized the references and polished the manuscript. All authors have read and approved the final version of the article.

Artificial Intelligence (AI) Statements

During the preparation of this manuscript, the authors used AI-assisted tools only for language polishing, grammar checking, structural organization, and readability improvement. All scientific content, references, interpretations, and conclusions were reviewed and verified by the authors, who take full responsibility for the manuscript.

Informed Consent Statement

Not applicable.

Conflicts of Interest

The authors declare no conflicts of interest.

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