Sustainable Hydrogels from Cellulose

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Hydrogels are three-dimensional, hydrophilic networks composed of a variety of polymeric materials linked by chemical (covalent bonds) or physical (non-covalent interactions) cross-linking. The unique hydrophilic nature of hydrogels provides a promising solution for food packaging systems, specifically in regulating moisture levels and serving as carriers for bioactive substances, which can greatly affect the shelf life of food products. In essence, the synthesis of cellulose-based hydrogels (CBHs) from cellulose and its derivatives has resulted in hydrogels with several appealing features such as flexibility, water absorption, swelling capacity, biocompatibility, biodegradability, stimuli sensitivity, and cost-effectiveness.

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1. Synthesis of Cellulose-Based Hydrogels (CBHs)

Cellulose-based hydrogels (CBHs) are synthesized from various sources including cellulose and its derivatives such as methyl cellulose (MC), hydroxypropyl cellulose (HPC), hydroxypropyl methyl cellulose (HPMC), and carboxymethyl cellulose (CMC). These derivatives have modified chemical structures that can improve the solubility, viscosity, and other properties of cellulose, making them useful in a range of applications [1][2]. Furthermore, ester derivatives of cellulose such as acetate trimellitate, acetate phthalate, hydroxypropyl methyl phthalate, hydroxypropyl methyl phthalate acetate succinate, etc., can also be used to create CBHs [3][4]. These derivatives have different properties and can be used to tailor the properties of CBHs for specific applications. CBHs are also synthesized by combining cellulose with other polymers such as polyelectrolyte complexes, or blending with other polymers [5][6]. This provides the potential to improve the mechanical and other properties of the resulting CBHs. One of the challenges of synthesizing hydrogels from cellulose or its derivatives is that they are not easily soluble in common solvents, particularly native/pure cellulose [1][3]. This makes dissolving and processing these materials into hydrogels more difficult. To address this challenge, researchers have developed various methods for dissolving cellulose or its derivatives. Some of the most commonly used solvents include alkali/urea or thiourea, LiCl/dimethylacetamide, N-methylmorpholine-N-oxide, and ionic liquids. Alkali/urea or thiourea solvents function by disrupting the hydrogen bonds between the cellulose molecules, whereas LiCl/dimethylacetamide can dissolve both cellulose and some of its derivatives [7][8]. N-methylmorpholine-N-oxide is another solvent that has been used to dissolve cellulose, and ionic liquids are a relatively new class of solvents that have been shown to dissolve cellulose and its derivatives [8][9].

The process of synthesizing CBHs has evolved over the years, with different approaches being employed depending on the specific crosslinking and processing techniques used. This has been conducted in order to achieve the desired properties and applications of the hydrogel, which can vary depending on the intended use of the material. Principally, there are various approaches including physical and chemical crosslinking, and **Table 1** depicts the synthesis methods, crosslinking mechanisms, and characteristics observed for different types of cellulose used in the fabrication of CBHs for various applications. These methods can improve the viscoelasticity and mechanical properties of hydrogels, altering the properties of the hydrogel to match specific criteria for packaging applications. Physical crosslinking involves the formation of CBHs through non-covalent interactions such as hydrogen bonding, van der Waals forces, or the physical entanglement of polymer chains [1][10]. This method has gained increasing interest in recent years due to the wide range of properties and applications it offers compared to chemically crosslinked polymers. In addition, physical crosslinking does not require the use of any chemical reagents, making it a simple and cost-effective method for producing hydrogels. Such crosslinking can be employed through different methods including heating or cooling a polymer solution, complex coacervation, freeze–thawing, hydrogen bonding, ionic interaction, or self-assembly. One example of physical crosslinking is the synthesis of CBHs using freeze–thaw cycles [11][12]. In this method, cellulose is dissolved in a suitable solvent, and the resulting solution is subjected to repeated cycles of freezing and thawing. This process results in the formation of a hydrogel due to the aggregation of cellulose chains. In one study, a high-performance CMC-based hydrogel film with desirable stretchability, UV-blocking, antioxidant, self-healing, and adhesion features was developed using the freeze–thaw method and was found to be effective for food packaging [13].

Table 1. Various methods for the development of cellulose-based hydrogels.

Synthesis Routes	Crosslinking Mechanism	Types of Cellulose	Characteristics	References
Physical crosslinking		Native cellulose	One-pot supramolecular bio-based hydrogels with high strength and pH sensitivity	[11]
	Freeze thawing	Carboxymethyl cellulose (CMC)	An eco-friendly method of repeated freeze–thaw cycles to develop hydrogel composites based on pineapple peel CMC, polyvinyl alcohol, and mesoporous silica SBA-15	[12]
	Ionic interaction	Cellulose microfibrils	Synthesis of cellulose microfibrils hydrogels using the TEMPO-oxidation system with increased storage modulus, compression strength, and surface area	[14]
		Nanocellulose	Development of sugarcane bagasse nanocellulose-based hydrogel as a colorimetric freshness indicator for detecting chicken breast deterioration	[15]

Synthesis Routes	Crosslinking Mechanism	Types of Cellulose	Characteristics	References
	Hydrogen-bonding interaction	Native cellulose	Facile and low-cost cellulose-based nanocomposite hydrogels with improved mechanical characteristics and adsorption to heavy metal ions utilizing hydroxyapatite (HAP) nanoparticles	[16]
	Complex coacervation	Carboxymethyl cellulose (CMC)	Amaranth protein isolate/CMC complex coacervates develop betanin-containing microcapsules for the creation of edible gelatin films with low light transmission and high antioxidant activity	[17]
	Hydrophobic interaction	Bacterial cellulose (BC)	Synthesis of a sodium alginate-bacterial cellulose-based nanocomposite hydrogel with multi-layered porous surfaces capable of swelling, releasing, and being biocompatible for substrate use	[18]
Chemical crosslinking	Grafting	Hydroxyethyl cellulose (HEC)	Synthesis of self-assembled supermolecular hydrogels based on HEC with potential applications as bacteriostasis materials	[19]
	Crosslinking agents	Carboxymethyl cellulose (CMC)	Crosslinked CMC/gelatin hydrogel films loaded with ZnO nanoparticles using glutaraldehyde as a crosslinking agent with antibacterial and antioxidant characteristics for sustainable food packaging applications	[20]
		Hydroxyethyl cellulose (HEC)	Development of citric acid cross-linked antimicrobial hydrogel films based on HEC and ZnO for food packaging applications	[21]
		Bacterial cellulose (BC)	Crosslinked bacterial cellulose hydrogels with improved mechanical properties and increased water retention capacity employing citric acid and epichlorohydrin as crosslinking agents	[22]
	Enzyme crosslinking	Bacterial cellulose (BC)	Transglutaminase-enzymatic crosslinking of BC/fish collagen composites with increased tensile strength and water vapor permeability	[23]
	Radical polymerization	Carboxymethyl cellulose (CMC)	Fabrication of a superabsorbent hydrogel for water retention and	[24]

Synthesis Routes	Crosslinking Mechanism	Types of Cellulose	Characteristics	References
			sustained release in advanced agricultural applications	
		Native cellulose	The synthesis of remarkably stretchable and compressible cellulose ionic hydrogels for flexible strain sensors	[25]
	Radiation crosslinking	Hydroxypropyl Methylcellulose (HPMC)	Developed biodegradable HPMC hydrogels with increased strength and swelling qualities using high-energy radiation from electron accelerators.	[26]
		Carboxymethyl cellulose (CMC)	An effective method for synthesizing CMC hydrogels with tailored swelling behavior by varying the radiation dose and the degree of carboxymethylation for targeted applications	[27]

chemical for many and have [28][29]. In contrast, ch… because they [30][31] Chemical within the including enzyme chemical [31][32] ables like aches are used to create … ation rate. The crosslinking agents react with the functional groups on the polymer chains such as the hydroxyl groups on cellulose to form covalent bonds, resulting in a three-dimensional network structure. An example of the use of chemical crosslinking in CBHs is the synthesis of hydrogel films based on hydroxyethyl cellulose (HEC) and zinc oxide (ZnO), which employed citric acid as a crosslinking agent. These hydrogels revealed excellent swelling and hydrophilicity as well as potential antibacterial characteristics, making them appropriate for application as food packaging materials [21].

Furthermore, grafting is another promising approach in the development of CBHs for food packaging applications [33][34]. Principally, the grafting method for CBHs entails the covalent attachment of polymer chains such as polyethylene glycol (PEG) or poly (acrylic acid) (PAA) to the surface of the hydrogel network [35][36]. This covalent attachment is achieved through various techniques such as chemical grafting, radiation-induced grafting, or enzymatic grafting. Grafting allows for the modification of the surface properties of the hydrogel, improving interactions with food packaging and enabling the controlled release of bioactive molecules in active packaging applications. Additionally, grafting improves the mechanical properties of the hydrogel, making it more suitable for food packaging applications that require specific mechanical properties [37][38]. Moreover, grafting is used to introduce functional groups into the hydrogel, allowing for the development of smart hydrogels that respond to external stimuli such as pH or temperature, which can be beneficial for food packaging applications where environmental conditions need to be controlled to maintain food quality and safety [39]. Other chemical crosslinking techniques employed in the development of CBHs, as described in **Table 1**, include radiation crosslinking and radical polymerization [24][25][26][27]. Radiation crosslinking involves exposing the cellulose-based hydrogel to ionizing radiation such as gamma rays, X-rays, or electron beams, which creates free radicals that react and form crosslinks between the polymer chains [40]. The advantage of radiation crosslinking is that it does not require any chemical crosslinking agents, and it can be used to crosslink hydrogels in a controlled and homogeneous manner. While radiation crosslinking is a useful technique for synthesizing CBHs, it does have limitations such as requiring specialized equipment and facilities, potential damage to polymer chains, is a time-consuming process, and high cost [31][41]. Radical polymerization, on the other hand, involves the use of chemical initiators that generate free

radicals, which then react with the monomers to form polymer chains that crosslink with the cellulose-based hydrogel network [42][43]. The advantage of radical polymerization is that it allows for a high degree of control over the crosslinking process, allowing for the precise tuning of the properties of the resulting hydrogel [44].

In general, the selection of a crosslinking technique for the synthesis of CBHs is determined by the specific application and the desired properties of the hydrogel. Although physically crosslinked hydrogels do not require crosslinking agents or chemical modification, they need further modifications to improve high mechanical strength. Alternatively, while chemical crosslinking is an efficient approach for synthesizing CBHs with better mechanical and thermal properties, its toxicity and lack of biodegradability make it undesirable for use in food packaging applications. Instead, researchers are more interested in investigating alternate methods for creating hydrogels that are safer, more sustainable, and more suited for packaging applications through the integration of cellulose and its derivatives with novel synthesis techniques.

2. Characterization of CBHs

The utilization of bio-based hydrogels in food packaging in various forms such as coatings, films, and composites offers a promising solution to the environmental issues related to the usage of non-renewable and non-degradable petroleum-based polymers [45]. However, when utilizing such hydrogels for food packaging, it is crucial to consider their functional properties for the intended application. In this context, the fundamental characteristics of the hydrogel such as its mechanical strength, high absorption or superabsorbent capacity, wettability, and barrier properties are crucial factors to consider alongside cost-effectiveness, sustainability, and biodegradability, regardless of whether CBHs are utilized for traditional, active, or smart packaging purposes [30][31]. In general, numerous factors such as the cellulose source, crosslinking method, crosslinking density, degree of substitution, and processing conditions, influence the performance of CBHs for food packaging applications [46]. The hydrogel characteristics including high water absorption capacity, biocompatibility, biodegradability, mechanical strength, and functional properties can be tailored to meet the needs of various food packaging applications by identifying and optimizing these factors. One of the key features of CBHs is the swelling behavior, which is an important characteristic that determines its water uptake capacity and retention ability. CBHs have exhibited tremendous potential in absorbing large amounts of water, up to 1000 times of their original dry weight, making them ideal for use in food packaging applications where moisture control is crucial [3][47]. For instance, a CMC-based hydrogel film prepared with polyvinylpyrrolidone (PVP) as a binding agent demonstrated significant water retention as well as water uptake capacity during a swelling and deswelling study and was referred to as a promising food packaging material for fruits and vegetables to keep them fresh for longer periods. In addition, the presence of PVP in the CMC-based hydrogel improved the mechanical characteristics during the hydrothermal stage in the various testing temperature and relative humidity conditions [48]. Furthermore, the swelling behavior of CBHs is controlled by varying the crosslinking density, degree of substitution, and type of cellulose used. Crosslinking density affects the degree of swelling, with a higher crosslinking density resulting in a lower degree of swelling [33]. Wettability, or the ability to interact with water or other fluids, is another crucial property affecting the features of CBHs for food packaging applications. For applications requiring fluid absorption or adhesion, high-wettability is preferred for

hydrogels, whereas hydrophobicity or repellency is preferred for low-wettability applications [49]. **Table 2** summarizes some of the key properties and significance of CBHs that are important for food packaging applications.

Table 2. Key properties, techniques, and significance of CBH characterization in food packaging.

Property	Main Characteristics	Analytical Techniques	Significance	References
Swelling index	Assessment of CBH performance in fluid absorption and swelling behavior	Gravimetric analysis; swelling ratio	Effectiveness of the packaging in preserving the food product by absorbing excess amount of liquid, typically water, and creating a protective barrier around the food product.	[48][50]
Wettability	Examining the degree of interaction between CBHs surfaces and fluids	Contact angle measurement; surface energy measurement; (AFM)	Consideration of wettability of CBHs surface for designing food packaging materials with emphasis on barrier properties and prevention of food product loss or contamination	[51]
Mechanical strength	Investigation of CBHs endurance and performance under specific environmental constraints	Tensile strength; elongation at break; compression strength; rheological characteristics	Preventing physical damage during handling, transportation, and storage by effectively protecting the food product while maintaining the structural integrity and functionality of packaging material under diverse loading conditions	[13][52]
Thermal stability	Thermal analysis under specific temperature conditions	Thermogravimetric analysis (TGA); differential scanning calorimetry (DSC)	Essential for designing food packaging materials to withstand high temperatures during processing, storage, and	[13][20]

Property	Main Characteristics	Analytical Techniques	Significance	References
			transportation without compromising the quality and safety of the food product	
Physical/morphological characterization	Visualizing the structural features of CBH networks	Scanning electron microscopy (SEM); AFM; field emission scanning electron microscopy (FESEM)	Designing suitable food packaging materials with the optimal porosity, barrier characteristics, and mechanical strength to maintain food quality and safety by visualizing the structural aspects of CBH networks	[48][53][54]
Chemical characterization	Identifying the chemical structures, molecular arrangements, and functional groups of the developed CBHs	Fourier transform infrared (FTIR) spectroscopy; X-ray diffraction (XRD); (NMR) spectroscopy; Raman spectroscopy	Optimizing the performance of CBHs using chemical composition, degree of crystallinity, and molecular orientation of the hydrogels	[48][55][56]

[31][33]

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their intended function under a range of temperatures and humidity levels. In this context, a highly elastic and versatile hydrogel film comprised of CMC, polyvinyl alcohol (PVA), poly(ethylene imine) (PEI), and tannic acid (TA) for food packaging applications possessed a remarkable tensile strain of up to 400% without rupture [13]. Furthermore, several studies have revealed that a range of factors including the degree of polymerization of the cellulose chains, type, and concentration of the crosslinking agent, choice of solvent, and fillers as well as the method of preparation and processing have a substantial effect on the mechanical strength of CBHs [57]. These factors have a vital influence in determining the mechanical properties of CBHs, and identifying their effects facilitates the development of hydrogels with improved mechanical strength. Similarly, the thermal stability of CBHs is critical for developing food packaging materials that can endure high temperatures during processing, storage, and transportation without compromising the quality and safety of the food product. Different analytical techniques including differential scanning calorimetry (DSC), dynamic mechanical analysis (DMA), and thermogravimetric analysis (TGA), are used to analyze this. These methods offer important insights into the thermal behavior of CBHs including their glass transition temperature, thermal degradation temperature, and storage modulus as a function of temperature. One study that investigated the synthesis of a CMC-based hydrogel packaging film used TGA and DSC analyses to establish how thermal stability improved when tannic acid was incorporated. This investigation additionally confirmed that CMC and tannic acid interacted effectively together to produce the hydrogel [13]. In addition, structural features as well as the chemical compositions, molecular configurations, and functional groups of the developed CBHs can be examined as well as identified using physical, morphological, and chemical characterization approaches [53][54][55][56]. Such properties offer significant insights into interpreting the characteristics of CBHs and enhancing their functionality as food packaging materials.

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