Recent applications of regenerated cellulose films and hydrogels in food packaging

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Highlights

- The applications of regenerated cellulose films and hydrogels in food packaging are promising.
- The mechanical and barrier properties of regenerated cellulose materials can be modulated to fulfill different requirements.
- Regenerated cellulose films and hydrogels provide versatile matrices for active and intelligent packaging construction.
- Regenerated cellulose materials are generally safe but the migration of functional fillers should be considered carefully.

Abstract

Nowadays, much attention has been paid to combat international plastic pollution. Especially, the nondegradable plastic packaging materials have created significant disposal and pollution issues threatening human health and development. In addition to the promised 100% recyclable packaging, the utilization of biodegradable packaging materials can also help address the issue. Cellulose is abundant and renewable, and cellulose-based materials have been regarded as an alternative to petroleum-based plastic food packaging. With the development of cellulose solvents, various regenerated cellulose films and hydrogels have been fabricated for different applications. In this review, we summarize the recent progress in the preparation of regenerated cellulose films and hydrogels, and highlight their potential applications as biodegradable packaging, active packaging, and intelligent packaging. Finally, the biodegradability and safety of cellulose-based materials are stated, and future opportunities and challenges in this active research area are described.

Keywords: Food packaging; dissolution; regeneration; cellulose; films; hydrogels

Introduction

Food packaging waste comprises approximately one-third of all Canadian household waste, and only 20% is recovered for reuse and recycling. Among them, synthetic plastics suffer from poor biodegradability and limited ways to reuse and recycle, and thus cause serious environmental issues. Food packaging developed from natural polymers provides an alternative solution and has attracted more and more attention. Cellulose is the most abundant renewable and biodegradable polymer on earth, and has several advantages such as inexpensiveness, low density, non-toxicity, versatility, and superb mechanical properties [1]. Cellulose-based food-packaging materials, for example paper and cloth, have been widely utilized. However, with the requirements of improved protection of food products, the exploration of novel food packaging with multiple functional properties is urgent. The development of various cellulose solvents enabled the processing of cellulose into different forms of materials. Especially, after dissolution, regenerated cellulose films and hydrogels can be fabricated by modulating physical and chemical interactions. They possess three-dimensional porous structures, which allow the incorporation of functional fillers and are thus applied as active or intelligent packaging materials [refs, 6-12?]. According to the Web of Science search result, 10,254 research papers on cellulose-based materials have been published in last two years, and 404 (3.94%) of them are related to food packaging applications. The utilization of cellulose-based materials in food packaging is promising and still has a huge potential. Several reviews have summarized different forms of cellulose-based materials such as cellulose films [2], fibers [3], hydrogels [4], microspheres [5], and composites [1], without the focus on food packaging purpose. Therefore, in this review, we have summarized recent strategies of fabricating regenerated cellulose-based films and hydrogels and their potential applications in food packaging. The considerations of biodegradability and toxicity are highlighted, and future opportunities, challenges, and research directions are described.

Recent studies on regenerated cellulose film and hydrogel production

Extensive studies have discussed various cellulose solvents including *N*-methylmorpholine-*N*-oxide (NMMO), LiCl/DMAc, ionic liquids (ILs), and alkali/urea solutions to develop a more efficient and environmentally friendly method for cellulose dissolution and regeneration. Figures 1 (a) to (d) illustrate the appearance of regenerated cellulose films produced from different raw materials and solvents. Recent research mainly focused on optimizing dissolution conditions and strategies of non-solvent regeneration that influence morphological and mechanical properties of regenerated cellulose films [13]. For instance, cellulose source [14], wood type (hard or soft) [9], pulping process (acid sulfite or kraft sulfate) [9], and coagulants used to regenerate cellulose were compared to investigate their effects on the morphology of cellulose films [15,16]. A novel cellulose pretreatment for the dissolution in LiCl/DMAc was recently proposed that used O_2 plasma irradiation to activate cellulose chains and eliminate glycosidic bonds, leading to the improved dissolution of cellulose and enhanced mechanical properties of regenerated films [17]. Zheng and co-workers applied three imidazolium-based ILs, namely AmimCl, BmimCl, and EmimAc, to dissolve cellulose , where BmimCl showed the best dissolution capacity to dissolve cellulose within 8 min at 90 ℃ [18]. However, the AmimCl dissolution system was the optimal option due to the short gel formation time (13 min) and the superior tensile strength of the produced film, revealing the good molecular network arrangements and strong intermolecular interactions. The effect of dissolution temperature was highlighted by Wei et al. that in the range of -2 to -12.5 ℃, a decrease in temperature resulted in more cellulose dissolution in NaOH/urea solution [19]. The regenerated cellulose film prepared at -10 ℃ was optimal due to its best mechanical properties, transparency, and thermostability.

For the production of cellulose hydrogels, the double-crosslinking strategy has been applied recently by combining chemical and physical crosslinking to obtain the hydrogels with dimensionally stable and recoverable double network structures [20,21]. For example, nanocellulose was crosslinked with epichlorohydrin (ECH) and metal salts ($FeCl_3$ or $CaCl_2$) to produce double-crosslinked hydrogels [22]. A three-

dimensional network was formed due to the interconnected nanofibrils with no aggregation in crosslinked domains. A highly deformable cellulose hydrogel (126% of tensile strain) containing chitin and chitosan was formed via free radical polymerization of synthesized allyl cellulose, and had good transparency and ionic conductivity at -20 ℃ working condition [23]. This hydrogel was further immersed in saturated NaCl solution to initiate double crosslinked network, which thus had an improved tensile strain (236%) and was still stretchable at -24 ℃, because the incorporation of physical crosslinking maintained the hydrogel integrity [24]. A similar cellulose hydrogel with excellent mechanical properties, good transparency, and anti-freezing capacity was also reported, where ECH was used to crosslink cellulose dissolved in benzyltrimethyl ammonium hydroxide solution [25].

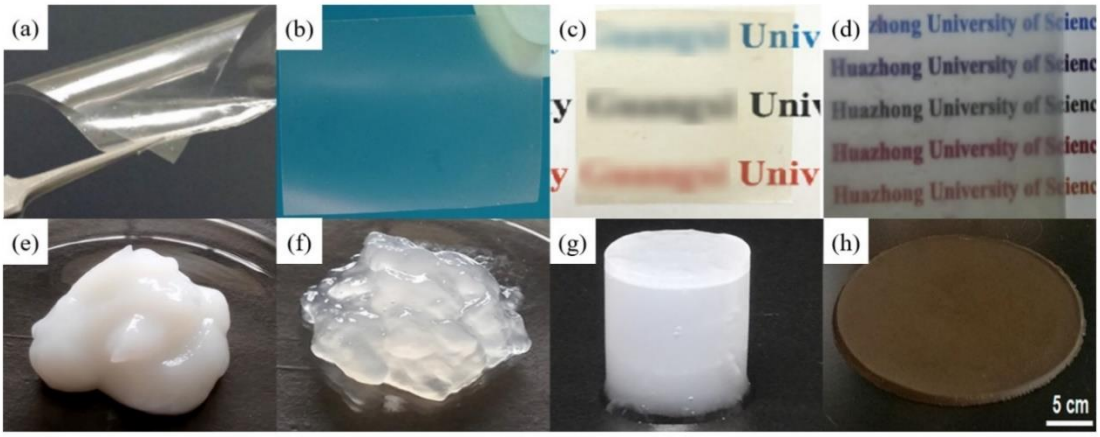

Figure 1. Photos of regenerated cellulose films (top) and hydrogels (bottom) prepared from (a) xxx in LiCl/DMAc [16], (b) xxx in NaOH/urea solution [26], (c) xxx in AmimCl [27], (d) xxx in BmimCl [28], (e) bagasse cellulose filaments (BCF) [29], (f) oxidized BCF [29], (g) Zn^{2+} crosslinked BCF [30], and (h) xxx cellulose with amino-terminated hyperbranched polyamic anchored nanosilver [31].

Potential applications in food packaging

Regenerated cellulose films

Cellophane is the commercial transparent film regenerated from a viscose solution; however, carbon disulfide and other by-products of viscose process cause serious environmental pollution [32]. Currently, regenerated cellulose films are prepared from eco-friendly solvent systems, and usually form composites with other polymers or additives to improve mechanical properties, optical properties, barrier properties, and thermostability. The applications of regenerated cellulose films in food packaging can be mainly classified into three aspects: biodegradable packaging, active packaging, and intelligent packaging. A pure cellulose film regenerated from durian rind dissolved in LiCl/DMAc was regarded as the alternative to cellophane [33]. It had a smooth surface, good transparency, high strength (44 MPa), good thermostability, and biodegradability (100% decomposed in 4 weeks). The cellulose film prepared from xxxxxx dissolved in ionic liquid showed comparable tensile strength and thermal stability, and the pineapple sample packaged by the regenerated film showed the decreased weight loss and well

maintained firmness and vitamin C content after 2 days [34]. Another similar cellulose film regenerated from xxxxxx dissolved in ionic liquid also demonstrated the capacity to extend the storage life and commodity rate of mango to about 7 days [35]. The incorporation of nanoparticles (NPs, nanocellulose, metallic nanoparticles, etc.) and polymers (chitosan, etc.) is the traditional way to reinforce the mechanical and barrier properties of regenerated cellulose films [38-40]. A recent study recommended that the addition of Ca^{2+} in cellulose/ionic liquid solution significantly improved the tensile strength (85.86 MPa) and thermal stability (351 ℃) of regenerated cellulose films; however, the transparency was reduced [27]. The modulation of polymer network structure is another way to enhance the performance. Ye and co-workers revealed that the well-ordered arrangement of polymers at nanoscale and macroscale facilitated the improvement of mechanical and optical properties [7]. A dual crosslinking approach was applied to control the aggregation of cellulose chains and resulted in a nanofiber-structured cellulose film with remarkable tensile strength (253.2 MPa) and top value of transparency (91%). Wang and co-workers obtained a similar film by using benzyltrimethyl ammonium hydroxide as solvent followed by a simple water evaporation strategy in regeneration [44]. The fabricated film had a dense network, high tensile strength (158 MPa), excellent transmittance (89.94%), and good gas barrier properties in a 40% humidity environment [44]. At the same time, some research works focused on the low elasticity and high UV transparency of regenerated cellulose films [36], which may lead to the ruptured packaging and deteriorated food [37]. For instance, the elongation percentage, water vapor permeability, and visual appearance of cellulose films were improved by the addition of glycerol and polyvinyl alcohol [36]. Besides, the incorporation of 2 wt% of graphene oxide (GO) in cellulose films endowed good visible light transmittance (78%) and UVA (66.7%) and UVB (54.2%) shielding property [41]. The enhanced UV shielding capacity and hydrophobicity of cellulose films were also observed after the addition of CeO_2 [42]. Typical properties and potential applications of some recently reported cellulose films are summarized in Table 1.

Table 1. Typical properties and potential applications of recently reported regenerated cellulose films and hydrogels.

Forms	Compositions	Tensile strength (MPa)	WVP (10^{-9} g/m·s·Pa)	Initial degradation temperature (℃)	Applications	Refs
Films	CMC-Cur-ZnO	41.8	1.67	210	Antioxidant and antimicrobial packaging	[45]
	Durian rind C	44	/	270	"Green" and low-cost packaging	[33]
	BC-ZnO	92.4	/	337	Highly flexible packaging	[46]
	RC-ZnO	126.61	5.42	270	Antimicrobial packaging	[47]
	GS-MCC	41.87	2.49	170.3	"Green" and low-cost packaging	[48]
	Gelatin-CNC	20	0.03	200	Biodegradable packaging	[49]
	Gelatin-BC-MgO	0.71	0.03	/	Egg packaging and preservation	[50]
	PVA-Gelatin-CNC	13.8	0.46	250	Biodegradable packaging	[51]
	Reactive CNF	47	3.40	260	Hydrophobic packaging	[52]
	CNF-ZnO-GSE	140	0.51	221	Antioxidant and antimicrobial packaging	[53]
	MC-CNF-SPA	46.6	0.02	230	Intelligent packaging	[54]
	FP-SCNF-FP	287	/	/	Low oxygen-permeable packaging	[55]
	OEO-Tween-CNF	24.63	/	/	Antimicrobial packaging	[56]
	OC-Nisin peptide	99.2	0.004	/	Antimicrobial packaging	[57]
	CMC-Starch	32.6	3.27	234	Transparent packaging	[58]
	GG-HEC-Lignin	39	2.18	148.2	Antioxidant and biocompatible packaging	[59]
Hydrogels	PVP-CMC (film)	24.4	0.23	/	Biodegradable packaging	[60]
	Collagen-CNC	0.90	/	/	Biocompatible and biodegradable packaging	[61]
	TOBCF-Zn^{2+}	0.28	/	/	Intelligent packaging	[29]
	C-PAAm	0.63	/	150	Highly flexible packaging	[62]
	$NaIO_4$-CNF-PVA	0.43	/	310.9	Thermostable packaging	[63]

Note: WVP, water vapor permeability; CMC, carboxymethyl cellulose; Cur, curcumin; C, cellulose; BC, bacterial cellulose; RC, regenerated cellulose; GS, *Lemang* bamboo; MCC, microcrystalline cellulose; CNC, cellulose nanocrystals; GSE, grape seed extracts; MC, methyl cellulose; CNF, cellulose nanofibrils; SPA, saffron petal anthocyanin; FP, fluoropolymer; SCNF, succinylated cellulose nanofibers; OEO, oregano essential oil; OC, 2,3-dialdehyde cellulose; GG, gellan gum; HEC, hydroxyethyl cellulose; PVP, polyvinyl pyrrolidone; TOBCF, TEMPO-oxidized bagasse cellulose filaments; PAAm, polyacrylamide; PVA, polyvinyl alcohol.

Active food packaging is one of many possible applications of regenerated cellulose films, in which antimicrobial agents could be incorporated to inhibit the growth of foodborne bacteria [Carbohydrate Polymers 241 (2020) 116256, 39,64,65]. In recent years, much attention has been paid to natural antimicrobials such as essential oils and extracts [66]. For instance, poacic acid as a plant-based antimicrobial agent was added in cellulose film to inhibit the growth of *S. aureus* [56], while oregano essential oil in the film showed an excellent inhibition rate (99.99%) against *E. coli* and *L. monocytogenes* [31]. A comparative study indicated that the cellulose films incorporated with Ag, ZnO, and CuO NPs inhibited the growth of *E. coli* and *S. aureus* through the release of reactive oxygen species [39]. Ag NPs were preferred due to their best inhibition capacity against both bacteria [39]. Gu and co-workers demonstrated that hyperbranched polyamide-amine could be used as a binder to ember Ag NPs and further control the release rate [31]. The produced film exhibited a microbial inhibition capacity and maintained the freshness of cherry tomatoes for 9 days [31].

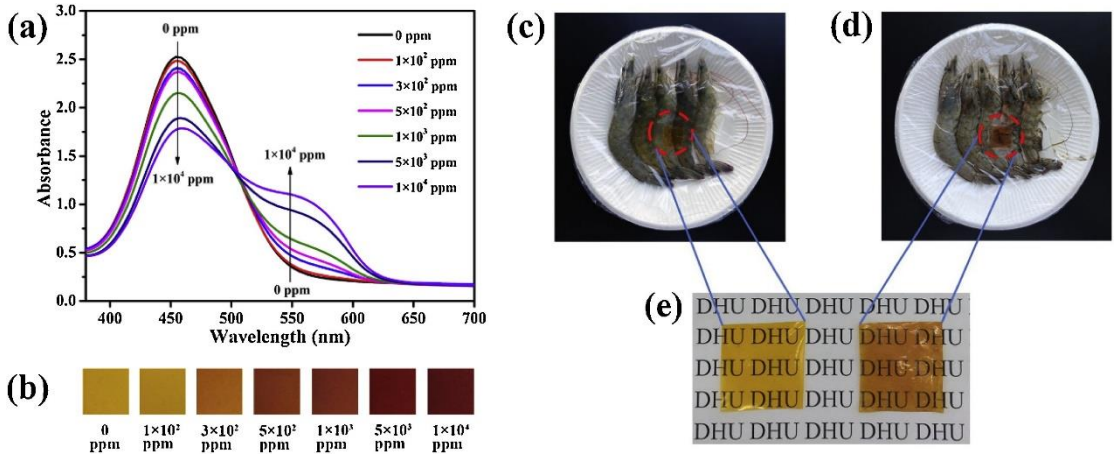

Figure 2. (a) UV-vis spectra and (b) optical images of intelligent cellulose films at different concentrations of ammonia solution. The color of cellulose films (c) before shrimp spoilage, (d) after shrimp spoilage, and (e) applied cellulose films [67].

Intelligent cellulose-based films were also designed and employed to monitor the quality and condition of packaged food. Ding et al. [67] revealed a pH-responsive film composed of regenerated cellulose, polyvinyl alcohol, and acidochromic dye, which showed a pH-responsive color change within pH 7-12, good tensile strength (35 MPa), thermostability (about 150 °C), and leakage resistance in acidic and basic environments. The methyl cellulose-based film loaded with saffron petal anthocyanin was reported with a wide pH-response range from 1 to 14, and successfully applied to indicate the freshness of lamb meat [54]. As shown in Figure 2, the prepared intelligent film could be used to detect ammonia formation and indicate the freshness of shrimps [67]. The anthocyanins-embedded cellulose/chitosan film had a wide pH response ranged from 2 to 12 and an excellent color stability for one month [68], while the similar cellulose/chitosan film embedded with alizarin was applied to monitor the spoilage of minced beef [69]. The pH value of minced beef elevated from 6.2 to 6.76 after storage at 4 °C for 4 days, and the color of the film changed correspondingly from brown to purple, revealing the high microbial load (>7 log CFU/g) beyond the acceptable limit [69].

Regenerated cellulose hydrogels

Recent studies on regenerated cellulose hydrogels are summarized in Table 1. Compared to regenerated cellulose films, the applications of hydrogels in food packaging are relatively less. Cellulose hydrogels can be regarded as an attractive absorbent material with superhydrophilicity, good structural stability after water absorption, and negligible influence on the sensory attribute of food [70,71], and be applied in food packaging to control the humidity and water activity of food. A superabsorbent cellulose hydrogel was prepared by crosslinking CMC with ECH, which had a desirable water retention value of 725 g water/g dry hydrogel [71]. Yang et al. provided a novel method to fabricate cellulose hydrogels by crosslinking with methylenebis acrylamide (MBA) in LiOH/urea solvents [72]. The interactions among water and -OH and -NH groups from polymer network endowed hydrogels with a good water absorption capacity of 220 g water/g dry hydrogel. The composite hydrogel of CMC and PVP at a ratio of 1:1 could absorb 1134% water, and delayed the deterioration of blueberries for 15 days, While the addition of guar gum slightly decreased the water absorption capacity (895%) but enhanced the mechanical properties [60]. A similar CMC-PVP-based hydrogel was applied to package table grapes, spinach, and tomatoes at room temperature for 30 days [73]. Alam and Cristopher noted that the water-absorption property of cellulose hydrogels was relevant to the concentration and chemical nature of polymers, and the optimal water absorption capacity (610 g water/g dry hydrogel) was found when cellulose hydrogel contained 25% of chitosan [74]. Another research focus was on the improvement of mechanical resistance by incorporating crosslinkers, polymers, and metallic compounds in cellulose hydrogels. Huber and co-workers [75] used undissolved micro-cellulose as a crosslinker to enhance the mechanical properties of hydrogels, resulting in lower gelation temperature and shorter heating time during the gelation process. The reinforcement of mechanical properties of cellulose hydrogels via the incorporation of Fe^{3+} was also elucidated, and the increase of Fe^{3+} content at low iron ion concentration levels formed more tridentate coordinates with the carboxyl groups of cellulose [76]. However, the iron ions at high

concentrations transformed tridentate into monodentate or bidentate, which deteriorated the toughness of hydrogels. Ye and co-workers constructed a dual-crosslinked cellulose hydrogel with the improved compressive strength (9.4 MPa) and tensile strength (1.7 MPa) by using two different crosslinkers namely ECH and polyethylene glycol diglycidyl ether [77].

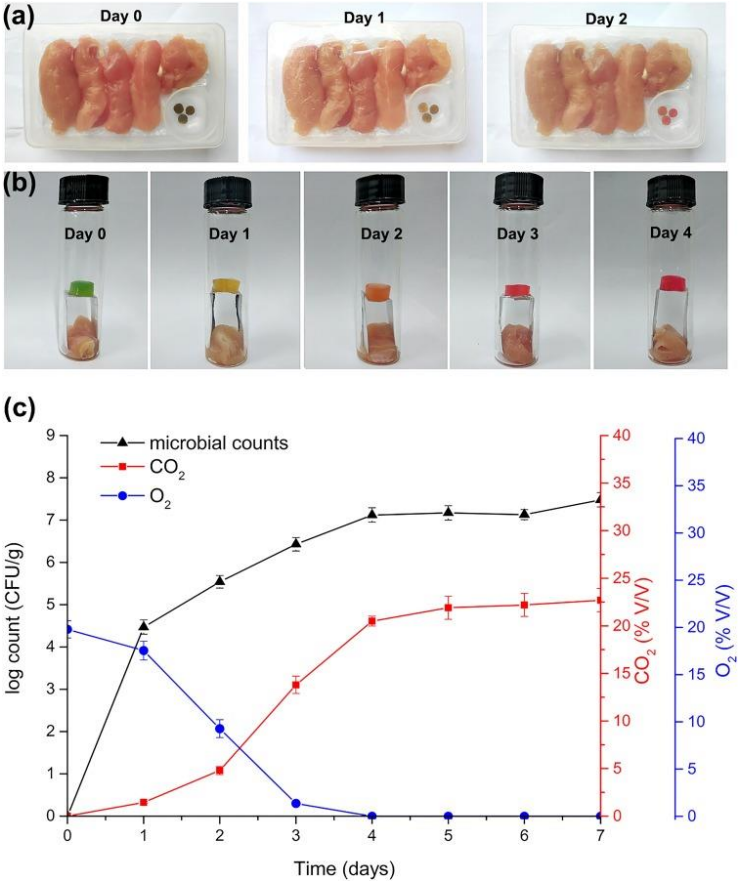

Figure 3. Color response of (a) cellulose-based hydrogels for intelligent food packaging application; (b) Changes in the color of hydrogel with chicken breast freshness in 4 days at 4 ℃; (c) Changes in CO_2, O_2, and microbial counts of chicken breast stored at 4 ℃ over 7 days [29].

Like regenerated cellulose films, cellulose hydrogels were also employed as a three dimensional matrix to fabricate antimicrobial or pH-responsive packaging by incorporating functional fillers. Compared to the dense structure of films, the porous network of cellulose hydrogels facilitates the transportation of bioactive compounds and enables quickly starting signaling pathways. The antimicrobial capacity was determined by the network density and concentration of active compounds [85].

Curcumin loaded in a bacterial cellulose hydrogel had a release rate of 76.99% in 6 h and significantly inhibited the growth of *S. aureus* [78]. The bactericidal effect was also found in the grapefruit seed extract-loaded cellulose hydrogel due to the existence of choline and ethanolamine [79]. Silver NPs were widely used to inhibit the growth of gram-negative bacteria (*P. aeruginosa*), gram-positive bacteria (*S. aureus*), and yeast (*C. albicans*) [83,84]. A novel strategy was proposed to produce hydrogels by using ECH to chemically crosslink bacterial cellulose and polyethyleneimine (PEI) in NaOH/urea solution [86]. The disruption of cell membranes by the polycationic nature of PEI caused a bactericidal activity against *S. aureus* and *E. coli*. The synergistic effect of several antimicrobial compounds was also investigated. For example, a hydrogel made from dialdehyde cellulose and chitosan was embedded with ZnO NPs and two bioactive compounds (quercetin and onion peel drug) to inhibit the growth of *S. aureus* and *T. rubrum* [81]. A comparable effect was found in the hydrogel filled with curcumin and ZnO NPs [82]. For the construction of intelligent packaging materials, as shown in Figure 3, two pH-responsive dyes (bromothymol blue and methyl red) were incorporated in a nanocellulose-based hydrogel as a food quality indicator to monitor the freshness of chicken breast [29]. The hydrogels showed a noticeable color change from green to red after three days due to the emission of CO_2 from microbial growth, indicating the detected microbial loads exceeded the acceptable limit for consumption (>6 log CFU/g). A tough, self-healing and pH-responsive hydrogel was produced from cellulose, polyvinyl alcohol, and borax, which could recover to the original state within 15 s after high shear strain and display a color change from yellow to red and purple when pH value increased from 5 to 12 [87].

Biodegradability and toxicity of cellulose-based materials

The biodegradability of cellulose may be affected by chemical modifications. Leppänen et al. reported that the biodegradation rate of regenerated cellulose depended on the degree of substitution (DS) [90]. Regenerated cellulose from ionic liquid was completely degraded after 4 weeks in a natural composting environment, while

cellulose acetate with DS 2.5 prevented the attachment of enzymes and showed no biodegradability in enzymatic hydrolysis and natural composting environment [90-91]. Haske-Cornelius et al. suggested to use esterase to degrade cellulose acetate with DS lower than 1.8, which could be enhanced by the synergistic effect of cellulase [92]. Cellulose nanocrystals are usually combined with regenerated cellulose to prepare all-cellulose materials; however, the incorporation of micro- and nanocellulose could lead to the decreased biodegradability [93,94]. It was because of that the increased crystallinity of all-cellulose materials hampered the water diffusion in polymer matrix and affected the disintegration kinetics [95,96]. Cui and co-workers demonstrated that okara cellulose hydrogels prepared by ECH crosslinking in LiOH/urea solution could completely decompose in 28 days [97]. The biodegradation process of crosslinked cellulose hydrogel was caused by the presence of microorganisms in soil, which initiated the cleavage of crosslinkages and disrupted covalent bonds, resulting in the decomposed network structure [98]. Regarding to the safety and toxicity of cellulose-based materials, various forms of cellulose including raw cellulose and regenerated cellulose, as well as it derivatives such as CMC, cellulose acetate, ethyl cellulose, etc., have been regarded as safe food substances according to the U.S. Department of Agriculture, and considered as the food additives by the FDA regulations [ref]. Nevertheless, the release and migration of functional additives from regenerated cellulose matrices should be carefully investigated. For instance, the minor release of Ag NPs didn't affect the survival rate of shrimp after an exposure of 24 h [100], but Ag NPs with high concentration (≥500 μg/mL) showed the significant cytotoxicity to Caco-2 and FHC human colon cells [101], and 100 ppm of Ag NPs remarkably decreased the vitality of hepatocellular carcinoma cells close to zero over 24 h [83].

Conclusions and outlook

Recent research focuses on optimizing dissolution conditions and developing new strategies to improve the properties of regenerated cellulose films and hydrogels, for example, the double-crosslinking method. Both cellulose films and hydrogels exhibit

potential to be applied in food packaging. Particularly, the regenerated films provide a solution for biodegradable wrapping materials, and the regenerated hydrogels can serve as a three dimensional matrix for absorption and quick signal response. Various organic and inorganic fillers can be incorporated in regenerated cellulose films and hydrogels to enable special functionalities, such as antimicrobial property and pH responsive color change. Generally speaking, regenerated cellulose materials are safe to be used for food packaging, but the release and migration of functional fillers should be evaluated. Future research in the following areas is required to promote their real applications:

1. The design of food packaging materials should be linked to a specific food product. Some research works just simply emphasized the improvement in mechanical and barrier properties, but did not mention the requirements for packaging materials. Especially for the development of active and intelligent packaging, various aspects such as initial status, storage condition and quality change of food product should be understood in advance.
2. The feasibility of new packaging materials should be considered. Processes should be available for mass production, and all the modifications and additives should be economically practical. The selection of cost-effective cellulose solvent system and the efficient recycle of solvent will help reduce the cost of regenerated cellulose materials.
3. The release and migration of functional fillers from regenerated cellulose packaging at different conditions (pH, temperature, solvent, food components, etc.) should be carefully studied. Although some active compounds are chemically bonded to cellulose matrices, the fate of these chemicals in the environment after the materials are disposed and degraded should be evaluated.

Conflict of interest statement

Nothing declared.

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