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Review

Sustainable polysaccharide and protein hydrogel-based packaging materials for food products: A review

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A R T I C L E I N F O

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A B S T R A C T

Sustainable food packaging is a necessary element to ensure the success of a food system, the accomplishment of which is weighed in terms of quality retention and ensured products safety. Irrespective of the raised environmental concerns regarding petroleum-based packaging materials, a sustainable analysis and a lab to land assessment should be a priority to eliminate similar fates of new material. Functionalized bio-based hydrogels are one of the smartest packaging inventions that are expected to revolutionize the food packaging industry. Although in this review, the focus relies on recent developments in the sustainable bio-based hydrogel packaging materials, natural biopolymers such as proteins and polysaccharides from which hydrogels could be obtained, the challenges encountered in hydrogel-based packaging materials and the future prospects of hydrogel-based food packaging materials are also discussed. Moreover, the need for 'Life Cycle Assessment' (LCA), stress on certifications and a sustainable waste management system is also suggested which can bring both food and packaging into the same recycling bins.

1. Introduction

The growing world population and changing lifestyles have resulted in the demand for more food production. This increasing demand for food production equally necessitates opting for better and improved food processing, logistics, and storage facilities to ensure food safety and food security. In addition, the commercialization and globalization of food supply make food production, preservation, and packaging to be more efficient to retain the quality of the products until it reaches the consumers. Nevertheless, food products are easily prone to microbial contaminations and can cause foodborne illness. Similarly, chemical contaminants accumulated in foods such as phytochemical residues and different metabolites from product decaying also invite associated risk factors. Therefore it is important to ensure hygienic handling and packaging of processed food products to retain their wholesomeness and overall quality. It is well known fact that the key function of food packaging materials is to maintain the food's hygienic quality and to preserve the shelf life throughout the transportation and storage. The conventional type of packaging materials routinely used are mainly petroleum-based which are linked with environmental pollution and remain biodegradable. Therefore it is important to develop environmentally friendly (bio-based) packaging materials to support consumers' health and promote a healthy environment. Globally, bio-based food packaging materials are gaining higher attraction due to their sustainable, storage-improved, and biodegradable properties [1,2]. A range of bio-based polymers have been explored by researchers across the globe to make food packaging to be much more effective [3–5]. Agricultural biomass that is high in lignocellulose content is the best resource for economic and environmental benefits. An estimated sum of 561–706 million tons of cellulose is expected to be extracted from major agricultural wastes from rice, wheat, corn, and bagasse [6]. Recent advances in materials science and engineering have developed high-performing superabsorbent hydrogels using raw materials from agricultural wastes [6]. Moreover, owing to their safety and eco-friendly properties, natural polymer-based hydrogels are preferred over synthetic ones [6–8].

Hydrogels obtained from agriculture-derived biopolymers offer good functional properties and present a three dimensional (3D) confirmation which allows their use as a safer packaging material in comparison with their synthetic counterpart [8]. Superabsorbent gels also known as hydrogels can be synthesized from the indigestible polysaccharides obtained from agri-food wastes and by-products [5,6,9,10]. Hydrogels

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have been extensively studied and implemented in various fields such as cosmetics, medical applications, water adsorbents, food agents, and as bio-composite fertilizers [11,12]. Hydrogels can hold more water molecules that contributes to their biocompatibility and are applied in tissue engineering applications. For example, cellulose-based hydrogels show excellent biocompatibility than conventional hydrogels that hold acrylamide monomers [13]. Application of hydrogel in food industry is still in infancy stage. The main feature of hydrogel required for packaging material application is its humidity control efficiency inside food packaging as exudates from the food products, especially fish and meat products and vegetables and fruits via transpiration, may release inside the package that could possibly allow microbial growth in food and thus spoil the quality [2]. Hydrogels can also offer opportunities for the design of efficient biopolymer packaging materials by including bioactive extracts with desirable properties such as antimicrobial for eliminating food-borne contaminants and antioxidant activity to remove intracellular ROS [2,9]. In this review, a brief overview of the current status of biopolymer-based hydrogels as packaging materials, the agricultural resources from which natural hydrogels could obtain and its applications are described. At the same time, we elucidate the limitations of hydrogel-based packaging materials, existing challenges and research gaps. We hope to provide novel research ideas and future prospects to the researchers who are interested in hydrogel-based packaging materials.

2. Hydrogels and its classification

In common terms, hydrogels are hydrophilic, colloidal substances made of 3D polymer networks [14]. Hydrogels can hold water molecules in the 3D network that swells and can also release water molecules and shrink, but they cannot dissolve in water. These characteristics makes them suitable for biomedicine, tissue engineering, agriculture, food packaging and bio-machine applications [14,15]. The 3D structure of hydrogels is able to withstand the pressure developed during swelling. The hydrophilic groups present in the polymers contribute to the water-absorbing ability of polymers, at the same time the nature and density of the polymer used for hydrogel preparation can significantly influence the water absorption capacity of hydrogels. Hydrogel are capable of controlling the water stored in its 3D structure. A hydrogel can absorb water more than 400 times its weight. When the surrounding environment is dry out it can release water and shrink. Hydrogels could dispense about 95 % of its stored water [16]. Like soft tissues, hydrogels can form a 3D environment and allow the diffusion of growth factors and nutrients. The term natural hydrogels define the natural polymer-based materials, e.g. proteins, lipids, and polysaccharides [2]. Hydrogels are classified based on their polymer type and, composition, physical appearance, ionic charge involved, types of cross-linking, stimulus responsiveness, and on biodegradability (Fig. 1) [17]. Hydrogels are classified into natural (e.g. chitosan, alginate), synthetic (polyacrylamide, polyethylene glycol), or as a combination of both (commonly referred to as hybrid hydrogels) based on the nature of polymeric matrix. Hybrid hydrogels are prepared by integrating one type of macromolecule with another so that the second polymer could strengthen the stability and functionality of the resultant hydrogels by fetching new functional groups or by manipulating the entire polymerization process. Hybrid hydrogels could be biologically active peptides, proteins, or micro/ nanostructures that are interconnected by chemical or physical crosslinking methods. These hybrid hydrogels can have demand in biomedical and functional food applications [18]. A novel chitosan-silica hybrid hydrogel has been studied for cell encapsulation and drug delivery application. These types of hydrogels are able to accommodate high protein loading efficiency [19]. With recent advances in material science and engineering technologies new product development technologies have been developed to recover and produce high-performing hydrogels from agricultural biomass suitable for agricultural and biomedical applications [20]. It is suggested that the

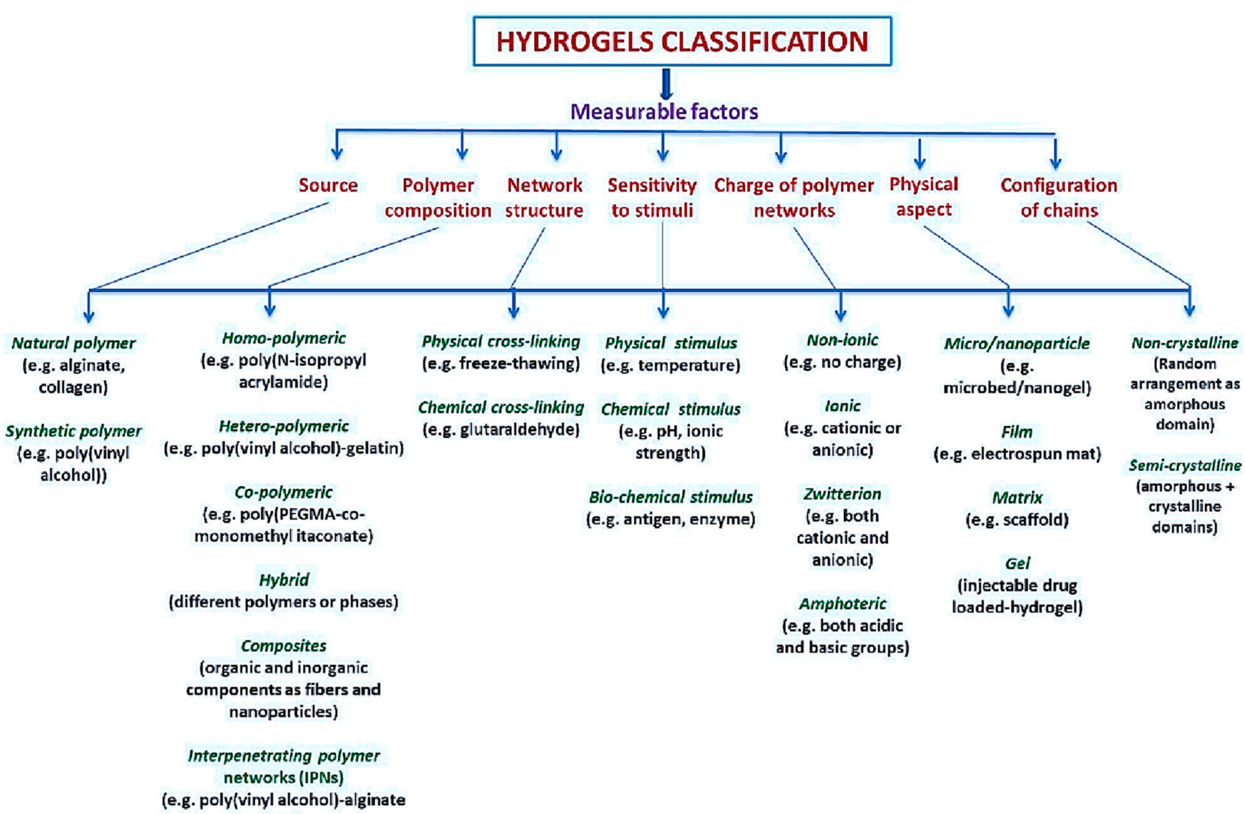

Fig. 1. Classification of hydrogels (Figure has been reproduced from Vasile et al. [17]).

polymer interactions can be strengthened by testing combinations of biopolymers with either bio-based or bio-dedegradable polymers with different structures and also via introducing charge interactions rather than hydrogen bonding. Some examples are; combinations of starch-cellulose fibers, chitosan-pectin, gelatin-pectin, zein-starch, methylcellulose-whey protein, gelatin-sodium alginate, and whey protein isolate-mesquite gum [21–23].

Furthermore, the second criterion of hydrogel classification is based on its cross-linking nature, i.e., physically cross-linked hydrogels and/or chemically cross-linked hydrogels [24]. Physically cross-linked hydrogels are reversible and unstable due to their monomers being connected by weaker interactions. For instance, cellulose-based hydrogels are preferred in biomedical applications owing to inter and intramolecular hydrogen bonding features of cellulose hydrogels that prevent them from any external binding. Whereas, chemically cross-linked (via free radical polymerization, Michael addition reaction, and esterification) hydrogels are irreversible and stable in forms [13]. For example, natural gum-based hydrogels are used to protect food products from moisture are modified by crosslinking with a suitable crosslinking agent that can strengthen the 3D structure of the gel [25,26]. One of the drawbacks of chemical cross-linking is the non-environmental friendly agents used for cross-linking. This can be overcome by the process of physical cross-linking via radiation processing methods such as gamma or electron beams. In a recent study, a combination of conformational structures of gelatin-derived hydrogels showed higher structural strength based on physical interactions and chemical cross-links [27].

Based on the polymer composition, hydrogels are categorized into homopolymeric hydrogel, copolymer hydrogel, and as multipolymer Interpenetrating polymeric hydrogel (IPN). Homopolymer hydrogel holds single species of monomer units which are uniform all over the hydrogel network. Whereas copolymer hydrogels are synthesized using two or more different monomer species arranged in blocks, random or alternating configurations along the polymeric network chain [15]. IPN is a special category of multipolymeric structures, formed by two independent cross-linked natural or/and synthetic polymer components [14].

Hydrogels are further categorized into different groups based on their electric charge, such as cationic/anionic, neutral/non-ionic, amphoteric having both basic and acidic groups, and zwitterionic with anionic and cationic groups in each structural repeating unit located on the cross-linked chains [14]. These electric charges have a high influence on the hydrogel water retention capacity. For example, cationic hydrogels have a good property of swelling at low pH [28].

Other features of hydrogels are their biodegradability, physical appearance, and stimulus responsiveness. For packaging applications, these features are rather important. Natural polymers are more easily biodegradable than synthetic ones which remain in the environment for many years. In addition, biodegradable hydrogels can degrade in an aqueous environment in a short time period and thus avoiding the requisite for removal after their use. Proteins and polysaccharide-based hydrogels have better biodegradability. Cellulose hydrogels produced from okra showed biodegradability in soil within 28 days leaving no cytotoxicity [29]. Depending on the applications and the technique of polymerization involved hydrogel appearance varies as matrix, film, or microsphere. This can be observed visually as well as with the help of different methods such as scanning electron microscopy, atomic force microscopy, etc. [29]. Similarly, hydrogels are fabricated based on their controllable response to external stimuli so that hydrogels can shrink or expand their structure. Moreover, hydrogels can perform volume transition changes in response to physical (temperature, magnetic/electric field, light, pressure, sound) and chemical stimuli (ionic strength, pH, solvent composition, and molecular species) [14].

In principle, good mechanical resistance, better moisture retention capacity, and swelling potential are the main features of a best-performing hydrogel and are having high demands in packaging applications, cosmetics, biosensors, biomedicines, and waste water treatment [30,31]. Chitosan-based hydrogels offer better packaging qualities for storing fresh fruits and meat products [32]. Similarly, responsive-stimuli chitosan hydrogels isolated from shrimp shell, which is cross-linked with different weights of 1,6-diisocyanatohexane in refluxing 1 % glacial acetic acid showed better flexibility in folding and storing meat and chicken pieces [33]. When dissolved in solvents the hydrogels act as colloidal gel with rheological properties and viscoelasticity, this again depends on the versatility of the synthesis and the nature of the precursor reactants [34,35]. Precise information on the classification of hydrogels necessary for packaging is documented in previously published articles [23,36].

3. Functional properties of hydrogels required for packaging applications

In the context of packaging applications, the functional properties to be considered for hydrogels are the water absorption capacity, mechanical strength, porosity, visco-elastic properties, thermal resistance, and morphological characteristics [36]. These functional properties are influenced by various factors such as the chemical structure of the polymers, composition, and preparation, degree of cross-linking between the polymers, and external stimuli responsiveness as discussed earlier in this review [2,37]. Below, we have discussed two main functional properties, i.e. water absorption capacity and mechanical strength of hydrogel-based packaging materials.

The core property of hydrogel is its water absorption capacity and swelling equilibrium, which can be manipulated according to the need of packaging application by selecting the polymer type and its polymerization process [23,38]. Hydrogels can absorb larger volumes of water from the environment and retain inside their polymeric matrix for a certain period under different conditions including mechanical pressure. The absorbed water retained in the polymer matrix prevents water loss and maintains swelling equilibrium, i.e. equilibrium water content (EWC) that can be studied by measuring the water uptake capacity of the gel at selected time intervals [39]. Hydrogels can retain water and swell in an aqueous solution (up to 1000-fold) of its dry weight. The hydration phenomena of hydrogels are an important feature in the modulation of drug release studies [40]. The water absorptions and the subsequent swelling properties of hydrogels are a multi-step process as described by Karoyo and Wilson [40]. In the first step, when the water molecules come in contact with polymers, they bound with the hydrophilic active sites of the hydrogel matrix and form primary-bound water which is strongly bound to the hydrogel. In the subsequent step, the hydrophobic sites come in and interact with the water molecules to form hydrophobically bound water (secondary). Thus, both primary and secondary bound water together form total bound water with intermediate properties. In the dry state, the hydrogel inside the matrix has high osmotic pressure due to the strongly linked ions in the polymer network, so that the excess amount of water can be easily absorbed and filled between the spaces in the polymeric network, which helps to reduce the osmotic pressure. So, an additional amount of water can be easily absorbed by the hydrogel and fills the space between the polymeric networks thus reducing the osmotic pressure. The swelling capacity of the hydrogel is influenced by the nature of the polymer based on its textural properties, the ambient temperature of the medium, and the interactions between polymer chains and water molecules. For example, swelling capacity varies between starch, chitosan, and cellulose due to the difference in their glycan structure, textural strength, and hydration properties [40,41].

Another relevant property to be taken into consideration for hydrogel-based packaging materials is their mechanical resistance. Conventional gels have lower mechanical strength and stiffness when subjected to external stress making them practically a weaker material for food packaging and tissue engineering application [42]. Because of the low mechanical properties of hydrogel limits its load-bearing applications. The mechanical properties of the hydrogel are further influenced by the polymer composition, particle size of the gel, and degree of cross-linking. Studies shows that the mechanical properties of hydrogel can be increased by reinforcing it with electro-spun fibers [42,43]. For food packaging applications hydrogel should fulfill the mechanical protection to the food product. In a research report, the mechanical strength of hydrogel was improved when using a combination of polyvinylpyrrolidone (PVP) with carboxymethylcellulose (CMC). From the stress-strain analysis, the authors found that tensile strength, strain at break as well as *E*- modulus were improved when they used CMC alone. It could be due to the cross-linking of the CMC chain with PVP upon drying. The temperature also influenced the cross-linking reaction and resulted in increasing the stiffness of CMC from 2940 to 3260 MPa at 25 °C [44]. Similar reports also indicate increased mechanical strength with the addition of nanocomposites, cross-linking agents, and pH-based regulation [45,46].

Besides the above-mentioned properties, hydrogels show different responses to physical, chemical, and biological stimuli. The response behavior of hydrogels is an appropriate feature for considering food packaging applications. Stimulus-responsive hydrogels are a good option in the delivery of active compounds, especially for antimicrobial agents or antioxidants. These hydrogels release the bioactive compounds under different controlled response conditions and hence improve food preservation [47]. Stimulus-responsive hydrogels can be applied in smart packaging systems so that the food quality alterations by microbiological activity or organoleptic quality changes can be monitored easily. Therefore, pH, temperature, and mechanical-responsive hydrogels are found to have relevant importance in developing food packaging applications [23]. There are stimulus-responsive smart hydrogels, also called shape memory hydrogels that could restore their original shapes with the help of a physical or chemical response code (for example, pH-stimulated DNA hydrogels) [48]. Other remarkable properties to be considered for hydrogel-based packaging materials are wettability which determines the level of hydrogel surface interaction with fluids and thermal resistance, which determines the thermal resistance of hydrogels at different temperatures [2]. We think that the challenging part of developing a packaging material is tuning its functional property. Therefore, it is advisable to prepare the packaging materials based on the food product to overcome the challenges to an extent.

4. Bio-polymers from agriculture wastes for hydrogel production

The growing population and urbanization has resulted in increased waste generation. The accumulation of waste not only creates environmental concerns but also leads to economic loss. As of the information published by UNEP Food Waste Index Report, about 931 million tonnes of food waste was generated in the year 2019, of that 61 % emanated from households and the rest from food services and retail sectors (UNEP Food Waste Index 2021). Hence by 2030, the European Union (EU) Commission has been expecting to meet the Sustainable Development Goals (SDGs) to halve per capita food waste at the retail consumer level (EEA report, 2020). Another thoughtful challenge is associated with agriculture intensification that generates 100 to 800 million tons of waste from rice, wheat, corn, and bagasse. Landfilling these wastes create carbon dioxide (CO_2) emissions. The best alternative is to reuse the waste materials for value-added products. Based on the revised waste legislation by the EU for the sustainable management of bio-waste, around 65 % of municipal waste will be recycled by 2035 [49]. Agri-wastes are rich in cellulose, lignin, and other polymers and their composition depends on the plant species [3]. Among the cell wall-derived polymers, cellulose is present in 35 to 65 % approximately (Sarkar et al., 2012). There is a higher amount of cellulose present in fruit pomace that can be fractioned by a mild acid-alkali condition. Cellulose content is higher in cucumber (16.13 %) and low in tomato pomace (8.6 %) [3]. It is estimated that, about 250 million tons of waste and by-products are generated per year in the EU, of which 30 to 50 % wastes produced from fruit and vegetable sector [3,50]. Hence, it is necessary to find alternative methods to avail and use these indigestible waste materials. The natural polymers obtained from agri-food biomass offer excellent properties to form hydrogels and are detailed below.

4.1. Polysaccharide-based hydrogels

4.1.1. Cellulose-based hydrogels

Cellulose has a chemical structure consisting of a linear homo-polysaccharide of β-d-gluco-pyranose units linked by β-1,4-linkages [51]. The glucose units of cellulose aggregate via hydrogen bonds as well as Van der Waal's force to form a thread-like crystalline cellulose microfibrils [52]. The strong hydrogen bond determines the durability and insolubility of cellulose in water and other organic solvents [6,52]. Cellulose is highly soluble in strong acidic or alkaline conditions. In nature, cellulose occurs as a linear polymer bound to hemicellulose and is cross-linked with water-impermeable and biodegradation-resistant lignin [53]. The extraction and separation of cellulose from non-cellulose components are performed via different chemical processes. The extracted cellulose fibers are further processed to develop cellulosic fines or nano-cellulose (i.e. cellulose nanocrystal and/or nano-fibrillated cellulose) for the production of superabsorbent hydrogels [6,38]. The potential features of cellulose include its renewability, biodegradability, biocompatibility, non-toxicity, and insolubility in most solvents [54]. Natural cellulose-based hydrogels are non-toxic and offer better wound-healing properties (mainly useful in drug delivery applications). They render excellent fluid absorbance ability without trailing the actual shape due to their weak interactions such as ionic interactions, H-bonding, and Van der Waals interactions as well as strong chemical interactions, i.e. via covalent bonds. This property has benefited diapers and feminine hygienic products [55].

A recent study shows the application of sugarcane bagasse (SCB) based pristine cellulose hydrogel for the delivery of grape pomace polyphenol drug [56]. The cellulose content in SCB is around 35–45 % and is considered a good source for cellulose-based hydrogel production. The study proved that the hydrogel network was an effective carrier for the adsorption of polyphenol drugs to yield maximum loading efficiency and also showed high biocompatibility and antibacterial activity in the polyphenol-loaded hydrogel [56]. A similar study was performed by Coelho et al. [57] using cellulose nanocrystals from grape pomace and used for the development of starch-based nanocomposite films. The incorporation of cellulose nanocrystals increased the tensile strength and Young's modulus in the films and indicates its application in food packaging [57]. In another work, the extraction of cellulose nanocrystals from grape pomace with deep eutectic solvents is reported [58]. They fabricated a self-healing nanocomposite hydrogel by radical polymerization of polyacrylic acid by adding cellulose nanocrystals as the renewable reinforcing agent, enhancing the dynamic crosslinking for self-healing and stable crosslinking for increasing mechanical strength and toughness in nanocomposite hydrogels [58]. Another study was performed by Ding et al. [59] using cellulose-based hydrogel-nano-metal composites from wheat straw for hydrogel generation from NaBH4 hydrolysis. Their study proves that wheat straw cellulose-based hydrogel-metal composites have good prospects in the applications of the catalytic direction [59] and also confirms its application for the growth of corn plants. The cellulose-based hydrogels on an application in the soil helped retain the water-holding capacity and significantly facilitated the controlled release of water into the soil under drought conditions [60]. Similarly, cellulose-based superabsorbent hydrogels were extracted from rice husk, oat husk, cotton, hemp, linen, wood fibers, tea, bamboo, and linseed [61–63].

Cellulose obtained from bio-based sources are used in various forms. Cellulose nano crystals (CNC), cellulose whiskers, cellulose fibers, methyl cellulose (MC), carboxymethyl cellulose (CMC), bacterial cellulose (BC), hydroxypropyl cellulose (HPC) and hydroxypropylmethyl cellulose (HPMC) are some of the derivatives [64]. Cellulose based hydrogels (CBH) are biocompatible, eco-friendly, non-toxic and a substitute to conventional packaging materials [65]. They can be created by reversible gel formations. The use of cellulose in its unmodified form has been used in different applications and has also been the subject of study, throughout the world. The hydroxyl group present in the cellulose structure gives it a new prospect in tailoring the cellulose based hydrogels made from ionic, covalent, complex, and hybrid cross links with other polymer materials [66]. The swelling properties of single CBH may result in the degradation and disintegration of the films. This problem can be solved by combining different biopolymers with cellulose where polymer-polymer interactions can supersede the polymer-water interactions [67].

4.1.2. Bacterial cellulose (BC) based hydrogels

BC can be produced from a variety of agro waste sources like apples [68–71], oat hulls [72], corncob and sugarcane bagasse [73], Semi-dry date fruit wastes and fig fruit wastes [74]. BC is synthesized by non-pathogenic microbial strains as a primary metabolic product as hydrogel in its natural form [75]. The ultra-fine BC structure has high tensile strength, high water holding capacity, high biocompatibility and high surface area to mass ratio [76]. Moreover, BC as a biomaterial are more popular for its versatility and can be used for extensive applied scientific applications. When the culture conditions are static with homogeneously distributed nutrition and oxygen, the bacteria produce BC nanofibers in all the directions with a diameter of 100 nm. As a result a randomly distributed fibrous layer is formed in the air-liquid interface where the bacteria lives and grow [77]. The BC fibrous layer in the radial-transverse plane (Fig. 2) also grows in thickness with time holding large amount of water. Interestingly, only about 10 % of the total 99 % water present in the BC structure behaves like free bulk water, the rest are tightly bound to the BC. Since, the life cycle of bacteria is completed within the BC network structure, each BC nanofiber is produced by an individual bacterium, and producing nanofiber is a continuous process; thus, resultant BC nanofibers are long. Moreover, the whole process can't be controlled at the micro level, thus weak links along a through-thickness direction (Fig. 2). With the formula of $(C_6H_{10}O_5)$ n, BC nanofibers compose of carbon, oxygen, and hydrogen, and BC nanofibers have few branches only [77]. BC hydrogel films lacks thermoplasticity, which can be introduced by mixing with other biopolymers, for its use in packaging [68,70,71].

4.1.3. CMC-based hydrogels

Although there are many reports on CMC-based hydrogels, the only formulation reporting its application in food packaging is with PVP [78]. The initial development of polyvinyl pyrrolidone-carboxymethyl cellulose (PVP-CMC) based hydrogels was done for adding thermo-responsiveness to PVP for its biomedical applications, especially tissue engineering and wound dressing [79]. The polymer films are smooth, flexible and transparent in nature [78]. The hydrogels have high water holding capacity and water holding properties. The water molecules are present inside this hydrogel as freezing bound water, the water that gets frozen inside the hydrogel structure with the temperature drop below freezing point. The volume phase transition temperature (VPTT) of PVP-CMC hydrogels has been discussed by Lu et.al [79], where they have stated that such phase changes for polymer gels is important for smart drug delivery from the thermodynamic point of view. When the temperature is above VPTT, then inter- and intra-hydrogen bonds breaks between water molecules and PVP/CMC causing dehydration and change of phase, while the hydrogen bonds between PVP and CMC

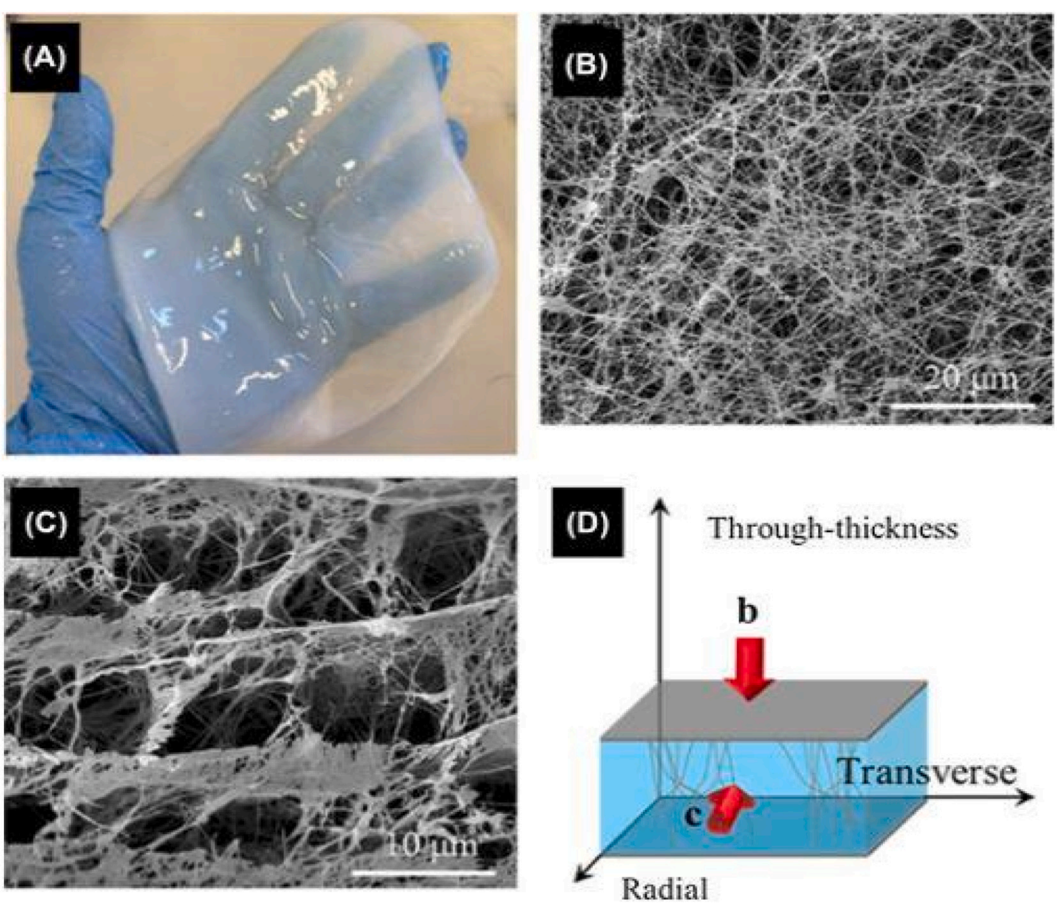

Fig. 2. (A) Bacterial cellulose hydrogel with a random distribution of fibers (B) in a plane of a fibrous layer and a multilayer structure (C) with some weak links along a through-thickness direction. (D) Schematic diagram of radial, transverse, and through-thickness directions (Image reproduced from Gao et al. [77]).

strengthens. More importantly, the balance between the hydrophobic and hydrophilic caused by the hydrogen bonds and hydrophobic moieties have created the stable reported PVP-CMC hydrogel structure [79]. With the increase in temperature, the balance breaks and the hydrophobic interactions increases causing the gel to shrink.

4.1.4. Guar gum-based hydrogels

Guar gum (GG) is a polysaccharide comprised of β-1, 4- mannose side linked to β-1, 6 galactoses at every second mannose, which can be abundantly found in nature. GG is a non-toxic, low cost and biodegradable polysaccharide promising for bioplastic preparation. Due to the presence of hydroxyl groups present in GG, which bonds with other structures by hydrogen bonds GG has an easy miscibility with other polymers. The hydroxyl groups present in GG also improves the water absorption capacity of the final polymer product [80]. GG has strong hydrogen bonding tendency in water with hydrated molecules and cellulose derivatives, but in cold water GG hydrates slowly [81]. This property can be applied to design hydrogel films that can prevent dehydration of the food products. In this case, hydrogels will absorb water slowly from the food in touch, and preserve the quality of food.

GG can be applied as an additive in pharmaceuticals, paper industry, textile industry, explosives, oil drilling and the cosmetic industry [82]. GG-based hydrogels have wide application in research and industry, mainly in the domain of water purification, medicine, agriculture and the food industry [81]. The regenerative quality of guar gum hydrogels makes it a better alternative for the treatment of wastewater. In fact, the non-ionic nature of GG makes it a suitable material for further modifications. Although edible food packaging materials are also developed from GG [83] the poor mechanical and barrier properties are constraints for its further use [84]. Thus the use of GG as filler to other cross-linked polymeric structures can be symbiotic to escalate the properties of the entire polymeric network.

4.1.5. Starch-based hydrogels

Starch is one of the abundant polysaccharides obtained from renewable sources such as cereals and tubers and has high demand in industrial-level applications due to its wide availability, low cost, edibility, and biodegradability [85,86]. In structure, starch is composed of amylose (20–25 %) and amylopectin (75–80 %) representing about 98–99 % dry weight of starch. Amylopectin is a high molecular weight (107–109 g/mol) branched linear chains of (1,4)-α-linked D-glucosyl units connected by (1,6)-α-linkages whereas, amylose has linear longer chains and is a minor component with low molecular weight (105–106 g/mol) [86]. Depending on the source of starch, the molecular structure, composition, shape, and size of the starch granules can be changed which can influence its processing features [87]. The crystallinity (varying from 20 to 40 %), wide availability, and cost of production of starch made it an attractive material for technological application. Though starch is inherently a hydrophilic material, the physical and mechanical properties of it mainly depend on the water content. For hydrogel applications, starch cannot be utilized in its native form as the three-dimensional stability, mechanical properties, and thermal stability are low. Therefore, starch requires to be modified by chemical or mechanical interactions [88]. Starch can form suspensions with good film-forming ability and offer several advantages as they are tasteless, colorless, and odorless. They offer good oxygen barrier properties, nutritional value, and edibility [89]. A recent study reported starch/clay nanocomposite-based biodegradable packaging materials with good mechanical properties [90]. A cross-linked cassava starch with epichlorohydrin polymer used for food packaging showed enhanced physicochemical, thermal, and retrogradation properties [91]. Better packaging properties were observed with starch-based films incorporated with citric pectin and feijoa peel (*Acca sellowiana*) flour for apple packaging [92]. Similarly, starch-based films were accounted properly for the packaging of cakes (cassava starch), salmon (cowpea starch), chicken breast (rye starch), and cherry tomato (pea starch) [93–95].

4.1.6. Chitosan-based hydrogels

Chitosan is a polysaccharide and a natural cationic copolymer that presents a lot of interest in hydrogels [96]. Chitosan is present in the natural state as an ingredient in the marine crustaceans and cell walls of certain fungi (e.g. *Fusarium solani, Lentinus edodes*) [97,98]. Chitosan is one of the widely used unique polymers with cationic characteristics, which enables it to bind with negatively charged materials (eg. anionic polymers, nucleic acids, and enzymes) [99]. Chitosan is a biodegradable, non-toxic, and hydrophilic biopolymer of glucosamine and *N*-acetyl glucosamine connected by (1–4) linkage [100,101]. Chitosan-based hydrogels are efficiently synthesized by using its $-NH_2$ and –OH functional groups for graft polymerization reaction and are well studied for food packaging applications [102]. Researchers have fabricated ultra-thin non-woven mats for food packaging applications using nanofiber technology [103]. Due to the excellent physical and mechanical properties chitosan nanofibers are excellent materials for packaging applications. Moreover, by the antibacterial and antioxidant potentials of chitosan polymer, the shelf life of delicate food products can be increased throughout their distribution and storage. To prolong the quality of unprocessed red meat chitosan nanofibers are developed as the inner part of multilayer packaging [96,104]. A chitosan-based hydrogel film prepared with the addition of β-acids and silicon through covalent/non-covalent bond interaction showed enhanced anti-ultraviolet ability of the film as well as improved antibacterial activity [103].

4.2. Protein based hydrogels

4.2.1. Gelatin-based hydrogel

Gelatin-based biopolymers are prepared by the partial hydrolysis of collagen from bovine and porcine skin, bones, and fish scales [105]. Gelatin is a macromolecule composed of bioactive polypeptides. Glycine, proline, and 4-hydroxy proline residues contribute to 60 % of the total amino acids in it [105,106]. It is a translucent, colorless, and nearly tasteless powder. Depending on the method of processing via acidic or alkaline pre-treatments gelatin has been categorized into type A and type B. Gelatin has extensive applications in the food, cosmetics, and pharmaceutical industry [107]. It has excellent film-forming properties due to its linear structure and limited monomer composition. Gelatin provides, elasticity, stability, and viscosity, therefore used as an emulsifier, biodegradable film-forming material, colloid stabilizer, foaming agent, micro-encapsulating agent, etc. Though gelatin has good mechanical, functional, and barrier properties, are sensitive to moisture and show poor barrier properties against water vapor [105]. Studies have proven the addition of plasticizers to improve the water sensitivity of gelatin films [108]. The addition of antimicrobial/bioactive compounds/nanoparticles in gelatin has significantly increased the antimicrobial activity against foodborne pathogens bacteria and showed strong antioxidant activity along with improvement of the UV protection, water vapor barrier, and mechanical properties [109–111]. An antibacterial hydrogel based on gelatin, chitosan, and 3-phenyllactic acid confirmed the extended shelf-life of chilled chicken [112]. A smart hydrogel was synthesized by Alpaslan and co-authors to be utilized as a multi-functional food packaging material from N, N dimethyl acrylamide (DMAAm), gelatin, and citric acid (CA) red apple peel extract (RApE) [113].

5. Recent advances in hydrogel applications in the food packaging system

Hydrogels can be a potential and smart source for packaging applications [2,23]. Due to their eco-friendly nature, bio-based materials are gaining interest in packaging material research at the same time seeking attention from the public. The success of hydrogel applications in biomedical and related fields has gained enough attention from researchers and encouraged researchers to expand the applications in other domains. Biodegradable food packaging from hydrogels is one of the demanding and progressing areas of research, especially in the field of active and intelligent packaging of food products. Active packaging performs functions that could improve food quality and safety, whereas intelligent packaging provides information on the quality and environmental changes inside the packaging with the help of sensors or indicators. The smart packaging systems are intended to minimize food losses, and also to provide information on the quality and safety of the food, so that food quality will be preserved throughout distribution until use [114]. A diverse range of hydrogel-based hygiene products, wound care, and tissue engineering products are out there in the market but hydrogel-based food packaging materials are not available yet. It is expected that soon we would be able to see smart hydrogel packaging materials with improved functional properties that ensure better food quality. One of the main features of packaging material is moisture control inside the food packaging. In that aspect, hydrogels can absorb excess moisture from the food package and thus slow down the growth of food-spoiling microbes on the food, especially ready-to-eat meals and hygroscopic food products [115]. The properties expected from functional hydrogel-based packaging materials are; renewability, cost of production, biodegradability, mechanical strength, transparency, breathability, and high absorption property. Numerous studies report the application of hydrogels as biodegradable packaging materials, but the durability, mechanical strength, and barrier properties remain poor as compared to synthetic packaging [110].

In the food packaging system itself, hydrogel-based absorbent pads that have water-removal features are of interest as these hydrogel pads can absorb the exudates from meat and vegetables and preserve the internal moisture of the package low. Therefore the rate of food spoilage can be reduced. The main reason for food spoilage inside the package is due to exudates that are generally released via the transpiration of fruits and vegetables, or the permeation of water vapor due to environmental changes or physicochemical changes in the packaged food [116]. An efficient absorbent pad is designed in a way that the exudates released from the food material should be held within the 3D structure of the absorbent pad and maintain a good visual presentation and sensory attributes for the food material. The absorbent pads also help keep the structural integrity of packages along with increasing the shelf life of the food material [2]. Thus, hydrogels can help to reduce the growth of food-damaging microbes such as moulds, and bacteria on food [117]. Hydrogels can absorb moisture up to 100 % of their weight, whereas super adsorbent type hydrogels are capable of absorbing over 100 % of their dry weight [117]. The absorbent pad used in packaging is made of two permeable layers and an inner layer holding the absorbent material. The external layers are generally cellulose-based or thermoplastic material with little pores, which is impermeable and protects the absorbent materials and their direct contact with the packaged food. Through the pores of the external layer, the exudates could flow to the absorbent hydrogels [2,118]. The main requirement of absorbent material for packaging application is the capacity to retain the exudate in the three-dimensional structure; good and clear visual presentation of the packed food, sensory attributes of the packed food, structural integrity of the food packaging system, increased shelf life of the packed food products and finally antimicrobial properties [118,119]. Like absorbent pads antimicrobial sachets or pouches are also placed inside the food packaging are one kind of active food packaging (Fig. 3). The antimicrobial compounds or antioxidants present in the hydrogels could defend the microbial activity in food [118,120]. An absorbent flexible gel sheet fabricated by researchers from the United States also shows similar characteristics. The gel sheets can absorb large quantities of aqueous fluids and can be folded, rolled up, and trim down to desirable sizes. This could be an excellent alternative to water-absorbent cloths or paper towels. The authors claim that these gel sheets could be used in hospitals, and labs to clean up biological fluids and also in the home kitchen to clean spills [121].

Hydrogels in food packaging are designed in the form of films or absorbent pads. Also water activated release of the antimicrobials can be coupled with hydrogels for food packaging. Hydrogel film production includes the casting of hydrogel without the addition of chemical cross-linking agents. Hydrogels developed by this method are transparent, flexible, biodegradable, and highly permeable. Cellulose-based hydrogels are of excellent material for food packaging application as it has high biocompatibility, mechanical properties, and biodegradability. Cellulose hydrogels obtained from plant or bacterial-based cellulose are applied as a thin layer onto the polymers to improve the visual appearance, transparency, biodegradability, and resistance to environmental agents. Cellulose-based hydrogels can perform excellent water absorption properties, swelling, and stimuli responsiveness. Therefore, it can be used in smart and intelligent packaging [36]. When using cellulose with polyethylene glycol, polyvinyl alcohol, gelatin, etc., they showed better packaging properties [122,123]. A detailed overview of fabricating regenerated cellulose-based films and hydrogels for food applications is documented [30,36].

Fig. 3. An illustration of hydrogel application as absorbent pads and antimicrobial/antioxidant pouches for food products.

The functional properties of hydrogels could be further enhanced by embedding nanoparticles, bioactive compounds or antimicrobial agents could further improve its properties [65]. Wang and Rhim [124] developed an agar/alginate/collagen ternary blend incorporated with silver nanoparticles (AgNPs) and grapefruit seed extract (GSE) for food packaging applications. They observed that hydrogel films incorporated with AgNPs exerted stronger biological activity against pathogenic bacteria such as *Listeria monocytogenes* and *Escherichia coli*. It is a promising observation to develop antimicrobial films for food packaging applications [124]. Similar observations were reported by other researchers using chitosan, gelatin, and carrageenan-based hydrogels incorporated with nanoparticles like AgNPs (Table 1). Studies showed that nanoparticle-incorporated hydrogels have high antimicrobial potential and also prevent fogging and greening of vegetables caused by post-harvest respiration [124,125].

Several polysaccharide-based hydrogels are proven to be useful in the formulation of food flavor carrier systems, and can bring newer developments in the food processing sector. For example, pectin, starch, cellulose, alginate, and carrageenan has been already in use for packaging film application. The main advantage of polysaccharide-based hydrogels have been noticed is the properties of being odor free, colorless, biologically absorbable, immune to oxygen, as well as semi-permeable towards CO_2 has potential applications in food industries [126–128]. Polymers like pectin and xanthan gum help in developing complexes suitable for the generation of gels as sheets, membranes, and coatings on the other hand, acacia gum produces round-shaped structures like nanocapsules through which active substances will be encapsulated and get integrated inside the packaging films [128].

Another type of hydrogel developed for food packaging applications is colorimetric hydrogel as an indicator for food spoilage. These hydrogels are worked based on stimuli response mechanism i.e., significant color shifts could be detected based on the change of medium

Table 1
Recent examples of natural polymer-based hydrogels used for food packaging applications.

Biomass (agri-food wastes)	Polymer obtained for hydrogel	Food material	Properties and application	Reference
Sugarcane bagasse	Carboxylated nanocellulose	Chicken breast	Colorimetric freshness indicator	[162]
Commercial grade	Gelatin, chitosan, and 3-phenyllactic acid	Chicken meat	Improved the antibacterial performance and prolong the shelf life	[112]
Sugarcane bagasse	Nanocellulose/nisin hybrid film	Ham	Antimicrobial performance against L. *monocytogenes,* Good light transmission, high tensile strength, low oxygen permeability and low water vapor transmission	[163]
Cotton linter pulp	Cellulose	NA	Cellulose film presents outstanding optical, thermal and mechanical properties	[164]
Rice husk, sugarcane bagasse	Carboxymethyl cellulose	NA	Better opacity, moisture content, tensile strength and solubility of the packaging film	[165]
Waste paper	Cellulose	NA	High tensile strength, antimicrobial compounds were easily loaded into cellulose film to make active packaging.	[166]
Eucalyptus sawdust	Carboxymethylcellulose	Banana	Humidity/ethylene absorbent in banana packaging and increased shelf life	[167]
Corn stalk	Regenerated cellulose films	NA	High tensile strength, ability to block ultraviolet rays and inhibitory effect on *E. coli* and *S. aureus*	[103]
Commercial grade	Bacterial Cellulose, guar gum, polyvinyl pyrrolidone, carboxymethyl cellulose	Blueberries	Improvement in elastic and load-bearing capacity, biodegradability, better barrier and hydrophobic properties	[70]
Bovine gelatin	Gelatin+ dialdehyde carboxymethyl cellulose		Increased tensile strength (TS) and thermal stability, decreased elongation at break, very transparent and excellent barrier properties against UV light	[168]
Commercial source	Carboxymethyl cellulose (CMC), polyvinyl alcohol (PVOH)	Chicken meat	Mechanical, permeability and antimicrobial properties were improved	[169]
Softwood kraft pulp	Cellulose nanofibers (TOCN)/cationic guar gum (CGG)		Improved the mechanical and barrier properties of the paper; provided the oil resistance, but maintained the mooncake's freshness	[133]
Shrimp shell	Chitosan	Chicken meat	High flexibility biodegradable film	[33]
Commercial grade	Carboxymethylcellulose	Tart cherries	Preservation and delivery of tart cherry anthocyanins and polyphenols	[119]
Wheat straw	TEMPO-oxidized nanofibrillated cellulose	Mango	Thermo/pH- -responsive preservative delivery in climacteric fruit packaging	[170]
Commercial source	Konjac glucomannan (KGM)/carboxymethyl cellulose	Tilapia	A pH-intelligent response fish packaging film with antibacterial properties and improved mechanical strength	[171]
Carp skin gelatin	Furcellaran,carboxymethyl cellulose, gelatin hydrolysate	Cherry tomatoes	Highest thermal stability of film, antibacterial activity as well as the increased shelf life of cherry tomato	[172]
Grape pomace	Celllulose and cellulose nanocrystals. NA		Increased the tensile strength and Young's modulus in the films, improved the mechanical properties and decreased water vapor permeability	[57]
Grape pomace	Cellulose Nanocrystals	NA	Successfully fabricated of self-healing nanocomposite hydrogels	[58]
Feijoa peel flour	Starch	Apple	Biodegradable packaging with antimicrobial and antioxidant properties was developed	[92]
Cassava roots	Cassava starch	NA	Increased tensile strength, lower water absorption, biodegradability, non-toxicity, low cost, and good thermal stability	[173]
Cowpea	Cowpea starch, maqui berry extract	Salmon	Used to develop antioxidant rich packaging film	[94]
Rye	Rye Starch, Containing Rosehip Extract	Chicken Breast	An active packaging material to retard lipid oxidation in foods	[174]
Commercial grade	Cellulose nanofiber, clove bud natural essential oil		Gelatin/agar-based films with improved physical and functional properties	[111]
Commercial grade	Agar/alginate/collagen with AgNPs and grapefruit seed extract		Antifogging packaging films for highly respiring fresh agriculture produce with high antimicrobial activity	[124]

pH. Based on this concept, a novel hydrogel was formulated by Alpaslan [129] from *N*, *N*-dimethyl acrylamide (DMAAm), gelatin, citric acid (CA), and basilicum extract (BE). The aim of the study was to create an advanced packaging hydrogel that could detect food spoilage and thus be applied as an instant food quality monitoring device [129]. A cellulose nanofiber (CNF)-based color indicator film was developed by adding shikonin extracted from dried roots of gromwell. The developed film showed significant color change based on the pH change. The developed CNF/shikonin composite film showed improved mechanical properties and remarkable UV-blocking properties [130]. In one of the studies, the authors [131] present the chitosan-based hydrogels and their physical responses to different pH levels. They revealed the electromagnetic characterization of the chitosan-based hydrogel in ultra-wideband (UWB) frequencies, and its use as a pH sensor with chipless radio frequency identification (RFID) resonators. Chipless RFID sensors are relatively low-cost and offer enormous design flexibility in diversified applications such as cold chain, and smart packaging [131] (Fig. 4).

Breathable hydrogels are another category of hydrogels formulated for food packaging applications, mainly for fresh-cut fruits and vegetables storage. Roy et al. [78] reported an approach to develop a breathable hydrogel film for food packaging. They used poly-vinyl-pyrrolidone, carboxymethyl cellulose, and agar. It is also transparent, flexible, and completely biodegradable. A similar finding was reported by Saha et al. [132]. The quality of grapes was monitored by the changes in the resistances of modified multi-walled carbon nanotube sensors. Compared to conventional food packages PVP-CMC-based hydrogel showed more breathability.

Paper-based packaging material has been developed by Dai et al. [133] using 2,2,6,6-tetramethylpiperidine-1-oxyl radical -oxidized cellulose nanofibers (TOCN)/cationic guar gum (CGG) hydrogel film. The authors modified the conventional paper with TOCN/CGG hydrogel film in layers on paper so that the mechanical properties, oil resistance, and barrier properties of the paper improved along with maintaining the freshness of the food materials inside the packing. The modified paper showed a tensile strength of 34.03 MPa and a burst strength of 510 kPa which were 26.78 MPa and 388 kPa with unmodified paper, respectively. The authors checked the packaging performance for the mooncake packaging test and the results showed oil resistance and freshness of the mooncake [133].

6. Conventional vs bio-resourced packaging materials

To date, conventional materials like petrochemical-based plastics, metal, glass, and paper have been used in food packaging. The

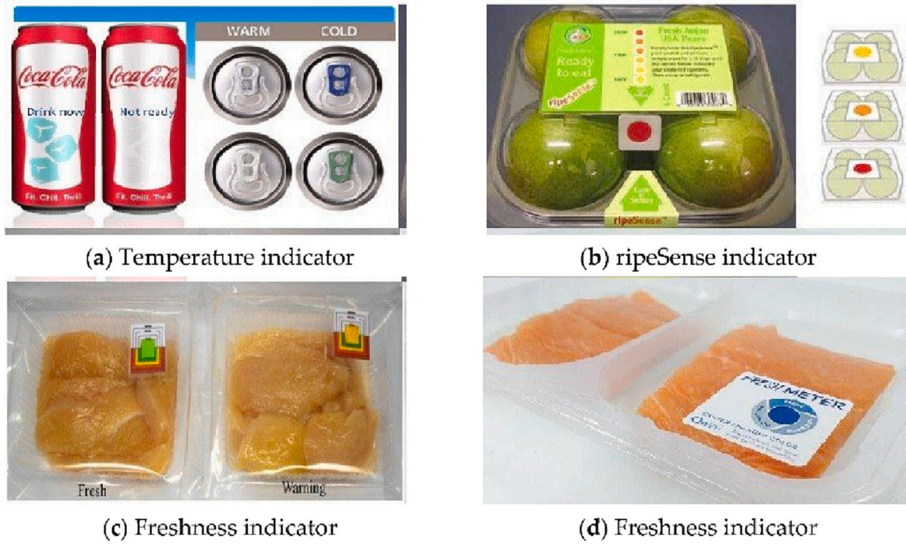

Fig. 4. Examples of intelligent packaging systems for food (Images has been reproduced from Nešić et al. [127]) (a) Temperature indicator; (b) ripeSense indicator; (c) Freshness indicator (d) Freshness indicator.

petroleum-based plastics (for example, polyvinylchloride, polyethylene terephthalate, polyethylene, polyamide, polystyrene, etc.) perform excellent packaging features such as gas and water barrier properties, tensile strength, and flexibility. After all, they are low-cost materials and easy to produce. Despite these benefits, petrochemical-based packaging materials result in a lot of environmental issues, especially greenhouse gas emissions due to incinerating such materials and also affecting the food chain thus causing a threat to the natural ecosystem. Therefore, eco-friendly solutions for food packaging are the mainstream study now. The above-mentioned biomaterials have remarkable properties that could be beneficial for packaging applications.

Though several studies have been performed with natural-based hydrogel in food packaging applications, the development of completely sustainable and biodegradable materials may take a longer time for availability in the market. One of the main drawbacks of hydrogels is they are not always compatible with all packaging materials and easily susceptible to chemical breakdown. At present, acrylates and their derivate monomers have often been used as raw materials to form hydrogels [86]. A copolymerization between acrylate monomers and natural polysaccharides would be an option to enhance the properties of hydrogel [134]. Another possibility is the use of nano-materials to improve hydrogel performance [2]. Different nano-materials such as cellulose nano-crystals, carbon nano-tubes, cellulose nano-whiskers, chitin whiskers, montmorillonite, micro-fibrillated cellulose, and starch nanocrystals have already been applied in food packaging [135]. Extensive research has to be conducted to study the shelf-life extension of food products by hydrogel-based packaging material from natural polymers, as this approach is promising and can add value to packaged foods due to its biocompatible and non-toxic nature. The present-day food packaging system, which is based on sachet technologies, uses materials from non-biodegradable sources and poses health risks and environmental concerns [2,134]. Due to the global climate change crisis and the concerns over health issues, and providing a sustainable environment for the future generation, there is a high increase in consumer acceptance of biodegradable food packaging [136].

7. Sustainability assessment

Due to the growing interest in environmental awareness and the sustainable use of natural resources, it is important to assess the environmental impact of materials used in packaging applications. Here comes the importance of Life Cycle Assessment or LCA of different biopolymers that are used for sustainable packaging materials to have information on the environmental impacts caused by them. LCA evaluates the potential negative environmental impacts and the resources used for overall product life cycle which includes raw material extraction to product design and production process, use phases and finally, waste management processes [137]. Though packaging sectors are using LCA assessment tools to examine the environmental footprint by packaging material, more solid data is needed for the packaging materials and the process used to convert them into packaging. LCA is mainly developed to study the environmental impact of a product throughout its life cycle starting from the extraction of the raw material from the earth and its return back to the earth as a waste product. The LCA gives an overview of life cycle inventory and related environmental consequences through impact assessment methodologies [138]. Not many studies have been performed on the life cycle of biopolymer-based hydrogel as packaging materials. The available information is mainly based on the biopolymers from feedstock. Moreover, previous LCA and environmental assessment methods were primarily focused on global warming potential and fossil fuel depletion impact categories as biopolymers could defeat the side effects caused by plastic packaging materials [139]. At the same time few studies draw attention to the negative impact of biopolymers. Though biopolymers are considered as an environmental friendly alternative to petrochemical polymers due to the renewable agricultural feedstock used to produce them but the farming practises used to grow the feedstock often because environmental burden and the energy spent for the production could be higher compared to fossil based polymers [138,140]. Researchers have performed LCA studies on plastics obtained from fossil fuels, bio plastics, bio-degradable and non-biodegradable plastics [140,141]. But the overall LCA assessment shows that the substitution of fossil-based polymers with bio-polymers does not always promise sustainability, instead waste management being one of the key elements to sustainable development. Recycling the packaging materials would be more promising than biodegradation [140]. One of the study's authors from Brazil analysed the energy and environmental performance of manufacturing cassava starch-based film. Their study revealed that the main energy and environmental impacts were mainly due to cassava cultivation, production of additives (fossil glycerine and ethanol additives) used in the producing the film and electricity consumption during the manufacturing of the film (by casting method) [142]. Though hydrogels meet the basic principles of green chemistry aspects such as biodegradability, biocompatibility and renewability they are considered safe and eco-friendly. There are available information on extraction of biopolymers, casting, and additives used and biodegradability data, but information on LCA of hydrogels is still scarce.

A recent review focused on the use of life cycle assessment (LCA) for evaluating the sustainability of thermoplastics, wherein the authors made a comparison of the LCA assessment of thermoplastic with other packaging materials. The authors disclose that though the bio-based biodegradable packaging materials could overcome the limitations of conventional plastics, the environmental impact caused by the agricultural feedstock needed for obtaining bio-based polymers is high. Moreover, the GHG emission rate that occurs as a result of biodegradation can also contribute to environmental impact. In addition, setting up appropriate infrastructure and the expenses required for composting process can cause additional expenses. One alternative the authors suggest is by using improved agricultural practices, such as using renewable electricity for cultivation, using feedstock that requires minimal energy, nutrients, and fertilizers, and employing superior agricultural nutrient management practices [140].

Another study by Gujjala and Won [143], reports the LCA of lignin hydrogels that has promising applications in environmental remediation. They studied process design and techno-economic analysis. From the study, they estimated that the hydrogel process itself resulted in emitting 2.8 kg CO_2 equivalent and 1.1 kg oil equivalent/kg lignin hydrogel. Heat integration during the process could lead to 78.9 % in heating and 83.4 % savings in cooling energy requirements. So that the authors suggest that the strategy they used for developing lignin-based hydrogel could be integrated with conventional biorefineries to develop sustainable alternatives for conventional scenarios [143]. The development of natural-based hydrogels requires adequate selection and synthesis of the biomaterial, water and energy for the casting process, solvents included, cross-linking agents, and additives such as crystalline nanofibers and nanocomposite materials. For example, the life cycle of cellulose-based hydrogels involves the processes that start from the separation of cellulose from other compounds present in biomass, such as lignin and hemicelluloses, the solvent used for its production, cross-linking agents and additives used, energy consumption for hydrogel casting process and biodegradation. All these processes should be considered in hydrogel LCA studies. This could provide opportunities to develop ideas for smart processing methods, identify critical inputs, the space for improvement, and intelligent end-of-life strategies for the product [144]. Therefore, a holistic approach of assessing the sustainability of a product, its socio-economic aspects in addition to the environmental impact is necessary before designing the biopolymer-based hydrogel and its competence with conventional packaging materials.

8. 3D-printed hydrogels and food packaging

3D printing is an emerging technology showing wide prospects not only in the food packaging industry but also in different fields. It has a better advantage than conventional processing methods to fabricate hydrogels with complex structures. 3D printing offers precise structural control over the fabrication process, facile operating methods, customizable characteristics of printed architectures, fineness in the structure of fabricated material, and better cost-effectiveness [145]. Though hydrogels have weak mechanical properties, 3D-printed hydrogels gain momentum and are expected to approach mainstream adoption and be explicitly used in diverse fields, especially biomedical fields [145]. 3D printing technology has been preferred nowadays in intelligent food packaging systems and intelligent packaging uses smart components such as food spoilage indicators, sensors, electronic tags, etc. to report the freshness of the food materials in the food packages, changes in the food storage conditions and track the products from point of origin to use [146]. With 3D printing, it is possible to fabricate intelligent packaging with highly sensitive, self-indicating, and multifunctional components using biocompatible nontoxic materials that are more economical than conventional fabrication methods. These advantages of 3D printing in intelligent packaging systems would help minimize food waste, food fraud, managing food quality control and package integrity control [146,147].

3D printing-based intelligent packaging is one of the main streams of technology being explored because of the assurance of food quality, and safety and as a proof of food authenticity. In a recent study the suitability of kappa-carrageenan (κC) and agar thiamine-loaded hydrogels for hot extrusion 3D printing was performed and compared their casting structures as well as release properties. Compared to conventional cast cylinders, the 3D printed cylinders release higher fractions of hydrogel improving the physical properties and highlighting the possibility of the formation of food-related structures under 3D printing technology [148]. A similar study was performed with a formulation of bio-ink combining pectin with sodium carboxymethyl cellulose (CMC) incorporated with zinc oxide nanoparticles (ZnONP) and compared their physicochemical and functional properties under the solution casting method as well as 3D printing. 3D printed scaffold exhibited a more compact and organized structure with better mechanical properties, moisture barrier, and antimicrobial activity than the solution casting method [149]. It is expected that 3D printing technology could contribute to food packaging applications by producing hydrogels with better functional features, especially to create food quality sensors, indicators, or electronic tags. Moreover, with the 3D components, full 3D spatial and optical sensing is possible as also the detection of microorganisms and low-power remote sensing of the product location.

9. Challenges and research gap

At the European level, 92 million tons of plastic packaging are produced which could increase to 1124 million tons of plastic material by 2050 if the use of plastic materials goes into the current flow [150]. Therefore it is important to find alternative solutions for developing sustainable packaging materials. Due to their wide spectrum property, hydrogels are applied in various disciplines including food packaging applications. Though hydrogels are one of the intelligent solutions to the use of petroleum–based polymeric materials, their shelf-life, cost of production, mechanical strength, and transparency are of real concern when it comes to commercial scale productions. Any attempts that can improve such properties are worth the attention. Nanotechnology offers the possibility to improve hydrogel performances. Hybrid hydrogels like nanocomposite hydrogels containing nanoparticles are proven to have better mechanical, thermal and water resistance properties and thus could overcome the main barriers in food packaging applications [151]. Similarly, the addition of cellulose nano-fibrils from cotton fibers to a polymer matrix based on chitosan-graft-poly acrylic acid significantly improved swelling capacity [152]. Numerous nanomaterials are already applied or are under trial to improve the properties of hydrogels and their applications in the biomedical and food packaging sector; carbon nanotubes, cellulose nano-whiskers, chitin whiskers, montmorillonite, micro-fibrillated cellulose, and starch nanocrystals are some examples [2,135]. After all, the nano-packaging effect on food depends on the composition of the nanomaterial that is involved in the packaging film. Though using nanotechnologies for improving packaging materials applications is likely to be expensive, researchers world-over are trying to develop new concepts to develop commercially successful products. Similarly, developing smart packaging systems could reduce food loss to some extent. Smart packaging is one of the novel concepts that involves the idea of both active and intelligent packaging. In the smart packaging concept, the protection of food from spoilage is promised and the quality is assured to meet consumer demand including its disposal [153].

Perforated packaging is another category where hydrogels could be applied. Perforated packages are mainly used for perishable food products to extend their shelf-life [154]. In the perforated packaging micro- or macro-sized holes are added so that it facilitates the smooth flow of oxygen and carbon dioxide gases from the food products produced by respiration process. The pores could also help in good circulation of cold air during transportation and storage [155]. In this context, the cooling efficiency of the hydrogels for the horticultural produces are scopes for future research. For example, perishable fruits and vegetables like cucumber, grape, tomato, avocado, berries etc. are mainly stored in perforated packages at low temperatures, so that shrivelling and drying of the grapes may be avoided by packaging them in perforated mono PET or multilayer boxes inside the fruit crates. Though the plastic-based perforated packaging method helps for reducing the food loss by extending the shelf-life of food products it also harms the environment with plastic accumulation. Therefore, we suggest that use of dry and to-be water-swollen hydrogels membranes could be taken as an alternative for packaging perishable foods because they have a three-dimensional cross-linked porous structure that allows gas and moisture exchange. Grapes and blueberries could be target food for hydrogel-based packaging [156] replacing the PET or r-PET boxes (Fig. 5) as shown by Bandyopadhyay et al. [70].

The concept of developing hydrogels that could permeate the gas and moisture, mainly for quickly respiring food products (sweet corn, spinach, mushrooms, and most berries) that release more moisture and CO_2 gas, could be an option. Also, the modification of hydrogels for regulated and desired transmission rate can be an avenue for further studies. A model based on the study could pack different products as per standards and norms. This method could be applied in food packaging applications, still this hypothesis need more solid proof and experimental performance. A recent study by researchers from Texas University revealed the use of salt-friendly hydrogels that could extract large amounts of fresh water from the air. The hydrogel was developed from zwitterions which carry both positive and negative charged functional groups that could help the polymer to become responsive to lithium chloride salt. Their study shows that while combining with hygroscopic salt the gel gains the ability to absorb and retain water in the hydrogel. The research team was able to store around six litres of pure water per kilo of hydrogel material in 24 h from the air with 30 % relative humidity. This is a promising study that could efficiently be applied in many fields that need to store water, especially arid regions. Similar concepts can be applied in food product packaging as the excess moisture formed in the packages due to metabolic processes can lead to microbial contamination. If such hydrogels can be used as an absorbent pad, between barrier layers, to absorb moisture from the air inside the package by keeping between a barrier layer exposed to air could be another promising alternative step for food shelf life and preservation [157].

Effective sorting of waste is the key for sustainable environment. All the possible after life scenarios of the bio compostable and biodegradable films are already mentioned by Taneepanichskul et.al [158]. Therefore, effective sorting is equally important for all the claimed biodegradable hydrogel-based packaging materials to help improve composting rates of these materials and reduce the contamination of recycling waste streams. A proper TÜV AUSTRIA BELGIUM labelling or DIN CERTCO certification is essential before a material is launched commercially. Also, as mentioned by Paul-Pont et al. [159] there is a gap in assessing the materials under the light of real life evaluation. The destiny of biodegradable material or the pace at which it decomposes in the environment are still questionable. The toxicological evidences found by different authors [160,161] on bio-based and biodegradable materials can also be a prerequisite for hydrogel based films when they may become available in commercial scale. Since there are no current regulations making it compulsory to get certified it is best to collect the materials together with food waste. Organizations like BioPak New Zealand are already bringing this concept into reality, in future if all kinds of compostable packaging including hydrogel films need to be connected to a responsible recycling stream.

10. Conclusion

Biopolymer-based hydrogels offer a number of noteworthy features including biodegradability, biocompatibility, and sustainability which mark them a better alternative for synthetic polymers. In order to industrialize such polymers for food packaging or pharmaceutical applications, the drawback of these biopolymers such as low mechanical strength, flexibility barriers, shelf life, etc. needs to be overcome by chemical modification, mixing, blending or by functionalizing with bioactive compounds and nanomaterials. This could also be contributing to developing the antimicrobial properties at the same time, raising challenges on the reproducibility of the results. Developing intelligent materials which can be responsive to changing environmental conditions is a better possibility in the packaging industry. More research and development are needed to design hydrogel-based packaging according to the customization of fresh fruits and vegetables. It is expected that by 2050, bio-based food packaging material could successfully contribute to reducing the waste problem created by plastic packaging materials. Utilizing food and fodder crops for packaging material could have an additional burden on the environment. Rather using the leftover farm crops and residues could reduce the environmental burden, provided the end-of-life of the products is also managed properly. Further environmental assessment regarding utilizing leftovers along with modelling

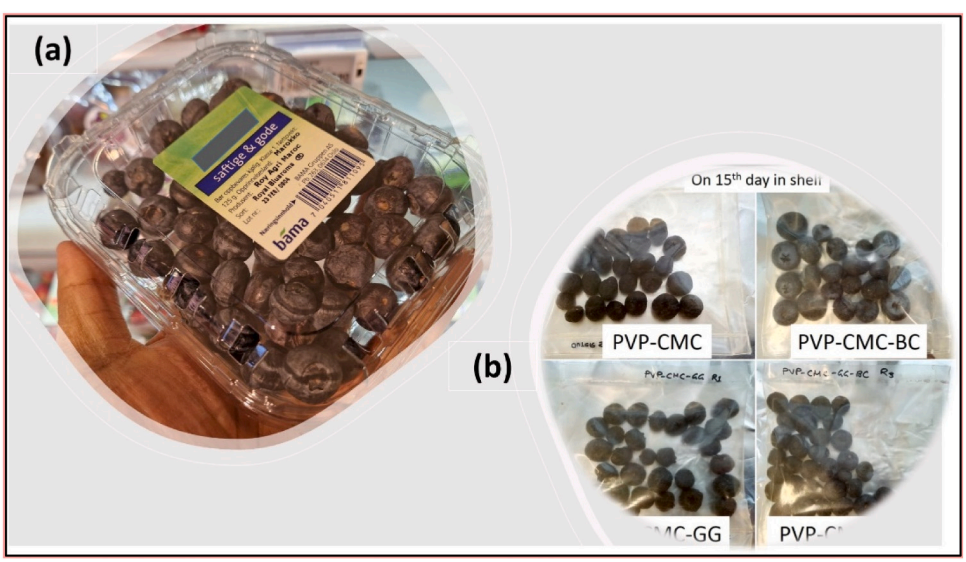

Fig. 5. (a) perforated rPET boxes (b) hydrogel packaging of berries (Bandyopadhyay et al. [70]).

the hydrogel-based packaging materials to fit in the standards are essential.

CRediT authorship contribution statement

The authors have equally contributed to the conceptualization, data curation, original draft preparation and editing work.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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