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Review

Recent Advances in the Preparation, Characterization and Applications of Locust Bean Gum-Based Films

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Abstract: Locust bean gum (LBG) is a typical galactomannan isolated from carob bean (*Ceratonia siliqua* L.) cultivated in the Mediterranean area. Due to its superior biodegradable, rheological and film-forming properties, LBG has been used alone or combined with other biopolymers (e.g., polysaccharides, proteins and lipids) to develop films. Till now, different kinds of functional ingredients (e.g., montmorillonite, bacteriocins, antibiotics, plant extract, essential oils and microbial cells) have been added into LBG-based films. Notably, the physical and functional properties of LBG-based films are affected by many factors, such as the structure of LBG, the type and content of other biopolymers and functional ingredients, and the physical treatment of film-forming solution. LBG-based films can be not only used as active packaging and edible coating in food industry but also used as wound dressings in pharmaceutical industry. For the first time, this review focuses on the recent advances in the preparation, characterization, physical and functional properties, and potential applications of LBG-based films.

Keywords: Locust bean gum; film; preparation; characterization; application

1 Introduction

Traditional plastic packaging materials are non-biodegradable due to their high molecular weights and stable carbon–hydrogen and carbon–carbon bonds. With the increasing awareness of sustainable development, the exploration and utilization of new packaging materials based on renewable bio-sources is becoming a major concern [1]. Up to now, natural and renewable biopolymers including polysaccharides, proteins and lipids have been widely used to fabricate packaging materials [2–4]. These biopolymers are normally derived from natural plants, animals and microorganisms that can be consumed by human beings without causing heath risk and environmental pollution [5]. Therefore, the developed bio-based packaging films have shown potential applications in food and pharmaceutical industries [6–8].

The term "gum" represents a group of polysaccharides isolated from various parts of plants (e.g., cell walls, tree exudates, seeds and tuber/roots). A number of plant seeds (e.g., balangu, basil, cress, wild sage, fenugreek, chia and mesquite seeds) are considered as valuable sources of gums [9]. In food industry, plant gums are widely used as thickeners, emulsifiers, stabilizers, dietary fiber, gelling agents, coating agents and packaging films [10]. Galactomannans, mainly obtained from the endosperm of

Leguminosae seeds, are one category of plant gums. Galactomannans are heterogeneous polysaccharides composed by β-(1→4)-linked D-mannopyranosyl backbone with single D-galactopyranosyl units attached *via* α-(l–6) linkage as side branch. It has been demonstrated that galactomannans are important resources of packaging film production due to their edibility, biodegradability and good film-forming ability [11]. The substitution degree of galactose in mannan backbone depends on the plant source of galactomannans and remarkably affects the solubility and the film-forming ability of galactomannans [12].

Locust bean gum (LBG), isolated from carob bean (*Ceratonia siliqua* L.) cultivated in the Mediterranean area, is a typical galactomannan with the mannose/galactose (M/G) ratio of about 4:1 (Fig. 1) [13]. LBG is partially soluble in cold water and normally needs to be heated to reach maximum solubility. Due to its superior biodegradable, rheological and film-forming properties, LBG is suitable to be developed into packaging films [14−16]. The semi-permeable barrier provided by LBG-based films is benefit to reduce moisture migration, gas exchange, respiration and oxidative reaction rates, thereby extending the shelf-life of food products [14]. Meanwhile, LBG-based films can serve as the carrier of functional ingredients to develop active packaging and wound dressings [17]. However, there is not a specific review concerning LBG-based films up to now. Therefore, this review focuses on the recent advances in the preparation, characterization, physical and functional properties, and potential applications of LBG-based films.

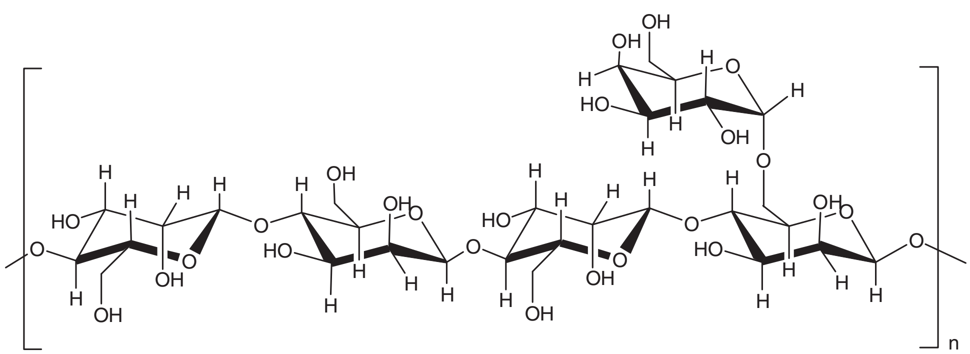

Figure 1: The chemical structure of LBG

2 Preparation and Characterization of LBG-Based Films

The formulation, preparation method, and conditions for dry and storage of LBG-based films are summarized in Tab. 1. Till now, LBG-based films are mainly prepared by solvent casting method. Solvent casting is a wet method for film preparation, which does not require specialized equipment. In general, film forming solutions are prepared by thoroughly dissolving LBG in water at about 80°C for 30 min. Meanwhile, LBG is often blended with other biopolymers (e.g., polysaccharides, proteins and lipids) to produce films with improved physical properties [18–33]. In addition, other functional ingredients including plasticizers, hydrophobic substances, natural active substances and microbial cells are also incorporated into the solutions to enhance the physical and functional properties of the films [11, 20, 24, 26, 28, 30–35]. To obtain homogeneous film forming solutions, LBG and other film components are completely dissolved and mixed in the solutions. The resultant film forming solutions are normally degassed by ultrasonic treatment and then poured onto flat plate. The solvents are allowed to evaporate by drying at 20–45°C for a few hours. The obtained films are peeled off from the plate and conditioned under controlled temperature and relative humidity (RH) before use. Notably, commercially used packaging films are normally prepared by extrusion method that employs high temperature and shear forces in film preparation. Extrusion is a dry method for film preparation that has some advantages over solvent casting method, such as industrially scalable and solvent-free. However, the high temperature and shear forces of extrusion method can affect the stability of bioactive substances in the film. Therefore, extrusion method has not been used in the preparation of LBG-based films up to now. In order to solve this problem, the thermal sensitive substances can be first encapsulated in nano materials and then incorporated into the films by extrusion method. In the future, extrusion technique can be compared with solvent casting method for the development of LBG-based films.

Table 1: The formulation, preparation method, and conditions for dry and storage of LBG-based films

Film matrix	Plasticizer	Functional addictives	Film preparation method	Conditions for film dry and storage	References
LBG/κ-carrageenan blend	Glycerol	Natamycin	Solvent casting	Dried at 25°C for 48 h and then stored at 20°C and 53% RH	[11]
LBG/agar blend	Glycerol		Solvent casting	Dried at 60°C overnight and then stored at 25°C and 38 ± 1% RH	[18]
LBG/κ-carrageenan blend			Solvent casting		[19]
LBG/κ-carrageenan blend	PG	Montmorrillonite and curcumin	Solvent casting	Dried at 35°C for 16 h and then stored in a desiccator	[20]
LBG/corn starch/PVA blend	Glycerol		Solvent casting	Dried at 30 ± 2°C for 36 h	[21]
LBG/xanthan gum blend	Glycerol		Solvent casting	Dried at 40°C for 24 h and then stored at 24°C and 43% RH for at least 48 h	[22]
LBG/κ-carrageenan blend	Glycerol		Solvent casting	Dried at 35°C for 16 h and then stored at 20 ± 1°C and 54 ± 1% RH	[23]
LBG/κ-carrageenan blend	Glycerol	Montmorrillonite organoclay	Solvent casting	Dried at 35°C for 16 h and then stored at 20 ± 1°C and 54 ± 1% RH	[24]
LBG/tragacanth gum blend			Solvent casting	Dried at 30°C for 24 h and then stored at 23°C and 53% RH	[25]
LBG/κ-carrageenan blend	Glycerol	Hemicellulose	Solvent casting	Dried at 35°C for 16 h and then stored at 20 ± 1°C and 54 ± 1% RH	[26]
LBG/whey protein blend	Glycerol		Solvent casting	Dried at 35 ± 2°C and 50 ± 1% RH and then stored at 25 °C and 53% RH	[27]
LBG/κ-carrageenan/whey protein blend	Glycerol	*Lactobacillus rhamnosus GG*	Solvent casting	Dried at 37°C and 50% RH and then stored at 25 or 4°C and 54 or 59% RH	[28]
LBG/native or alkali-modified agar blend			Solvent casting		[29]

(Continued)

Table 1 (continued).

Film matrix	Plasticizer	Functional addictives	Film preparation method	Conditions for film dry and storage	References
LBG/egg white albumin blend; LBG/collagen blend	Glycerol	Emulsified maize oil	Solvent casting	Dried at 25 ± 1°C and 55 ± 5% RH for 48 h	[30]
LBG/pullulan/ xanthan gum blend	Glycerol	Cinnamaldehyde, eugenol, and thymol emulsions	Solvent casting	Dried at 21°C and 40% RH for 24 h	[31]
LBG/pullulan/ xanthan gum blend	Glycerol	Sakacin A	Solvent casting	Dried at 25°C and 40% RH for 24 h	[32]
LBG/κ-carrageenan blend	Glycerol	Cranberry extract	Solvent casting	Dried under airstream at room temperature and stored in a desiccator	[33]
LBG	Glycerol	*Wickerhamomyces anomalus* killer yeast	Solvent casting	Dried at room temperature for 48 h and then stored at 25°C and 75% RH	[34]
LBG	Glycerol, PG, sorbitol and PEG	Stearopten and beeswax	Solvent casting	Dried at 25 ± 5°C and 60 ± 5% RH for 1 day	[35]
LBG	Glycerol		Solvent casting	Dried at 45°C for 12 h and then stored at 25°C and 53% RH for at least 48 h	[36]
LBG	Glycerol		Solvent casting	Dried at 40°C for 15 h and then stored at room temperature and 43% RH	[37]
LBG	Glycerol		Solvent casting	Dried at 35°C for 16 h and then stored at 25 ± 1°C and 53% RH	[38]
Carbamoylethyl LBG	Glycerol		Solvent casting	Dried at 45°C for 24 h	[39]
LBG	PEG		Solvent casting	Dried at room temperature for 1 day	[40,41]

LBG, locust bean gum; PEG, polyethylene glycol; PG, propylene glycol; PVA, polyvinyl alcohol; RH, relative humidity

As shown in Fig. 2, the structure of LBG-based films is normally characterized by scanning electron microscopy (SEM), Fourier transform infrared (FT-IR) spectroscopy and X-ray diffractometer (XRD). Among different techniques, SEM is employed to observe the microstructure (i.e., surface and cross-section) of the films. The compactness and homogeneity of the films are mainly affected by the miscibility of LBG and other film components. Since LBG is hydrophilic, it is well miscible with water soluble biopolymers (e.g., agar, κ-carrageenan, xanthan gum and tragacanth gum) and functional ingredients (e.g., anthocyanins) [19,22,25,29,33]. The existence of intermolecular interactions (especially hydrogen bonds) occurring between LBG and other film components is confirmed by FT-IR spectroscopy. The broad band at 3500–3300 cm^{-1} (O–H stretching vibration of hydroxyl groups in LBG) often shifts after the incorporation of other biopolymers, such as κ-carrageenan, tragacanth gum, egg white albumin and collagen [11,19,25,30]. The intermolecular interactions are closely related to the physical properties (e.g., water sensitive, barrier, mechanical and thermal properties) of the films. XRD is often employed to analyze the crystallinity and miscibility of the blended films. If blended films have low miscibility, each film component will exhibit its own crystal character. Since LBG is in the amorphous phase, the crystallinity of the films is generally influenced by the character of other film components as well as their interactions [20,22,23,24,36].

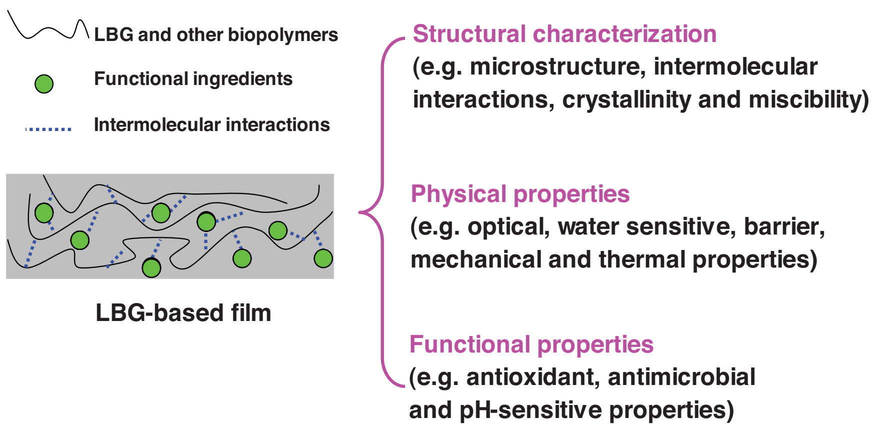

Figure 2: The structural characterization, physical and functional properties normally determined for LBG-based film

The physical properties of LBG-based films including optical (e.g., color, transmittance and opacity), water sensitive (e.g., moisture content, contact angle, solubility, water sorption and swelling), barrier (water vapor permeability (WVP) and oxygen and carbon dioxide permeability), mechanical (tensile strength (TS), elongation at break (EAB) and elastic modulus (EM), folding endurance and puncture resistance) and thermal (glass transition and thermal degradation) properties are normally determined (Fig. 2). In general, LBG-based films possess good optical, barrier and mechanical properties at low RH conditions. However, the high hydrophilicity of LBG makes the films very sensitive to water and moisture, which greatly limits the application of the films at high RH conditions [11,20,25,27,30]. Notably, the incorporation of other biopolymers and functional ingredients can greatly improve the physical and functional properties of LBG-based films. It has been demonstrated that the addition of montmorillonite organoclay, natamycin, sakacin A and essential oils can significantly enhance the antioxidant and antimicobial activities of LBG-based films [11,24,31,32]. In addition, LBG-based films became pH-sensitive when anthocyanins-rich cranberry extract is incorporated into the films [33]. Factors affecting the physical and functional properties of LBG-based films are summarized in the next section.

3 Factors Affecting the Physical and Functional Properties of LBG-Based Films

3.1 LBG Structure

The physical properties (e.g., solubility and viscosity) of galactomannans depend on the M/G ratio. In general, galactomannans with a lower M/G ratio show higher water solubility but lower viscosity because galactose side groups can prevent galactomannans to form effective intermolecular interactions [36]. Therefore, the comparative study of galactomannans with different M/G ratios is a useful approach to reveal the structure-function relationship of LBG-based films. Kurt et al. [37] compared the physical properties of salep glucomannan, LBG and guar gum-based films. They suggested the moisture content, solubility, WVP, TS and EAB of the films were remarkably affected by the substitution degree of galactose along mannan backbone. In a similar study, galactomannans with different M/G ratios (1.3, 1.7, 2.9, 3.4 and 5.6) were isolated from five plant sources including *Adenanthera pavonina*, *Cyamopsis tetragonolobus*, *Caesalpinia pulcherrima*, *Ceratonia siliqua* and *Sophora japonica*, respectively. Then, the physical properties of galactomannan-based films with different M/G ratios were compared [38]. Results showed M/G ratio had a significant influence on the WVP, gas permeability, water affinity, mechanical and thermal properties of the films. Recently, the film-forming properties of three different galactomannans including guar gum (M/G ratio = 2), tara gum (M/G ratio = 3) and LBG (M/G ratio = 4) were compared by Liu et al. [36]. Results showed the film network became denser and more compact when the M/G ratio of galactomannans increased, resulting in the improved mechanical and water vapor barrier properties. The possible reason is that fewer side chains and long blocks of unsunstituted mannose units of galactomannans can facilitate the intermolecular chain interactions.

The chemical modification of LBG is another way to tailor structures and impart functions of LBG-based films. Carbamoylethyl LBG is synthesized by substituting the hydroxyl groups of LBG with amide groups ($-CH_2CH_2CONH_2$) [39]. The amorphous nature, non-Newtonian flow and shear-thinning behavior of native LBG were retained in the carbamoylethyl derivative of LBG. The carbamoylethyl LBG-based films showed a quite low WVP, indicating the films were highly resistant towards water. However, carbamoylethyl LBG-based films have not been compared with unmodified LBG-based films for their physical properties. In the future, other structural characters of LBG (e.g., molecular weight, the distribution of D-galactosyl units along mannan backbone, the type of substitution group and substitution degree) on the physical properties of the films can be further investigated.

3.2 Other Biopolymers

The blending of LBG with other biopolymers can effectively change the rheological property of film-forming solutions and improve the physical properties of corresponding films [22,25,27,29]. Till now, LBG has been blended with several other polysaccharides (e.g., agar, *κ*-carrageenan, starch, xanthan gum, tragacanth gum and pullulan) and proteins (e.g., whey protein, egg white albumin and collagen) to develop composite films (Tab. 1). Among different composite films, LBG/*κ*-carrageenan blend films have been most widely studied [11,19,20,22–24,26,33]. Due to the presence of abundant hydroxyl groups, LBG can form synergistic interactions with other biopolymers through hydrogen bonds. The presence of synergistic interactions between LBG and other biopolymers have been verified by different instrumental methods, such as textural profile analysis (TPA), rheology measurement, FT-IR spectroscopy, SEM and XRD techniques [19,23,25,27,29].

It has been demonstrated that the physical and functional properties of the blend films are mainly affected by the weight ratio of LBG and other biopolymers. Martins et al. [23] reported the highest TS and the lowest WVP of LBG/*κ*-carrageenan films were obtained when LBG/κ-carrageenan weight ratio reached 3:2. He et al. [19] found LBG/κ-carrageenan films with LBG/*κ*-carrageenan weight ratio of 1:3 had the highest gelling and mechanical properties, which could be further developed into hard capsules as pharmaceutical excipients. The effect of LBG/xanthan gum weight ratio on the WVP and mechanical properties of the films was investigated by multiple response optimization technique. The optimal film formulations were determined as LBG 89.6%, xanthan gum 10.4% and glycerol 20% [22]. The impact of LBG content on the physical properties of LBG/corn starch/polyvinyl alcohol (PVA) films was measured. The incorporation of LBG increased the TS but reduced the EAB and enzymatic hydrolysis rate of the films. In addition, a higher content of LBG had a greater impact on the physical properties of the films [21]. Similarly, Mostafavi et al. [25] documented the moisture content and WVP of LBG/tragacanth gum films decreased but the TS and EAB of the films increased when the proportion of LBG in the films increased from 25 to 100%. Recently, Akkaya et al. [18] found the light transmittance, TS and EAB decreased but the WVP, swelling ratio and antibacterial activity of LBG-agar films increased when the proportion of LBG in the films increased from 25 to 75%.

3.3 Plasticizers

Plasticizer is a basic film component that can reduce intermolecular interactions and enhance the mobility of biopolymeric chains, thereby altering the mechanical properties of the films. The addition of plasticizer is essential to overcome the brittleness of the films. Due to the hydrophilic nature of LBG, it is essential to use of polar plasticizers for better compatibility [40,41]. As summarized in Tab. 1, different plasticizers including glycerol, polyethylene glycol (PEG), propylene glycol (PG) and sorbitol have been incorporated into LBG-based films. Bozdemir et al. [35] compared the WVP of LBG-based films containing different plasticizers (i.e., glycerol, PG, sorbitol and PEG 200). Obvious phase separation and physical exclusion of plasticizer were observed when sorbitol was added into the films. By contrast, the other plasticizers (glycerol, PG, and PEG 200) were more compatible with LBG. In general, the macro-phase separation of films indicates the less miscibility, while the micro-phase separation of films indicates the improved compatibility [42,43]. It was found that the films plasticized with PEG 200 had the lowest WVP. However, the films containing glycerol had the highest WVP because glycerol possessed high affinity towards water vapor. Aydinli et al. [40] examined the impacts of the amount and molecular weight of PEG (PEG 200, PEG 400 and PEG 600) on the WVP, mechanical and light transmittance properties of LBG-based films. Results showed that the WVP of the films increased but the light transmittance, TS and EAB of the films decreased when the amount and molecular weight of PEG increased. Therefore, the most suitable plasticizer of LBG-based films is PEG 200 and its content in the films should be carefully controlled to guarantee the film compatibility [41].

3.4 Hydrophobic Substances

The hydrophilic LBG-based film matrix can form strong chain-chain interactions which provide good barrier property to gases, such as O_2 and CO_2. However, these interactions are adversely affected by moisture absorption in the films. As a result, the barrier properties of LBG-based films generally decline with the increase of RH. Thus, it is essential to incorporate hydrophobic substances into LBG-based films. In this respect, hydrophobic lipids including emulsified beeswax, stearopten, and maize oil were added into LBG-based films [30,35]. The films added with emulsified lipids showed reduced WVP but enhanced TS and EAB as compared to the films without emulsified lipids. The improved water vapor barrier and mechanical properties of the films were attributed to intermolecular interactions between lipids and film matrix.

The addition of nano materials is a promising option to improve the hydrophobic and antimicrobial properties of polysaccharide-based films [44]. In this respect, nanocomposite films are prepared by homogeneously dispersing hydrophobic clay minerals in the films. Montmorillonite organoclay modified by the addition of quaternary ammonium salts was dispended in LBG/κ-carrageenan blend to develop biocomposite films [24]. Montmorillonite organoclay could form hydrogen bonds and/or electrostatic interactions with film matrix and block the micro-paths in the film network. Meanwhile, montmorillonite organoclay could release ammonium salts from surfactant-modified clay and exhibit antimicrobial activity against *Listeria monocytogenes*. Therefore, the incorporation of montmorillonite organoclay significantly reduced the WVP whereas increased the CO_2 permeability, TS, EAB and antimicrobial activity of the films. Moreover, the physical and functional properties of the films were closely associated with the content of montmorillonite organoclay. Recently, similar biocomposite films were developed by adding unmodified montmorrillonite into LBG/κ-carrageenan blends [20]. Results showed the addition of montmorillonite reduced the moisture content, folding endurance, swelling ratio and WVP whereas enhanced the TS and EAB of the films.

3.5 Natural Active Substances

Natural active substances are frequently incorporated into LBG-based films to develop active packaging materials [45]. Antimicrobial agents obtained from fermentation processes (e.g., bacteriocins and antibiotics) are commercially available. Natamycin, a polyene macrolide antimycotic agent produced by the actinomycete, was incorporated into LBG/*κ*-carrageenan blend to develop antimicrobial active packaging films [11]. Results showed the incorporation of free natamycin and natamycin-loaded poly(*N*-isopropylacrylamide) nanohydrogels significantly changed the optical and mechanical properties of the films. However, further studies are needed to evaluate the release behavior of natamycin from the films into different food model systems. Trinetta et al. [32] loaded sakacin A (a bacteriocin) into LBG/pullulan/xanthan gum blend films and verified the films had strong antimicrobial activity against *L. monocytogenes*. The incorporation of sakacin A had little impact on the light transmittance of the films, however, significantly reduced the TS of the films. Nonetheless, the effect of sakacin A content on the physical and functional properties of the films was not determined and needs further investigations.

Natural plant extracts commonly possess potent antioxidant and antimicrobial activities due to the presence of abundant polyphenolic compounds [46]. However, studies on the development of active packaging films based on LBG and polyphenol-rich plant extracts are very limited. Anthocyanins, one category of phenolic compounds, are the largest and most important water-soluble pigments in fruits, vegetables, flowers and cereals. Anthocyanins-rich films are promising active and intelligent packaging materials that can be applied in food and pharmaceutical industries [47]. On one hand, anthocyanins possess excellent antioxidant and antimicrobial activities, which can be used in active packaging to extend the shelf life of food products. On the other hand, anthocyanins are pH-sensitive and normally change their colors as a function of pH, which can be utilized in intelligent packaging to monitor the freshness of protein-rich food products (e.g., fish, shrimp, chicken, pork, milk and cheese) [48,49]. Recently, pH-sensitive films were developed by added anthocyanins-rich cranberry extract into LBG/*κ*-carrageenan blend [33]. The films containing cranberry extract could respond to pH changes in different buffer solutions and continuously monitor bacterial infections in wound. In the future, anthocyanins-rich LBG-based films can be used as active and intelligent packaging materials in food industry.

Essential oils extracted from plants and spices are natural antioxidant and antimicrobial agents for active packaging development [50]. The film microstructure is normally weakened by the addition of essential oils due to partial replacement of strong chain–chain interactions by weak polymer-oil interactions in the film network. However, the water barrier property of the films is improved by essential oils due to their hydrophobic characters. Recently, natural emulsified essential oils including thymol, cinnamaldehyde, and eugenol were incorporated into LBG/pullulan/xanthan gum blend to develop active packaging films [31]. Results showed thymol had the highest release rate from the films, which was followed by eugenol and cinnamaldehyde. This study suggests essential oils loaded films can be used as active packaging materials with controlled release of active compounds. Nonetheless, the impact of essential oils on the physical and functional properties of LBG-based films can be further determined.

3.6 Microbial Cells

Green mould caused by *Penicillium digitatum* is one the major postharvest diseases of citrus fruits, which limits the shelf life of harvested fruits. The application of microbial antagonists is an effective way to control postharvest fruit infections caused by *P. digitatum*. Aloui et al. [34] incorporated *Wickerhamomyces anomalus* killer yeast into LBG-based films and evaluated the efficiency of the films to maintain viability and antifungal potential of incorporated microbial cells. Results showed that LBG films were able to maintain more than 85% of the initial *W. anomalus* yeast population and to completely inhibit the growth of *P. digitatum* in synthetic medium. Notably, the incorporation of *W. anomalus* yeast had little impact on the optical, mechanical and barrier properties of the films.

Probiotics are living non-pathogenic microorganisms that confer health benefits to human hosts. Due to the low stability of probiotics under common food processing conditions (e.g., heat, pH, osmotic pressure and redox potentials), the use of edible films to embed viable probiotics has received increasingly attention. Soukoulis et al. [28] examined the potential of LBG/κ-carrageenan/whey protein blend films as the vehicle of probiotic *Lactobacillus rhamnosus* GG. The films possessed compact microstructures and exhibited acceptable mechanical and barrier properties. Moreover, the films were able to stabilize *L. rhamnosus* GG during storage at 4 and 25°C for 25 days. In the future, the survival of probiotics throughout ingestion and gastrointestinal passage can be further studied.

3.7 Physical Treatment

Some physical treatments including gamma irradiation and thermal treatment have been used to improve the properties of LBG-based films. The impact of gamma irradiation on the properties of LBG/corn starch/PVA blend films was investigated by Kim et al. [21]. The film-forming solutions were irradiated by a cobalt-60 irradiator with different irradiation doses (0, 3, 6, 12 and 24 kGy). The application of gamma irradiation in the film-forming solutions resulted in intact and smooth films. Meanwhile, the enzymatic hydrolysis rate and WVP of the films decreased with the increase of irradiation dose. This study suggests the irradiation technology is a useful tool to cross-link LBG with other biopolymers, resulting in the improved functional properties of the films. However, the irradiation dose should be accurately control to guarantee the safety of the produced films. In another study, LBG/whey protein isolate blend film-forming solutions were prepared by two different thermal treatments before casting: 1) the film-forming solutions were heated until 75°C and immediately cooled back to room temperature; 2) the film-forming solutions were heated until 75°C, kept at 75°C for 10 min and then cooled back to room temperature [27]. Results showed that more severe heat treatment (the second thermal treatment) produced films with higher TS and EAB but lower solubility and gas permeability. The difference in the physical properties of the films prepared under different thermal treatments was caused by the distinct strength of intermolecular interactions between LBG and whey protein isolate

4 Potential Applications of LBG-Based Films

4.1 Food Packaging Films and Edible Coating

Fruits, vegetables, meat, fish and derived products are perishable food items with quick deterioration under improper storage. Polysaccharide-based packaging films are effective in preserving perishable food [51]. However, studies on the application of LBG-based films in food packaging are very limited up to now, which is probably because native LBG films are highly hydrophilic and normally loss their functionality under high RH conditions. In the future, it is essential to reduce the water sensitive property of native LBG films by adding hydrophobic substances or by chemically modifying LBG with hydrophobic groups. Moreover, natural active substances can be added into LBG-based films to develop active food packaging.

Apart from food packaging films, edible coating is another application form of LBG-based film-forming solutions. Edible coating can create a semi-permeable barrier against moisture and gas (e.g., O_2 and CO_2), thereby limiting the weight loss, oxidation and respiration rates of the packaged food. Meanwhile, it can act as the carrier of plasticizers, emulsifiers and natural active substances to enhance the functionality of the matrix [51]. Till now, LBG-based edible coating has been widely used in food preservation. LBG/lipid composite coating showed a better performance in controlling the weight loss and ethanol production during postharvest storage of ‘Fortune’ mandarins [52]. LBG-based coating enriched with *W. anomalus* yeast effectively reduced the weight loss, maintained the firmness and inhibited green mold formation during the postharvest storage of ‘Valencia’ oranges [34]. In a similar study, LBG-based coating enriched with *W. anomalus*, *Metschnikowia pulcherrima* and *Aureobasidium pullulans* species effectively controlled postharvest decay in mandarins that were artificially inoculated with pathogenic molds *P. digitatum* and *Penicillium italicum* (Fig. 3) [53]. Except for microbial cells, natural active substances (e.g., pomegranate peel extract and *Foeniculum vulgare* essential oil) were incorporated into LBG-based coating. Results showed that LBG-based coating with natural active substances had potent antimicrobial activity and could maintain the quality of white shrimps and globe artichoke slices during refrigerated storage [54,55]. Kharchoufi et al. [56] further demonstrated the *W. anomalus and* water pomegranate peel extract had synergistic effect on reducing the postharvest decay of oranges inoculated with *P. digitatum*. This study provides a new insight into the development of LBG-based coating by combining biocontrol agents and natural active compounds.

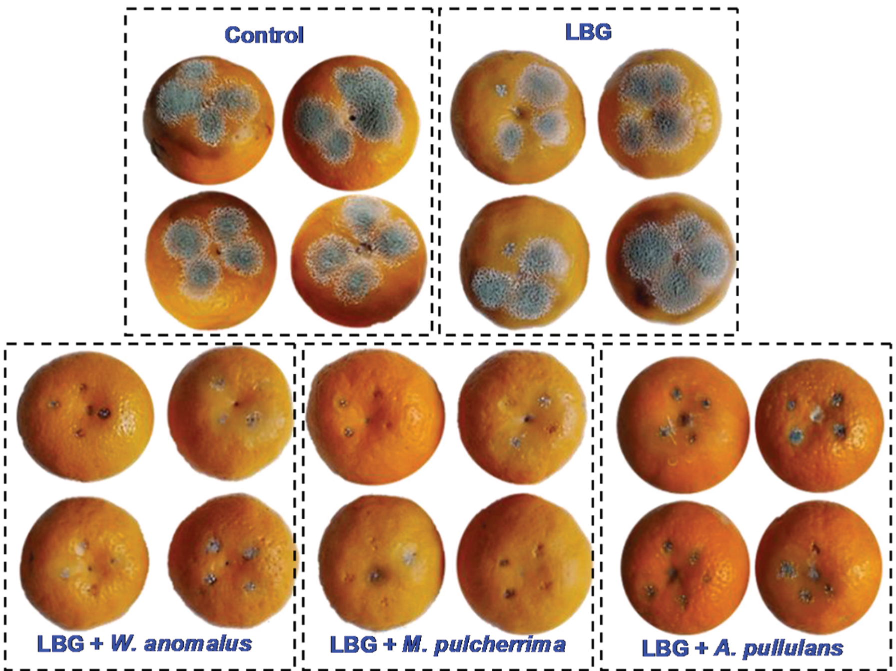

Figure 3: The application of LBG-based coating enriched with *W. anomalus*, *Metschnikowia pulcherrima* and *Aureobasidium pullulans* species to control postharvest decay in mandarins. Adapted from Parafati et al. [53] with permission from Elsevier, copyright 2019

4.2 Wound Dressings

Wound healing stands as a very complex and dynamic process, aiming the re-establishment of the damaged tissue's integrity and functionality. Wound dressings are biomaterials utilized to cover the wounds in order to absorb exudates, maintain the moisture balance, allow gas exchange and prevent the wounds from being infected [57]. Currently, wound dressings formulated from polysaccharides have gained great attention due to their non-toxicity, biocompatibility, biodegradability and easy processing and mouldability. In addition, polysaccharide-based biomaterials can be loaded with wound healing agents to promote wound healing in clinical applications [58]. The hydrophilic nature of LBG makes it possible to blend with other biopolymers and/or inorganic materials to prepare innovative wound dressings. Till now, some studied have been carried out to examine the wound healing effect of LBG-based films. Kaur et al. [20] developed LBG/κ-carrageenan/montmorrillonite biocomposite films and used the films for transdermal delivery of curcumin (a wound healing and anti-inflammatory agent). Results showed the films could constantly release 69.38% of curcumin in 24 h and showed good skin permeation and wound healing effect. Recently, Akkaya et al. [18] demonstrated LBG-agar gum films were non-cytotoxic to NIH 3T3 cells and could be used as antibacterial wound dressing candidates. In clinical applications, monitoring the pH of the wound is essential to know its healing status because the pH of bacterial infected wound normally rises from mildly acidic (pH 4.0–6.0) to alkaline values (pH 7.0–9.0). As shown in Fig. 4, smart wound dressings were developed based on LBG/κ-carrageenan films incorporated with anthocyanins-rich cranberry extract [33]. The developed films could respond to pH variations in different buffer solutions due to the presence of pH-sensitve anthocyanins. Thus, the films could continuously monitor bacterial infection in the wound. In the future, novel wound dressings can be developed by blending LBG-based films with wound healing ingredients.

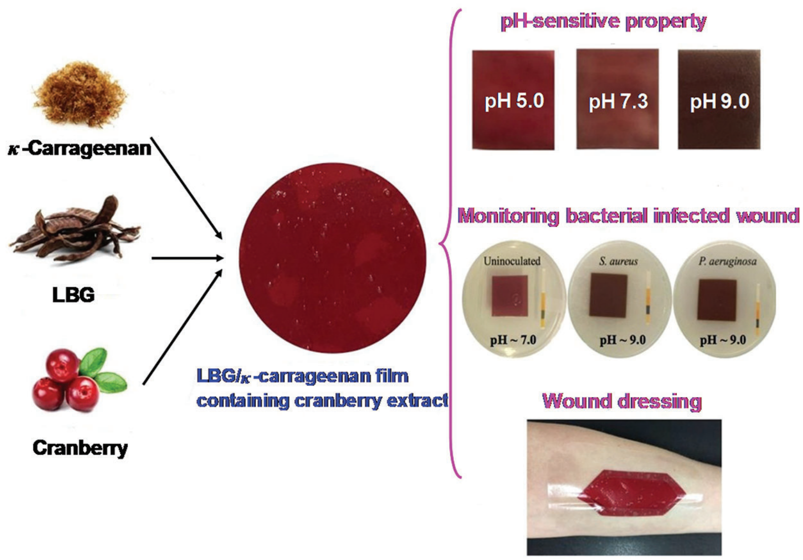

Figure 4: The application of LBG/κ-carrageenan films incorporated with anthocyanins-rich cranberry extract in smart wound dressing. Adapted from Zepon et al. [33] with permission from Elsevier, copyright 2019

5 Conclusions

LBG is suitable to be developed into packaging films due to its superior biodegradable and film-forming properties. Till now, LBG-based films are mainly prepared by solvent casting method due to its simplicity. However, industrially applied extrusion method has not been used in the preparation of LBG-based films. In the future, extrusion technique can be used and compared with solvent casting method to develop LBG-based films. In general, the structural characterization (e.g., microstructure, intermolecular interactions, crystallinity and miscibility), physical properties (e.g., optical, water sensitive, barrier, mechanical and thermal properties) and functional properties (e.g., antioxidant, antimicrobial and pH-sensitive properties) of LBG-based films are determined. The structure of LBG, the type and content of other biopolymers and functional ingredients, and the physical treatment of film-forming solution are considered as important factors affecting the physical and functional properties of LBG-based films. The comparison of galactomannans with different M/G ratios and the chemical modification of LBG are both useful to reveal the structure-function relationship of LBG-based films. In the future, LBG can be blended with other biopolymers and functional ingredients (e.g., nanoparticles, bacteriocins, antibiotics, polyphenol-rich plant extracts, essential oils, biocontrol agents and probiotics) to develop novel functional films. The developed films can be widely used as active packaging, edible coating, and wound dressings in food and pharmaceutical industries.

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