Contents lists available at ScienceDirect

Trends in Food Science & Technology

journal homepage: www.elsevier.com/locate/tifs



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ARTICLE INFO

Keywords: Shellac Natural polymer Molecular structure Property Food industry Application

ABSTRACT

Background: The promising applications of polymers have recently attracted increasing interest in the food industry, prompting the development and utilization of novel polymers with critical roles in future foods. One such polymer with significant advantages in food applications is 'shellac,' a long-established natural polymer furnished with distinctive properties, including pH-responsiveness, biodegradability, and biocompatibility.

Scope and approach: In this review, literature involving food-related shellac applications, selected from Web of Science, were used in content development. First, various shellac sources, extraction methods, and processing methods are introduced, and their chemical compositions and identification reviewed. Subsequently, the physicochemical properties of shellac are discussed in detail. Finally, the applications of shellac in the food industry are comprehensively reviewed.

Key findings and conclusions: The extraction and processing technology for shellac have matured, and the structure of shellac is now clearly understood. Based on the recognized structures and properties of shellac, its application is widely advantageous in all aspects of the food industry. However, the development of new food-related shellac applications is dependent on fabrication information based on existing research reports, hence the need for elucidation. In addition, the constraints of shellac, which results from self-polymerization, and their consequent effects on the stability of shellac-based food systems are rarely reported, and it is necessary to address these issues. In general, the application of shellac in the food industry shows great potential.

1. Introduction

Natural polymers, which include proteins, starches, and other polysaccharides, are used not only as ingredients or additives to endow food with specific structural and functional characteristics, but also as processing materials for food applications (Djagny, Wang, & Xu, 2001; Jobling, 2004; Wijaya, Patel, Setiowati, & Van der Meeren, 2017). The intrinsic characteristics of these natural polymers favoring their application, especially as processing materials, include their good biocompatibility, biodegradability, and non-toxicity (Rhim & Ng, 2007). In addition, global health and environmental awareness has caused a tremendous shift in consumer preferences, from synthetic to natural polymers. Hence, the subsequent creation of more natural products in the food industry. Interestingly, natural polymers show comparable or superior performance to synthetic polymers in specific applications. For example, konjac glucomannan, agarose, and sodium alginate have important applications in the processing of non-plastic packaging films (Wu et al., 2019; Yuan, Hong, et al., 2017; Yuan, Mu, et al., 2017), thermoreversible hydrogels (Yuan et al., 2018), and pH-responsive microspheres (Yuan et al., 2019), respectively. Shellac possesses a

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https://doi.org/10.1016/j.tifs.2021.01.031

Received 26 August 2020; Received in revised form 5 January 2021; Accepted 9 January 2021 Available online 15 January 2021 0924-2244/© 2021 Elsevier Ltd. All rights reserved.





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combination of these characteristics, including good film formability, pH-responsiveness, and amphiphilicity, making it a promising processing material in the food industry.

Shellac is a natural polymer refined from a resinous substance excreted by an insect, *Laccifer lacca*, which is parasitic on certain trees, especially in India, Burma, Thailand, and southern China (Wang, Ishida, Ohtani, Tsuge, & Nakayama, 2004). In these regions, shellac has been recognized for about 4000 years (Gibson, 1942) and was originally used as a natural dye for architecture, silk, and leather dyeing. Subsequently, the applications of shellac gradually extended to include the pharmaceutical field, because of its good enteric properties (Azouka, Huggett, & Harrison, 1993). Additionally, shellac is generally recognized as safe (GRAS status) by the Food and Drug Administration (FDA), allowing its use as a food additive and raw material in food coatings and film formulations (Farag & Leopold, 2011).

Structurally, shellac is a low-molecular-weight resin mainly composed of oxyacid polyesters. The oxyacids are divided into aleuritic acids and cyclic terpene acids linked by ester bonds, which, respectively, constitute the hydrophobic and hydrophilic components of shellac (Luo et al., 2016). Therefore, shellac possesses the additional advantage of exhibiting excellent amphiphilicity, compared to the biodegradability and biocompatibility of other natural polymers (Patel, Drost, Seijen ten Hoorn, & Velikov, 2013). Furthermore, shellac is characterized by carboxyl groups in its molecular structure, which are exempted from the cyclic terpene acid esterification reaction, thereby endowing shellac with weak acidity (about pKa 6). Consequently, shellac dissolves in alkaline solutions rather than in acidic solutions, prompting its pH-responsiveness (Buch, Penning, Wächtersbach, Maskos, & Langguth, 2009). Owing to these exceptional characteristics of shellac (amphiphilicity and pH-responsiveness), its application in the development of novel foods is on the rise. These novel food applications, which have attracted increasing attention over the last few decades, include food delivery systems, food foaming agents, oil-gelling agents, and food emulsifiers.

Although a substantial number of studies in recent years have captured the use of shellac in the food industry, to the best of our knowledge, there is a paucity of relevant reviews on the application of shellac in the food industry. Therefore, we retrieved related articles from Web of Science for a comprehensive review of the applications of shellac in the food industry. First, this review will introduce the sources, extraction methods, chemical composition, identification, and excellent performance of shellac. Then, the applications of shellac in the food industry will be comprehensively introduced. Finally, the health considerations and future perspectives of shellac, regarding its food-related applications, are discussed. Through this comprehensive review, we aim to provide more theoretical references and practical directions to inspire the further developments and applications of shellac-based food systems.

2. Sources, extraction, and processing

Generally, there are as many as 100–150 *Laccifer lacca* (the insect that produces shellac, as previously mentioned) larvae per inch of host-tree twig. To survive, these larvae move onto specific host trees for 2–3 days each, inserting their proboscis into the phloem tissue to reach the sap. Consequently, shellac is secreted to form cells around their bodies, aiding their adherence to the host-tree branches. After that, male insects are moved out from their cells, while females still live in them. The male insect subsequently fertilizes the female after about 8 weeks and then dies within a few days. The fertilized female continues to secrete large amounts of shellac, and produces an average of 200–500 larvae (Azouka et al., 1993), which continues the cycle of shellac secretion and breeding.

The raw shellac collected from trees twigs is known as 'sticklac' and is processed into shellac for further use. It is either processed by hand or by machine (Derry, 2012). Industrially, the sticklac is first separated

from impurities, such as wood materials, followed by crushing and washing. The washed lac is then dried in a drying chamber using hot airflow under strict temperature regulation to prevent it from melting. The dried seedlac is then processed into shellac either in a heating process, using a steam-heated hydraulic press with a lining cloth to squeeze out the melt lac, or in a solvent extraction process. The solvent extraction process involves dissolving the seedlac in ethanol, filtering the obtained solution through a fine cloth to remove any insoluble matter, and finally eliminating the alcohol.

The dark brown color of native shellac extracts limits its application, necessitating color removal (bleaching) using decolorizing agents before use in some fields (Derry, 2012). NaClO is the commonly used for the removal of lac pigments and waxes, thereby improving its appearance to fit a wider range of applications, including those in the pharmaceutical, food, and fruit processing domains (Liao, Zhou, Huang, & Li, 2015). However, during the bleaching process, chlorine is often added to the double bonds of shellac, forming combined chlorine. Combined chlorine reduces the properties of shellac; therefore, reagents such as sodium ethoxide (Liao et al., 2015), Pd/Fe binary metals (Liao, Xu, & Li, 2009), Pd/C catalysts (Liao, Chai, & Xu, 2011), Pd–Ni/y-Al₂O₃ bimetallic catalysts (Zhang, Wang, Liao, Guo, & Shi, 2019), and Pd-Ni@SiO₂ bimetallic nano-catalysts (Liao, Wang, Zhang, Wang, & Guo, 2020) are employed to terminate such reactions during the bleaching process. Recently, a novel chlorine-free bleaching method using H₂O₂ as a bleach was developed to fabricate bleached shellac. H2O2 bleached shellac possesses better properties, including excellent solubility in alcohol, thermal stability, a higher acid value, and a lower softening point, than shellac that has been commercially refined using NaClO (Li, Zheng, et al., 2016). It is worth noting that the bleaching conditions of shellac, including solvent type, bleach concentration, and time, affect its final properties, including its color, acid value, insoluble solids, and polarity. Saengsod et al. found that the optimal conditions for the conventional bleaching of shellac were sodium carbonate as the solvent, 20 mL NaClO, and a 2-h bleaching time (Saengsod, Limmatvapirat, & Luangtana-anan, 2019). Both NaClO and H₂O₂ bleach can cause bleaching damage to shellac, but through different mechanisms, which can provide a scientific basis for the subsequent targeted regulation of bleaching damage (Li, et al., 2020).

In short, the renewability, improved extraction technologies, and continuous advancement of bleaching methods have promoted the use of shellac as a natural polymer with great potential. Coupled with its excellent performance, it is expected that shellac will be widely applied in the food industry in the future. However, the popularity of shellac applications in the food industry will result in a demand for improvements in quality as well as their quantity. Therefore, suitable tree species need to be selected and cultivated for the production of shellac-reproducing *Laccifer lacca*. Subsequently, sources of high yield and quality shellac with specific properties may be obtained by changing the diet and living environment of *Laccifer lacca*. Moreover, the shellac secretion mechanism requires clarification, with important guiding significance for various production needs. Undoubtedly, highly intelligent and efficient shellac extraction technology and equipment also require development, to meet increasing demand in the food industry.

3. Chemical composition and identification

Shellac is an amphiphilic biomacromolecule with a distinctive molecular structure consisting of aleuritic acid and cyclic terpene acids, as mentioned earlier (Fig. 1). Depending on the differences between the R and R' groups, the cyclic terpene acid moiety in shellac can be aleuritic acid, shellolic acid, jalaric acid, laccijalaric acid, laksholic acid, laccishellolic acid, or laccilaksholic acid. The aleuritic acid and cyclic terpene acids are linked by ester bonds and act as the hydrophobic and hydrophilic components of the shellac, respectively (Azouka et al., 1993; Luo et al., 2016). Some recent studies using modern detection techniques have also confirmed the recognized structure of shellac. Flow



Fig. 1. Generalized amphiphilic molecular structure of shellac. Reproduced from (Luo et al., 2016) with permission from American Chemical Society, Copyright 2016.

injection analysis and high-pressure liquid chromatography, coupled with electrospray ionization quadrupole-time of flight mass spectrometry, were used to analyze the chemical structure of shellac. Eight groups of compounds with increasing masses were detected and identified as free acids, esters, and polyesters with up to eight units. In some cases, highly detailed information on the ester linkages was provided by the mass spectra, enabling the different isomers to be distinguished (Tamburini, Dyer, & Bonaduce, 2017). Furthermore, a combination of these different analytical techniques could also be employed to further explore shellac structure in detail. The structures of six new shellolic esters and four known lac dyes from lac were identified using a combination of IR, HR-ESI-MS, 1D, and 2D NMR. The shellolic ester data obtained from NMR spectral analysis further clarified their structures (Lu et al., 2018). Obviously, the distinctive molecular structure of shellac provides the basis for its identification and determines its wide application, based on the structure-activity relationship.

The complexity of the molecular structure and composition of shellac can be identified by the chemical analyses and chromatographic and spectrometric methods listed in Table 1. The C=C group in shellac can be determined using FT-Raman spectroscopy to evaluate the natural waxes and resins embedded in shellac obtained from archeological materials (Edwards, Farwell, & Daffner, 1996). Interestingly, different shellac samples can be distinguished by evaluating the average ratios of terpenic acids, aleuritic acid, and other fatty acids, using reactive pyrolysis-gas chromatography (Py-GC) in the presence of tetramethylammonium hydroxide ((CH₃)₄NOH, TMAH) (Wang, Ishida, Ohtani, Tsuge, & Nakayama, 1999). Furthermore, butolic acid, aleuritic acid and its derivatives, and the typical sesquiterpenoid compounds in shellac can be identified by gas chromatography-mass spectrometry (GC-MS) to characterize fresh and aged shellac (Colombini, Bonaduce, & Gautier, 2003). Several different grades of shellac, mostly known as generic shellac, are commercially available. Their applications are mainly determined by the ester structure of the shellac molecules, without further differentiation. Generally, the different grades of shellac have different colors, glass transition temperatures, and pKa values. Notably, the single acids and polyesters in shellac can be observed via MALDI-TOF-MS to distinguish different grades of shellac (Buch et al., 2009). Furthermore, shellac that contains sesquiterpenic acid can be better detected by ATR-far-infrared region spectroscopy, which has high accuracy in distinguishing the sesquiterpenic, diterpenic, and diterpenic with polymerized communic acid and triterpenic resins (Prati, Sciutto, Mazzeo, Torri, & Fabbri, 2011). Although the development of chromatography, spectroscopy, and mass spectrometry technology has provided more accurate strategies for composition analysis, the lack of international uniform standards and methods for the identification of shellac hinders the expansion of its applications. More effort should be made to develop analytical technologies and establishing relevant standards for shellac in the food industry and other industries in the future.

Table 1

Identification approaches for different shellac-based samples.

| 11 | | L . | |
|---|----------------------------|---|----------------------------|
| Objects | Identification approach | Identification results | References |
| Natural waxes and resins including shellac | FT-Raman spectroscopy | Shellac has identifiable v (C==C) components, around 1649 $\rm cm^{-1}$. | Edwards et al. (1996) |
| Natural resin shellac | Reactive Py-GC | The average ratios among terpenic acids, aleuritic acid, and the other fatty acids were about 53:34:14 for Indian shellac and 51:35:14 for Thailand shellac. | Wang et al. (1999) |
| Fresh and aged shellac | GC-MS | Butolic acid, aleuritic acid, and its derivative acids, together with typical sesquiterpenoid compounds, were the main molecules identified and used as markers for the molecular recognition of the resin in fresh and aged shellac. | Colombini et al. (2003) |
| Different shellacs | MALDI-TOF-MS | The presence of a wide range of shellac components with groups of single acids at lower masses and polyesters up to tetramers at higher masses. | Buch et al. (2009) |
| Natural resins including shellac | ATR-FarIR spectroscopy | The vibrations δ CCC ²⁵ , δ CO ³¹ , τ (CCC), τ (CCO) ²⁴ and τ (CH ₃) ^{26,27} have been tentatively identified in shellac. | Prati et al. (2011) |

4. Physicochemical properties

4.1. Thermal properties

Shellac is a hard amorphous resin that contains small amounts of wax, yellow coloring matter, and odiferous matter. The color of shellac ranges from extremely light blonde to extremely dark brown, with many varieties of brown, yellow, orange, and red in between. The color is influenced by the sap of the tree from which the lac is harvested from and the time of harvest. Shellac is heavier than water with softening and melting temperatures of 65-70 °C and 75-80 °C, respectively (Azouka et al., 1993). It is worth noting that the softening and melting points are not distinct, unlike those of crystalline substances, leading to a gradual transformation of shellac from a solid to a liquid state. In particular, heating shellac above its melting point over an extended period may lead to not only a gradual loss of fluidity but also the formation of hard, horn-like, insoluble products, mainly after passing through the rubbery stage. This is because the presence of free hydroxyl and carboxyl groups in shellac make it highly reactive and capable of undergoing interesterification at elevated temperatures (>70 °C). The reaction results in evaporation and the formation of insoluble polymerized products. Similar thermal performance was also revealed in a later study using differential scanning calorimetry (Ruguo et al., 2011). Notably, heating the insoluble polymerized products in water can reverse the reaction (Azouka et al., 1993). A conceptual illustration of the transformational changes that occur during shellac processing under different temperature conditions is shown in Fig. 2. Shellac is polycrystalline, with a crystallite size of 1.3-4.0 nm, as determined by X-ray diffraction and high-resolution transmission electron microscopy. Furthermore, the growth of spherulites at different temperatures during the cooling of molten shellac confirmed the two-step crystallization of shellac, which can also be confirmed by differential scanning calorimetry (Mondal et al., 2020).

4.2. Solubility

Shellac is insoluble in water, glycerin, hydrocarbon solvents, and esters but soluble in alcohol and organic acids (Ghoshal, Khan, Gul-E-Noor, & Khan, 2009). The solubility parameter evaluated using various means (including solubility, structural contributions, and intrinsic viscosity), revealed that shellac is insoluble in weak H-bonded solvents but soluble in moderate to strong H-bonded solvents (Banerjee,

Srivastava, & Kumar, 1982). Based on their different solubilities in alcohol and water, shellac nanoparticles can be easily prepared by the anti-solvent precipitation method (Sedaghat Doost, Muhammad, Stevens, Dewettinck, & Van der Meeren, 2018). Because of its chemical structure, shellac is an amphiphilic substance, but it tends to be more hydrophobic owing to its oil-gelling ability (Patel et al., 2014). Furthermore, the functional groups (-OH, -COOH, -CHO) present in shellac contribute to intermolecular and intramolecular interactions via H-bonding or electrostatic interactions (Pearnchob, Dashevsky, & Bodmeier, 2004). Thus, shellac is capable of self-assembly into nanosized vesicles, which can further aggregate to fabricate the shellac hydrogel via cohesion between the vesicles. This phenomenon was attributed to cooperation between the hydrophobic interactions and hydrogen bonding capacity of shellac (Li, Pan, et al., 2016). Because of its carboxyl groups, shellac exhibits the attributes of a weak acid with a pKa of about 6, being dissolvable in water with a pH > 7. Under acidic conditions (pH < 7), the protonation of shellac hinders its dissolvability (Bellan, Pearsall, Cropek, & Langer, 2012). The preparation of an alkali (sodium) shellac solution using 1 mol/L Na₂CO₃ solution produces a shear-thickening suspension capable of changing from a Newtonian fluid to a continuous shear-thickening non-Newtonian fluid under a high shear rate. However, lower concentrations (0.1 and 1.0 wt %) of this alkali shellac solution produce a viscous fluid, while higher concentrations (5, 10, and 15 wt %) produce an elastic gel (Basu, 1950). At higher shear rates, low concentration shellac solutions form gels, while high concentration shellac gels are destroyed. This phenomenon relates to hydrophobic interactions between sodium clusters, as illustrated by the disappearance of this shear-thickening behavior following the addition of polyethylene oxide (Gao et al., 2018).

4.3. Structure modification

Naturally derived shellac experiences aging during storage, caused by intramolecular polymerization between carboxyl groups and hydroxyl groups, which significantly affects its molecular structure and properties (Coelho, Nanabala, Ménager, Commereuc, & Verney, 2012; Farag & Leopold, 2009). Thus, the modification of the shellac structure is crucial to achieve better functionality and exploit its application. The distinctive molecular structure of shellac eases its modification by chemical reactions. The application of plasticizers, such as polyethylene glycol (PEG), prevents reactions between active groups such as –OH, thereby enhancing their film stability (Khairuddin et al., 2016).



Fig. 2. Conceptual illustration of the changes in shellac processing under different temperature conditions.

Ammonium hydroxide and 2-Amino-2-methyl-1-propanol can form a salt with carboxyl moieties of shellac, which is advantageous for prolonging the shelf life of shellac films (Luangtana-anan et al., 2007). Furthermore, shellac succinate, which is synthesized with succinic anhydride via a dry media reaction, can be used to improve the properties of shellac. Increasing the number of succinate moieties in succinate-derived shellac significantly affects shellac properties, for example by increasing shellac solubility in solutions with pH < 7(Limmatvapirat et al., 2008). Similarly, shellac phthalate is synthesized through a solid-state reaction that involves grinding shellac with phthalate, followed by their thermal activation under various conditions. Compared to native shellac, shellac phthalate shows increased solubility in solutions with pH < 7, enhanced thermal stability, and anti-aging effects (Panchapornpon et al., 2011). Grafting 2-hydroxvethyl methacrylate monomers into shellac molecules using ultraviolet irradiation produces films with enhanced mechanical properties (Arnautov, Korhov, & Faitelson, 2013). Furthermore, shellac can also be PEGylated to synthesize shellac-PEG, which is effective in nanoparticle preparation via nanoprecipitation and nanoemulsion-solvent evaporation. The resultant nanoparticles possess a higher solid content and improved stability in salt solutions (Prawatborisut, Seidi, Yiamsawas, & Crespy, 2019). Incorporating hydroxypropylmethylcellulose (HPMC) and acrylic acid in shellac can produce a novel pH-responsive polymer via radical polymerization (Barik, Patnaik, Parhi, Swain, & Dey, 2017). Labuschagne et al. processed shellac in supercritical CO₂ using particles from a gas-saturated solution process. In this process, the shellac dissolves in the supercritical CO₂, and porous shellac structures can be formed by regulating the pressure and temperature during operation. Essentially, the supercritical CO₂ accelerates the esterification between -COOH and -OH, preventing post-processing 'aging' and enhancing the storage stability, solubility, and dissolution rates of the shellac (Labuschagne, Naicker, & Kalombo, 2016).

5. Applications of shellac in the food industry

Owing to its distinctive structure and properties, shellac is widely used in the food industry. Fig. 3 depicts the various possible applications of shellac, including as a raw material for fabricating food waxes, food coatings, and biodegradable films. It is also applied as a food foaming agent, oil-gelling agent, and food emulsifier. Furthermore, shellac is employed in the preparation of food delivery systems, such as microcapsules, coated carriers, nanofiber films, nanoparticles, and microparticles.

5.1. Food waxes

In the food industry, shellac has long been used as a wax to enhance the postharvest preservation of fruits and vegetables. The use of protective wax on some fruits and vegetables is an effective postharvest technique that maintains their quality and extends their shelf life. In principle, the preservative effects of these waxes on fruits and vegetables are achieved by reducing the gas transmission rate, preventing external mechanical damage, and shielding them from microorganism attack. In this context, shellac is applied during the production of commercial food waxes, or in combination with other waxes to maximize their preservative potential (Jo et al., 2014). Studies on the effects of shellac wax applications have been carried out on fresh apples, revealing improved color and reduced weight loss (Drake & Nelson, 1990). Similarly, Jo et al. found that a shellac-based wax covering on 'Fuji' apples improved appearance and sensory values, prevented physical damage, reduced weight loss (from 7.22% to 4.82%), and maintained hardness. Moreover, it inhibited microorganism growth (from 2.67 log CFU/g to 1.30 log CFU/g for total aerobic bacteria and from 2.31 log CFU/g to 1.58 log CFU/g for yeast and molds), thereby extending the shelf life of the apples (Jo et al., 2014). Compound shellac wax, used as a fruit preservative, also showed a preservation prolonging effect on mangoes by adjusting their respiration rate from approximately 160 to 80 ppm kg⁻¹ \cdot h⁻¹. In addition, weight loss was reduced from approximately 16%-8%, subsequently lowering the rate of decay from 85% to around zero (Fu et al., 2013). Similarly, commercial wax (polyethylene-shellac) was found to decrease weight and firmness losses, modify the internal atmosphere, and lower the respiratory rate of clementine mandarins (Contreras-Oliva, Perez-Gago, & Rojas-Argudo, 2012). In terms of corrosion protection, the addition of antifungal agents to shellac-based wax improved the quality of citrus fruits (Kouassi, Bajji, & Jijakli, 2012). During the processing of Spanish-style green olives (Manzanilla olives), the mechanically harvested fruits were stored under nitrogen, coated, and then stored again under nitrogen. Commercial wax (mainly shellac) used in coating the olives suppressed the enzymatic hydrolysate oxidation of oleuropein under anoxic conditions, thereby decreasing the development of brown spots on the olives (Ramirez, Sanchez, Romero, & Brenes, 2015).

Although shellac as a food wax has been widely used in the preservation of fruits and vegetables, much work can still be carried out to develop more functional shellac-based food waxes. Under the premise of ensuring safety and health, the introduction of other waxes into shellac is an effective means of enhancing its preservation effects. In addition,



Fig. 3. The applications of shellac in the food industry.

Table 2

| Coatings | Foods | Application effects | References |
|--|---|---|---|
| Shellac coating | Huanghua pears (<i>Pyrus</i> <i>pyrifolia</i> Nakai, cv. Huanghua) | Respiration rates and weight loss were reduced. Changes in the ripening parameters such as cell membrane permeability, texture profile analysis, soluble solids contents, titratable acidity (TA), ascorbic acid levels, and taste, were delayed. | Zhou et al. (2008) |
| Shellac coating | Chinese water chestnut (Eleocharis tuberosa) | 3. Metabolism was inhibited, resulting in better quality. Weight loss, cell membrane permeability, and decay rate increased. However, peroxidase, hardness, and quality parameters | Peng, Li, and Yin (2013) |
| Shellac coating | 'Valencia' oranges | Decreased. Weight and firmness losses were reduced. Fruit TA and ascorbic acid content were decreased, and pH, TPC, and TAC were increased. Fruit Paces was improved. | Khorram, Ramezanian, and Hosseini (2017) |
| Shellac coating | Apples | Weight loss, respiration rate, fruit softening, ripening index, color degradation, and polyphenol oxidase activity were reduced. | Ali, Kanwar, Yadav, Basu, and Mazumder (2019) |
| Shellac coating | Apples (Rich Red) | Weight loss, respiration, color degradation, fruit softening, and ripening index were significantly reduced compared to uncoated apples for up to 30 days of torage | Ali, Basu, and Mazumder (2020) |
| Shellac coating | Eggs | Interior qualities [Haugh units (72.37), weight loss (1.37), and viscosity (30.10 mPa s)] improved. | Yüceer and Caner (2020a) |
| Shellac coating | Eggs | Strength value increased (3.63 \pm 0.54 kgf) and shelf life was enhanced. | Yüceer and Caner (2020b) |
| Shellac coating (combine with hot water treatment) | Hami melon (Cucumis melo var. saccharinus) | Melon softening was reduced. Cell wall hydrolase activity, cell membrane leakage, fruit weight loss, and the loss of cell wall constituents were reduced. | Zhou et al. (2015) |
| Shellac-methylcellulose coating | Mangoes | Respiratory rate and the development of external and internal color were reduced. Loss of firmness was retarded. Changes in acids were delayed. Fruit maturation was delayed. Fither and layed layed layed. | HOA, DUCAMP, LEBRUN, and BALDWIN (2002) |
| Carnauba–shellac coating and shellac coating containing oleic acid, etc. | Apples (var. Delicious, Fuji, Braeburn, and Granny Smith) | Fruitlike and apple-like volatiles concentrations were increased. Fermentation product accumulation was reduced. Flavor synthesis rates were regulated and flavor loss rates were restricted. | Bai, Hagenmaier, and Baldwin (2002) |
| Commercial shellac-based coating | Mature oranges (Maroc late) | p-limonene, methyl anthranilate, and 3,7-dimethyl-2,6-octadienal content was reduced, and linalool content was increased. | Moussaid, Caillet, Nketsia-Tabiri, Boubekri, and Lacroix (2004) |
| Commercial shellac coating and shellac coating containing morpholine, oleic acid, etc. | Oranges, bell peppers, and apples | Ethane permeance of orange and apple peels was significantly reduced. Markedly high internal CO₂ and low O₂. Internal gas concentrations in different fruits with the same conting different loading to differences in quality. | Hagenmaier (2005) |
| Commercial shellac coating | Fresh eggs | Albumen viscosity was increased. Shell strength was increased. Quality was improved | Caner (2005) |
| Shellac-Aloe gel coating | Apple (var. Fuji) slices | Respiration rate, ethylene synthesis rates, and electrolyte leakage were reduced. Polyphenol oxidase and peroxidase activity were reduced. L*, a*, and b* values, firmness, and microbiological and keeping availity showed reduced changes during storage | Chauhan, Raju, Singh, and Bawa (2011) |
| Edible composite coating composed of hydroxypropyl methylcellulose, beeswax, and shellac | 'Clemenules' clementine mandarins | Green and blue molds were inhibited. Disease severity was reduced. Weight loss was reduced and the rind firmness was maintained. | Valencia-Chamorro, Palou, Angel del Rio, and Perez-Gago (2011) |
| Edible composite coating composed of hydroxypropyl methylcellulose, beeswax, and shellac | 'Valencia' oranges, and 'Ortanique' and 'Clemenules' mandarins | Incidence and severity of both molds were reduced. Fruit weight loss was reduced and firmness was maintained during cold storage. | Palou, Perez-Gago, and Valencia-Chamorro (2014) |
| Shellac-based coating containing starch, EDTA, and sodium alginate | Fresh green chilies | Shelf life was extended. Weight loss was reduced. | Chitravathi et al. (2014) |
| Shellac-Aloe gel coating | Tomato fruits | Respiration and ethylene synthesis rates were restricted. Shelf life was extended. | Chauhan et al. (2015) |
| Shellac-gelatin composite film coating | Banana fruits | Weight loss, softening, and the acid and sugar amount were slowly decreased, and fruit storage time was extended. | Soradech, Nunthanid, Limmatvapirat, and Luangtana-anan (2017) |
| Shellac coating containing carvacrol and thymol | 'Ruby Red' grapefruit | Lesion development inhibited by 24%–59%, compared to shellac alone. Fruit decay from natural infections and chilling injury were inhibited compared to shellac alone. | Yan, Zhang, Hu, Deng, and Ritenour (2020) |
| Shellac coating containing cinnamon essential oil | 'Thomson navel' orange fruit | Shellac coating (10%) enriched with 0.5% cinnamon essential oil reduced weight loss by 52%, firmness loss by 38%, and ascorbic acid content loss by 39%, compared to untreated oranges. Incorporation of cinnamon essential oil appeared to have good compared a potential to minimize graen mold by almost 90%. | Khorram and Ramezanian (2020) |

the introduction of hydrophobic phytochemicals is also expected to significantly enhance the preservation effect of shellac-based food waxes. Unfortunately, previous studies on shellac food waxes mainly focused on its postharvest preservation effects. In fact, the uses of shellac-based waxes for pre-harvest preservation, reducing pests and diseases and increasing yield, have more obvious significance. The impressive barrier and biocompatibility characteristics of shellac could protect the fruits and vegetables without posing a health threat.

5.2. Food coatings

Food coating provides preservative functions by controlling gas and water vapor exchange between the products and the surrounding environment. It also improves the mechanical properties or structural integrity of fresh food, consequently extending its shelf life (Chitravathi, Chauhan, & Raju, 2014). Usually, food coatings are composed of additives (if needed) and edible polymers including proteins and polysaccharides, that form a continuous network structure. Fortunately, shellac is as malleable as other polymers; therefore, it can also be used to prepare food coatings to extend the shelf life of food.

As shown in Table 2, shellac-based edible coatings effectively improved the quality of foods (such as fruits and vegetables), protecting them from the external environment, and consequently extending their shelf life, as mentioned earlier. Their food preservation mechanisms involve not only the effective inhibition of gas exchange in fruits and vegetables, but also the reduction of respiration intensity. Thus, nutrients loss was reduced as well as evaporative water loss. In addition, a full and fresh appearance and high hardness was maintained. Edible coatings protect fruits and vegetables from invasion by pathogenic bacteria, thereby avoiding losses through product decay. Nevertheless, it is worth noting that self-polymerization significantly decreases the mechanical properties and stability of pure shellac coatings. Incorporating aminecontaining substances such as Jeffamine® (Bar & Bianco-Peled, 2020) can prevent self-polymerization through the amine-carboxyl reaction, improving the properties of shellac coatings. In addition, the incorporation of functional ingredients in the edible coatings can further enhance their preservative effects. The use of shellac-based emulsions loaded with active substances (such as essential oils) to fabricate shellac-based food coatings is expected to easily improve their preservative capacity. In addition, the current application of shellac as a food coating is mainly limited to the preservation of fruits and vegetables. However, the fact that other waxes (such as carnauba wax) have been widely used as a food coating in confectionaries (such as soft candy and chocolate) and instant foods (such as bread and cake) (de Freitas et al., 2019) hints at the promising and extensive application for shellac-based food coatings in the future.

5.3. Biodegradable packaging films

Food packaging films have been widely used to protect food from environmentally induced deterioration. Shellac is a degradable filmforming agent with good barrier properties to achieve food protection. However, the poor mechanical strength of shellac films greatly limits their application. Fortunately, shellac modification or the preparation of shellac-based composite films addresses these shortcomings (Soradech, Limatvapirat, & Luangtana-anan, 2013). Alternatively, shellac is applied as a layer on other biodegradable films such as agar and cassava starch films, thereby enhancing the performance of these films (Phan The, Debeaufort, Luu, & Voilley, 2008).

Generally, composite films maximize the advantages of various components, which can be easily adjusted according to specific applications. They thus require extensive study. In the preparation of shellac composite films, the use of a polyethylene glycol 400 (PEG 400) plasticizer at a 20% w/w concentration effectively protects the active groups of shellac. This protection is provided via hydrogen bonds between the active components of shellac and the hydroxyl groups of PEG, thereby

enhancing the properties of shellac films (Luangtana-anan, Nunthanid, & Limmatvapirat, 2010). Soradech et al. found that increasing the gelatin content in shellac-gelatin composite films improved the strength, flexibility, and coating efficiency of the films on a hydrophilic substrate, and provided good storage stability (Soradech, Nunthanid, Limmatvapirat, & Luangtana-anan, 2012). Despite the improved storage stability of the composite films, the mechanical properties of the films deteriorated during storage, as revealed in later studies. Fortunately, the use of 5% and 10% w/w PEG 400 prevents the polymerization of shellac and improves the mechanical properties of composite films (Soradech et al., 2013). Similarly, PEG 400 and diethyl phthalate plasticizers were used to prepare shellac and pectin composites to overcome the shortcomings of pectin films, which include poor water retention, moisture resistance, and mechanical properties (Luangtana-anan, Soradech, Saengsod, Nunthanid, & Limmatvapirat, 2017). Du et al. fabricated konjac glucomannan-shellac composite films with improved hydrophilicity, good mechanical properties, and thermal stability, suggesting possible applications as biodegradable packaging (Du et al., 2019). Correspondingly, composite films prepared by blending shellac with soybean protein isolate showed significantly improved water resistance compared to pure soybean protein isolate films (Zhang et al., 2020). A new natural composite film was also recently prepared using carboxymethyl cellulose, gelatin, and shellac. The composite film showed good uniformity, tensile strength, antibacterial properties, elongation, air permeability, and thermal stability, low porosity, and an inclination toward particle aggregation (Mohamed, El-Sakhawy, Nashy, & Othman, 2019). In the presence of two emulsifiers, stearic acid and lauric acid, shellac was successfully doped into hydroxypropyl methylcellulose to form a composite film with enhanced properties (Byun, Ward, & Whiteside, 2012). Similarly, shellac, stearic acid, and Tween-20 were also used to improve the properties of pea starch-guar gum composite films (Saberi, Chockchaisawasdee, Golding, Scarlett, & Stathopoulos, 2017). In addition to being used as a raw material for the fabrication of composite films, shellac can also be applied as a separate layer to enhance the properties of films. The production of edible bilayer films, composed of agar or cassava starch as the cohesive structural layer and an ethanol-cast shellac layer as a moisture barrier to enhanced hydrophobicity, were reported by Phan The et al. These bilayer films tend to be cracked and scaled off, but the addition of PEG 200 could eliminate such defects (Phan The et al., 2008). In another study, wrapping deacetylated konjac glucomannan film in a shellac/stearic acid emulsion efficiently produced water-resistant konjac glucomannan film with improved water/moisture resistance and mechanical properties. Such films have great potential for use in food preservation (Wei et al., 2015). Recently, Ariyanto et al. developed a novel functional packaging by coating paper with a 1-methylcyclopropene controlled-release system that used shellac to wrap a 1-methylcyclopropene-loaded α-cyclodextrin. This functional packaging could reduce the ethylene production rate of apples and delay softening during storage (Ariyanto, Chiba, Oguma, Tatsuki, & Yoshii, 2019). Studies have also shown that shellac modification during the preparation of films usually results in better performance. In the process of maintaining or improving its biodegradability, Ghoshal et al., observed that grafting various acrylic monomers on shellac using y-rays yielded better quality shellac films with longer shelf lives (Ghoshal, Khan, Khan, Gul-E-Noor, & Chowdhury, 2010).

In recent years, the emergence of active and intelligent packaging systems that can monitor the quality of packaged food have gained wide attention (Bhargava, Sharanagat, Mor, & Kumar, 2020). The incorporation of naturally derived food pigments and food waste-based pigments, such as anthocyanins, curcumin, betalains, carotenoids, chlorophyll, brazilin, and quercetin, into shellac-based packaging films will provide additional benefits such as pH sensitivity, time-temperature sensitivity, CO₂ sensitivity, and ammonia sensitivity. Notably, this could provide a reference for the development of shellac-based intelligent biodegradable packaging films in the future.

5.4. Food delivery systems

Delivery systems can comprehensively regulate the distribution of bioactive components in the body at specific locations, times, and doses. They effectively improve the stability and bioavailability of bioactive components, and are thus widely used in the fields of medicine and food (Zhou, Cheng, Liu, Li, & Li, 2019). Currently, the delivery of bioactive compounds is still the subject of intensive research, and many delivery systems have been proposed, including microcapsules, coating materials, nanofibers, and nanoparticles. Oral application remains the simplest and most direct way to ingest active ingredients, attracting attention to food delivery systems. Generally, the materials used in food delivery systems must be safe and edible, limiting the use of irritant and toxic synthetic polymers and drawing increasing attention to natural products (Patel et al., 2013). Shellac is a non-toxic, biodegradable, film-forming, pH-responsive, water-resistant, and acid-resistant natural polymer (Limmatvapirat, Limmatvapirat, Puttipipatkhachorn, Nuntanid, & Luandana-anan, 2007; Tang, Hong, Li, & Huang, 2019) approved by the FDA as a food additive (E904) (Farag & Leopold, 2011). These advantages present shellac as an ideal raw material for food delivery systems.

5.4.1. Microcapsules

Microencapsulation technology has great advantages in embedding and delivering active substances. In recent years, the application of shellac in microcapsules has shown great potential in the fields of food and drug delivery, as summarized in Table 3. Shellac-based microcapsules have been developed using various techniques, including solvent evaporation (Kong et al., 2017), extrusion (Ben Messaoud et al., 2016; Patel et al., 2013), hot-melt extrusion (Gately & Kennedy, 2017; Khor, Ng, Kanaujia, Chan, & Dong, 2017), fluidized bed drying (Silva et al., 2016), and spray drying (Berg, Bretz, Hubbermann, & Schwarz, 2012; Oidtmann et al., 2012; Zhou et al., 2019). Many natural food-derived bioactive ingredients in the food industry, such as probiotics, polyphenols, natural pigments, and enzymes, are sensitive to processing. They are therefore encapsulated in microcapsules to enhance their stability (Ahn et al., 2018; Silva et al., 2016), mask their unpleasant taste (Khor et al., 2017), and control their release behavior (Henning, Leick, Kott, Rehage, & Suter, 2012). To create a final product that meets the expected requirements, appropriate core-shell materials and microencapsulation technology should be selected. Shell materials should have specific properties, such as biocompatibility, non-toxicity, stability, and appropriate permeability, and the core materials should differ according to the product requirements, as the product may be a solid, liquid drop, or dispersion (Bah, Bilal, & Wang, 2020). In all aspects of performance, shellac has obvious advantages for use in preparing food-grade microcapsules.

As mentioned earlier, shellac is a weak acid with a pKa <7, which can be dissolved in alkaline solutions to form ionized shellac. Ionized shellac is protonated by H⁺ and crosslinked by Ca²⁺, causing it to solidify and promoting the formation of microcapsules in acid and calcium chloride solution (Xue & Zhang, 2009). However, the carboxyl groups of shellac could lead to polymerization, resulting in poor stability and low permeability. Therefore, some researchers have attempted to combine shellac with other natural polymers (such as proteins and polysaccharides) to prevent polymerization and control the final physical and chemical properties (Berg et al., 2012; Yang et al., 2018). Patel et al. prepared gelatin-shellac microcapsules under acidic pH conditions using the acid resistance of shellac and the interaction between shellac and gelatin. They successfully encapsulated and released bioactive substances, flavor substances, colorants, flavorings, and immobilized enzymes (Patel et al., 2013). Pectin and alginate are capable of being crosslinked by calcium ions to form the "eggbox model" (Braccini & Pérez, 2001). A composite capsule wall composed of shellac, alginate, and pectin was applied as an outer membrane to form coated capsules. In the composite capsule, the diffusion effects of calcium and hydrogen

ions in the mixed solution of pectin and shellac affected the mechanical properties and thickness of the capsule membrane (Leick et al., 2011). The co-extrusion of sodium alginate-shellac and coconut fat was found to reduce the porosity of microcapsules, effectively protecting *Lactobacillus paracasei* BGP-1, which reached a 95% survival rate under simulated gastrointestinal conditions (Silva et al., 2016). Microcapsules with sodium alginate as the inner layer and shellac as the outer layer, especially after the crosslinking of shellac and calcium, showed the pH-dependent release of vitamins. As the heating temperature of the coated capsule was higher than the glass transition temperature of shellac, the vitamin retention efficiency also increased. This is because of the thermoplastic nature of shellac, which can form a viscous polymer precipitate at such temperatures, creating a dense network with a porosity lower than that of an unheated capsule (Ben Messaoud et al., 2016).

Apart from the traditional extrusion and fluidized bed technology, spray drying is a promising technology expected to be widely used in shellac-based microcapsule production because of its low cost and suitability for large-scale production. In addition, emerging microen-capsulation technologies, such as layer-by-layer self-assembly, electrospray, supercritical fluid, and microfluid technologies with the advantages of green health, energy-saving, environmental protection, and customizable preparation (Bah et al., 2020) can also be used in the further development of shellac-based microcapsules.

5.4.2. Enteric coating materials

The low solubility of shellac in the intestinal fluid (pH~6.8) limits its use as an enteric coating polymer. Plasticizers and water-soluble polymers can improve the enteric coating properties of shellac. Shellac enteric coatings containing 5% w/w glycerol, 5% w/w sodium alginate, or up to 20% w/w polyvinylpyrrolidone (PVP), protected probiotics from gastric juice and provided the best release characteristics in simulated intestinal juices (Stummer et al., 2010). Oehme et al. prepared acylated pectin beads coated with shellac and HPMC for colon-targeted anthocyanin delivery. The resistance of the beads in simulated gastric fluid was improved and the release rate reduced below 5% after the addition of the water-soluble polysaccharide HPMC to the shellac coating. Therefore, coating composition is essential to achieving drug stability in the gastrointestinal tract and the targeted release of anthocyanin (Oehme, Valotis, Krammer, Zimmermann, & Schreier, 2011). Shellac can also be used as a potential enteric coating for lactase microcapsules, which helps to reduce the effects of lactose intolerance when incorporated in milk formulations (Ahn, Kim, & Kwak, 2019; Ahn, Lee, & Kwak, 2019). Notably, the inter- and intra-individual variability in gastrointestinal pH, gastrointestinal environment, gastric emptying, and gastrointestinal transit time will inevitably affect the activity of bioactive substances coated with shellac-based enteric coatings. Thus, designing shellac-based enteric coatings targeting specific sites (such as the mucosa or receptor proteins) and combining them with the gastrointestinal environment is of great significance to improve the biological activity of active substances.

5.4.3. Nanofiber films

Electrospinning is a simple, effective, and top-down processing technology for the production of nanofiber films (Thammachat, Tabcharoen, & Soradech, 2013; Yu, Li, Zhang, & Williams, 2017). The nanofiber films prepared by this technology have the advantages of a large surface area to volume ratio, high porosity, and good fiber uniformity. These properties have important application value in the fields of medicine and food (Thammachat et al., 2010). Recently, some researchers fabricated shellac nanofiber films through electrospinning, and applied them as food-grade delivery systems, including targeted delivery (Wang, Yu, Li, Bligh, & Williams, 2015) and edible packaging materials (Li, Dong, et al., 2020). Shellac and its derivatives can easily form nanofiber films by electrospinning, owing to the high concentrations of shellac polymer chains generating entanglement which allows

Table 3

| Techniques | Wall materials | Core materials | Results | References |
|--|---|---|---|--|
| Extrusion | Shellac, alginate | CaCl ₂ , glycerol, riboflavin | The coated capsules crosslinked with calcium showed a pH-dependent release of the encapsulated vitamins. | Ben Messaoud et al. (2016) |
| | Shellac, gelatin | Bioactives (silibinin and epigallocatechin gallate)/flavor (D-Limonene)/food-grade colorants (curcumin, indigo carmine, and purpurin)/pH-sensitive dye (alizarin)/ immobilized enzymes (or-amylase) | The microcapsule has a range of bio-related applications, such as the encapsulation and release of bioactive components, flavor substances, colorants, flavorings, and immobilized enzymes. | Patel et al. (2013) |
| | Ammoniacal shellac, amidated pectin | CaCl ₂ , glycerol, bilberry extract | Shellac-coated pectin capsules prolonged drug release and reduced it from 100% to 5%. | Henning et al. (2012) |
| Co-extrusion + fluidized bed drying/ lyophilization | Shellac, alginate | Sunflower oil/coconut fat, <i>Lactobacillus paracasei</i> BGP-1 | The microcapsules were 0.71–0.86 mm in diameter, and coconut oil and fluidized bed drying were more effective at maintaining probiotic activity. More than 90% of the encapsulated BGP- 1 survived 60 days storage at 25 °C. Over 95% of the encapsulated BGP-1 survived under simulated gastrointestinal conditions. | Silva et al. (2016) |
| Extrusion/co- extrusion + fluidized bed drying | Shellac, alginate | Sunflower oil, Lactobacillus acidophilus LA3 | The viability of the co-extrusion-encapsulated LA3 using alginate or a blend of alginate-shellac was higher than extrusion encapsulated LA3 after 60 days storage. The alginate-shellac blend increased probiotics survival rates in simulated gastrointestinal fluids. | Silva et al. (2018) |
| Cold-set gelation | Shellac, oat protein | Oat protein, riboflavin/ <i>Lactobacillus acidophilus/</i> amylase | Oat protein-shellac beads prevented the early release of riboflavin in gastric juices and protected the enzymes and probiotics from gastric juices. The encapsulated riboflavin was sustainably released into the simulated intestinal tract. | Yang et al. (2018) |
| Fluidized bed technology | Shellac | Sweet whey powder, trehalos/sorbitol/ maltodextrin, <i>Lactobacillus reuteri</i> DSM 20016 | After storage at 4 °C for 28 days, the encapsulated bacteria remained stable. The final survival rate of the encapsulated probiotics was $76.74 \pm 24.36\%$ under simulated gastrointestinal conditions, while that of the free cells was $17.74 \pm 10.51\%$. | Schell and Beermann (2014) |
| | Shellac, sodium bicarbonate/citric acid, HPMC/pea starch, glycerol | Riboflavin 5'-monophosphate sodium salt dihydrate/nicotinic acid/nicotinamide | The acid sub-coating with citric acid counteracted the premature release of nicotinamide, and the alkaline sub-coating with sodium bicarbonate delayed the release of riboflavin 5'- monophosphate sodium salt dihydrate and nicotinic acid. | Theismann et al. (2019) |
| Spray drying | Shellac | Pectin, maltodextrin, citric acid, caffeine, blueberry extract | The higher the microcapsule water-binding capacity, the lower the anthocyanins release in simulated gastric juices. | Berg et al. (2012) |
| | Shellac, pectin amide, citric acid, maltodextrin | Bilberry extract | Microcapsules could not prevent the early release of anthocyanins in simulated gastric juices, but could protect them from the degradation in simulated intestinal juices. | Oidtmann et al. (2012) |
| | Shellac resin ammonium salts | Resveratrol | The microcapsule possessed sustained-release characteristics in a simulated gastrointestinal tract, enhancing the stability and antioxidant activity of resveratrol. | Zhou et al. (2019) |
| Emulsification/ external gelation | Alginate/whey protein isolate/shellac/sucrose | Limosilactobacillus reuteri TMW 1.656 | The addition of shellac to the microcapsules resulted in larger particle size, a much denser structure, lower hygroscopicity, and slower solubilization in water, which decreased probiotic inactivation after freeze drying and simulated digestion, heating, and ambient storage. | Huang et al. (2021) |
| Emulsification/ inverse ionotropic gelation | Alginate/shellac/ maltodextrin | Peppermint essential oil | The introduction of secondary excipients (shellac and maltodextrin) into the capsule shells affected the size, shell thickness, and swelling properties of the capsules, but did not affect their essential oil encapsulation efficiency. The addition of shellac resulted in a thicker shell, which protected the essential oil from oxidation, prolonging shelf life. | (Foglio Bonda, Regis, Giovannelli, & Segale, 2020) |
| Hot-melt extrusion | Shellac/carnauba wax/ zein | Quercetin | The order of the taste-masking effect of the microencapsulated powder observed was zein > carnauba wax > shellac. The dissolution rates of the carnauba wax- and shellac-microencapsulated powder were similar to that of pure quercetin under simulated gastrointestinal conditions. | Khor et al. (2017) |
| | Shellac | Bifidobacterium bifidum | At pH 1.2 and 6.8, the degradation rate of the samples was <5% in 24 h. At pH 7.4, the samples were completely degraded after 10–11 h. After storage for 5 months, probiotic viability had decreased by 57.8%. | Gately and Kennedy (2017) |

the maintenance of a continuous solution jet (Thammachat et al., 2010; Thammachat et al., 2013). The concentration of the shellac solution was identified as the key parameter that determines spinnability. In addition, electrospinning can be used to generate various complex nanostructures, using the macrostructure spinneret as a template. This has great potential for the development of advanced materials.

In coaxial electrospinning, the concentration of shellac solution used as the core fluid is important. For example, Wang et al. prepared ferulic acid-loaded shellac nanofiber films with a release rate of <10% in a pH 2.0 solution. However, upon transfer to a neutral phosphate buffer solution, the shellac molecules absorbed water, causing the fibers to swell and gradually expanding the dense structure. The drugs were then continuously released into the solution (Wang et al., 2015). In addition, Li et al. used coaxial electrospinning to prepare a PVP/shellac fiber membrane loaded with eugenol. Because the PVP/shellac fiber membrane was furnished with good thermal stability and moisture resistance, it inhibited strawberry rotting and prolonged their shelf life (Li, et al., 2020). Because of the low production efficiency of electrospinning in the fabrication of shellac-based nanofibers, the technology requires urgent advancement in functions such as ordinary multi-nozzle electrospinning and electrospinning without needles to expand shellac-based nanofiber applications in food and medicine.

5.4.4. Nanoparticles

Recently, increasing attention has been granted to the functionality of shellac as a base material for nanoparticles, especially in the field of embedding and delivering food-derived active substances. The application of shellac nanoparticles for the encapsulation of hydrophobic bioactive chemicals is not only effective in controlling their release but also improves their stability. For instance, the loading of curcumin (a natural yellow colorant) in shellac nanoparticles showed various advantages, such as biocompatibility and enhanced stability, which enriched the applications of this natural colorant in drinks, with appealing color and health benefits (Kong, Chen, et al., 2019). However, shellac nanoparticles tend to aggregate at acidic pHs, therefore stabilizers are required to overcome this shortcoming (Patel, Heussen, Hazekamp, & Velikov, 2011).

Several techniques have been employed for the fabrication of shellac nanoparticles (Table 4). Because of the significant difference in the solubility of shellac in alcohol and water, the anti-solvent precipitation method has become the most common technique for preparing shellac nanoparticles. Patel et al. reported an anti-solvent precipitation method that uses an ethanolic shellac solution with an aqueous natural polymer (xanthan gum) solution to prepare shellac nanoparticles. The average particle size of the nanoparticles was controlled (approximately 150-300 nm). Additionally, the pH-dependent release of shellac nanoparticles produced by the anti-solvent precipitation method was demonstrated by studying the release of silybin-loaded shellac nanoparticles in simulated gastrointestinal conditions (Patel et al., 2011). In a study conducted by Dewettinck's group, a shellac ethanol solution was added to an almond gum solution using the anti-solvent precipitation method to obtain shellac-almond gum nanoparticles. The nanoparticles showed good stability at pH 1.2, with diameters and encapsulation efficiencies of approximately 197 nm and 78%, respectively (Sedaghat Doost et al., 2019; Sedaghat Doost et al., 2018). Structurally, the almond gum was adsorbed onto the surface of the shellac nanoparticles, and quercetin was converted from a crystalline to an amorphous structure in the nanoparticles. Interestingly, they also found that the addition of polysorbate 80 (Tween-80) or Quillaja surfactants effectively reduced the particle size and increased the encapsulation efficiency. Uniform shellac-almond gum nanoparticles using Tween-80 as a stabilizer were obtained with an encapsulation rate >97%. Furthermore, the quercetin encapsulated in the nanoparticles showed stronger antioxidant capacity, less degradation, and higher absorption rates than free quercetin under gastrointestinal tract pH conditions (Sedaghat Doost et al., 2018). Moreover, cinnamon extracts encapsulated using shellac-xanthan gum

nanoparticles can effectively improve the antioxidant activity and physical stability of chocolate beverages (Muhammad et al., 2019; Muhammad et al., 2020). In our recent research, curcumin-loaded shellac nanoparticles prepared by the anti-solvent precipitation method were used as a filler to enhance the performance of chitosan films. The films effectively preserved shrimp, confirming the potential application of shellac nanoparticles as film fillers (Yuan et al., 2021). The properties of shellac-based nanoparticles were further enhanced by using the anti-solvent co-precipitation method to form dispersible zein-shellac composite nanoparticles (Chen et al., 2018). Similarly, Spasojević et al. encapsulated carvacrol in zein-shellac composite nanoparticles using the anti-solvent co-precipitation technique (Spasojević et al., 2020). The anti-solvent co-precipitation technique has also been used to encapsulate chlorophyll in shellac nanoparticles. The chlorophyll-loaded nanoparticles were incorporated into calcite crystals grown from an agarose gel media. They enhanced the stability and promoted the targeted intestinal release of chlorophyll (Kong, Jin, et al., 2019). Interestingly, Kong, et al. developed a versatile microfluidic strategy to incorporate natural colorants such as curcumin in shellac nanoparticles with controlled physicochemical properties (Kong, Chen, et al., 2019).

The ability of shellac to solubilize in water at various pHs, suggests the possibility of shellac nanoparticle preparation via other techniques; for example, shellac nanoparticles can be obtained by adjusting the pH of an aqueous solution. Shellac and rice protein biopolymers were dissolved at a pH of 12, followed by a pH adjustment to 7 to fabricate the nanoparticles. The obtained composite nanoparticles ranged from 50 to 500 nm in size and were capable of being induced by Ca^{2+} to undergo a structural transition to ribbons or networks. These structures have shown positive effects in encapsulating and delivering apigenin with controllable cellular uptake in a HepG-2 cell model (Wang, Yang, Feng, Wang, & Chen, 2020). Shellac nanoparticles with sizes ranging from 100 to 300 nm were also prepared by ionic crosslinking between the cationic molecules of chitosan and the anionic molecules of shellac. The obtained nanoparticles showed high encapsulation efficiency for bovine serum albumin (Kraisit, Limmatvapirat, Nunthanid, Sriamornsak, & Luangtana-anan, 2013). Studies have also shown that shellac submicron particles (approximately 720 nm) can be produced by electrospraying (Yu et al., 2013). Interestingly, oat protein-shellac nanoparticles could be prepared via water-in-water emulsion, followed by cold-gelation. The resultant particles have the capability to protect resveratrol during transportation through gastric fluid and control its release in the colon. Furthermore, these optimized nanoparticles could improve the cellular uptake, transport, and bioavailability of resveratrol and control the effects of resveratrol, preventing liver toxicity induced by CCl₄ (Yang et al., 2019).

5.4.5. Microparticles (beads/spheres)

Larger-sized or micro shellac particles (beads/spheres) are prepared by controlling the preparation conditions. Recently, shellac microparticles have shown great potential in the food industry, not only owing to their potential for the encapsulation and delivery of active substances, but also for their ability to form multiscale structures, consequently enhancing their encapsulation and delivery effects. Weitz's group fabricated shellac microparticles loaded with β -carotene using the microfluidic method. The microparticles were capable of not only retaining the health-promoting effects of the natural colorant but also providing its food additive functions, i.e., controlling the color of food. β-carotene was uniformly dispersed in densely packed shellac microparticles, reducing its exposure to oxygen and thereby preventing its oxidation. In addition, the color of the designed particles could possibly be tailored by selectively changing the concentration of β -carotene dispersed in the particle matrix, thereby enriching the edible colorants (Chen et al., 2017). Zein-shellac composite colloidal particles, prepared by the anti-solvent co-precipitation method, showed significantly higher encapsulation efficiencies for curcumin than pure zein particles. At a 1:1

Table 4

Techniques used in fabricating shellac nanoparticles.

| Fabrication methods | Additions | Inclusions | References |
|---|---|--------------|--|
| Anti-solvent precipitation | Xanthan gum | Silybin | Patel et al. (2011) |
| | | Cinnamon | Muhammad, Saputro, Rottiers, Van de Walle, and |
| | | extract | Dewettinck (2018) |
| | | Cinnamon | Muhammad et al. (2019) |
| | | Cinnamon | Muhammad et al. (2020) |
| | | extract | |
| | Almond gum, Surfactants (Polysorbate80, | Quercetin | Sedaghat Doost et al. (2018) |
| | sucrosemonopalmitate, Quillaja Saponin) | | |
| | Almond gum, Tween 80 | Quercetin | Sedaghat Doost et al. (2019) |
| | / | Curcumin | Yuan et al. (2021) |
| Anti-solvent co-precipitation | Zein | / | Chen et al. (2018) |
| | / | Chlorophyll | Kong, Jin, et al. (2019) |
| | Zein | Carvacrol | Spasojević et al. (2020) |
| Versatile microfluidic strategy | / | Curcumin | Kong, Chen, et al. (2019) |
| Adjusting the pH of the aqueous solution | Rice proteins | Apigenin | Wang et al. (2020) |
| Ionic cross-linking | Chitosan | BSA | Kraisit et al. (2013) |
| Electrospray | / | Ferulic acid | Yu et al. (2013) |
| Water in water emulsion followed by | Oat protein | Resveratrol | Yang et al. (2019) |
| cold-gelation | | | |

mass ratio of zein to shellac, the curcumin in the complex particles exhibited significantly improved photochemical and thermal stability (Sun et al., 2017). Mei et al. fabricated chitin-shellac composite microspheres based on the self-assembly of the polymers caused by thermally induced phase separation. The porous structure of the composite microspheres differed from that of the chitin microspheres, which made it possible to immobilize the *veast* alcohol dehydrogenase with a loading capacity of 79.0 mg/g (enzyme/carrier). Moreover, the immobilized enzymes exhibited catalytic activity comparable to their free counterparts (Mei et al., 2018). Obviously, particles with multiscale structures are more attractive in food applications. As such, sodium shellac was used to solidify a complex vitamin E (VE) emulsion to generate multiscale particles using the H⁺, Ca²⁺, and spray-drying methods. However, because of the high solid content of the nanoscale emulsion particles, they were difficult to wrap using the spray-drying method. The particles solidified by H⁺ showed higher VE loading and encapsulation efficiency than those of the other methods. These multiscale particles can be widely used in the medical, food, and cosmetic industries (Luo et al., 2016). Similarly, an emulsion-encapsulated alginate-shellac composite formed microbeads with high oil encapsulation efficiency under Ca²⁺ induction. The microbeads prepared under optimized conditions, effectively encapsulated sunflower oil and controlled its release in the intestine. This system has important implications for the design of oil-encapsulated and oil-soluble compound delivery systems (Morales et al., 2017). Most of the above-mentioned micro-scale shellac particles prepared from pure shellac or shellac complexes with polymers are only used to encapsulate a single active compound. However, multiple active compounds incorporated in the micro-scale shellac particles would exhibit more interesting characteristics; therefore, shellac-based microparticles with hierarchical structures, such as dimer particles, trimer particles, and core-shell particles, are expected to act as carriers for multiple active compounds.

5.5. Food foaming agents

Food foaming agents have been widely used in the food industry to enhance the sensory properties of foods. Studies have reported that micro-rod shellac-based particles have stronger foam-stabilizing capabilities than traditional foaming agents (Campbell, Holt, Stoyanov, & Paunov, 2008; Campbell, Stoyanov, & Paunov, 2009). To expand the application of shellac in the food industry, Campbell et al. fabricated novel micro-rods containing a food-grade shellac wax and ethyl cellulose composite using shear emulsification in combination with solvent abrasion technology (Campbell et al., 2008). The edible micro-rods revealed good adsorption and fusion capabilities at gas-liquid interface, which enhanced the stability of gas bubbles in foam. Changes in the shear rate and polymer concentration of the dispersed phase could control the diameter and aspect ratio of the micro-rods. Moreover, the morphology of the ethyl cellulose micro-rods can be transformed, from flat ribbons to having long strings through the micro-rods, by changing the polarity of the solvent (Campbell et al., 2008). To further enhance the foam-stabilizing abilities of these micro-rods and explore their potential applications, Campbell et al. incorporated solid particles, including sporopollenin, silica (with a porosity of approximately 95%), hydrated baker yeast cells, and sodium bicarbonate micro-crystals into the micro-rods. The presence of lumpy inclusions in the micro-rods reduced the foam drainage rate, which led to higher foam stability. The attained stability is attributed to the entanglement of the lumpy micro-rods, as compared to smooth rods at the foam film surfaces. The lumpy micro-rods adsorbed at the foam film surface, as well as those accumulated in the Gibbs Plateau borders, obstructed the flow, retarding liquid drainage (Campbell et al., 2009). Notably, the incorporation of flavorants and functional substances in the shellac micro-rods may expand the applications of shellac-based food foaming agents to include enriching the taste, adjusting the apparent, increasing creativity, and extending the storage period of food.

5.6. Oil-gelling agents and emulsifiers

The preparation of oleogels by solidifying liquid edible oils has gradually attracted the interest of food researchers in recent years (Patel & Dewettinck, 2016). Patel et al. found that shellac can be potentially applied in the fabrication of oleogels with different textures and thermal properties by altering their concentrations (Patel et al., 2013). In a later study, it was reported that the oleogels resulted from the physical entrapment of liquid oil in crystal networks of shellac. This was achieved by cooling the hot oil-shellac dispersions to room temperature. In addition, the oleogels can be used in the fabrication of emulsifier-free emulsions with temperature-dependent melting behavior and excellent storage stability (Patel et al., 2013). Interestingly, by regulating the water content, the oleogels can be used in the preparation of edible emulsifier-free emulsions, including structured water-in-oil (W/O) emulsions (spreads), oil-binder replacements in chocolate paste formulations, and a shortening alternative for cake preparation (Patel et al., 2014). The properties of the shellac oleogels were significantly different from those of HPMC, which are known as high internal phase emulsion

oleogels. In short, the shellac oleogels exhibited pseudolipid structural properties owing to the reversible melting and crystallization of their wax crystals. The recovery of the shellac oleogels was significantly better than that of HPMC oleogels. Moreover, shellac oleogels showed good resistance to the incorporation of water and could therefore form stable emulsifier-free W/O emulsions, even at a water inclusion level of 60% by weight (Patel & Dewettinck, 2015). These emulsions were formed via the adsorption of shellac crystals at the oil-water interface to stabilize the droplets. Oil-in-water emulsions with pH-responsive switchable behavior were also prepared using a complex formed by the strong interactions between xanthan gum and shellac, which showed Pickering properties and had a strong stabilizing effect on the emulsion (Patel et al., 2013). Interestingly, Weitz's team fabricated novel biocompatible amphiphilic alginate/shellac particles using the microfluidic method to form colloidal surfactants that were able to stabilize both water-in-oil and oil-in-water emulsions (Chen et al., 2017). Similarly, biocompatible amphiphilic dimer particles consisting of a hydrophobic PLA bulb and hydrophilic shellac-PEG bulb were fabricated through controlled co-precipitation and phase separation. The easily adjustable size and ratio of the dimer particles made it possible to stabilize different types of Pickering emulsions, while the pH-responsive properties of shellac enhanced the characteristics of the entire emulsions (Sun et al., 2020).

Oil-gelling agents and emulsifiers have been the attractive applications of shellac in the food industry in recent years. However, apart from being used as a solid fat replacement, the application of shellac-based oleogels still requires expansion to achieve higher value addition. Compared to shellac-based oleogels, shellac-based emulsions are still underexplored. This may be because shellac is much more hydrophobic than hydrophilic. Therefore, the hydrophilicity of shellac needs to be regulated in future studies on shellac-based emulsions.

6. Health considerations

Shellac has been used in agriculture, medicine, and various other fields in the past few decades, increasing their chances of coming into contact with humans. Therefore, it is important to take cognizance of its health and safety features. Fortunately, shellac has been recognized as safe (GRAS status) by the US FDA since 1939 (Hagenmaier & Shaw, 1991), which has been supported by subsequent research.

Shellac used in cosmetics as a coating material is often topically applied, coming into contact with human skin without causing irritation (Rademaker, Kirby, & White, 1986). Shellac F, a new shellac-based varnish containing 5% sodium fluoride, possesses excellent cell compatibility and desensitization effects (Hoang-Dao et al., 2008). In addition, studies have also shown that shellac solutions can be used to embolize gangrenous tissue in selected diabetic patients (Alzahrani, Bedir, & Al-Hayani, 2013). In vitro experiments showed that it is impossible for shellac to be digested by the enzymes in simulated gastric and intestinal fluids (Kunkel & Seo, 1994). In the colon, shellac may be hydrolyzed by the intestinal environment and microorganisms, indicating the safety importance of its hydrolysate. Lu et al. evaluated the bioactivities of sesquiterpene acids, including seven sesquiterpene acids isolated from shellac. Cytotoxic and antibacterial tests were used to evaluate their bioactivities, revealing the absence of cytotoxic activity in the compounds. Shellolic acid A also exhibited inhibitory activity against Bacillus subtilis with an MIC value of 0.1 mg/mL (Lu et al., 2014). In their later studies, it was observed that 10 compounds isolated from a methanolic extract of Laccifer lacca secretions revealed the absence of cell growth inhibition and, concurrently, five compounds were found to be significantly active against some bacteria. Their study also revealed that lac resin is edible, non-toxic, and physiologically harmless at levels presently employed as excipients (Lu et al., 2018). In conclusion, to date, few studies have focused on the possible health risks of shellac; however, these studies revealed that shellac does not pose any health risk.

7. Conclusion and future perspectives

The numerous advantageous properties and potential applications of shellac have gained much recognition, especially in the food industry. Its origin as a natural resin, extracted from a wide range of sources distributed in the forests of several Southeast Asian countries, confirms its renewability. Currently, the relevant extraction technology is mature and, as such, the possibility of large-scale production exists. As introduced earlier, shellac has distinctive molecular structure, which is responsible for its exceptional properties, including its film-forming ability, pH-responsiveness, hydrophobicity, degradability, and biocompatibility. Importantly, shellac is a green label product, i.e., it is a nonbiotoxic polymer whose safety has been screened through cell experiments. These advantages, coupled with the current health theme awareness, have resuscitated the glory and popularity of shellac once overshadowed by synthetic polymers (which resulted from their advanced preparation technology). The applications of shellac in the food industry (as well as the medical and pharmaceutical industries) have advanced into various technologies, including food waxes, food coatings, biodegradable films, food-grade delivery systems, food foaming agents, oil-gelling agents, and emulsifiers. Moreover, shellac-related technological advancements in recent years have also captured their application in producing pH-responsive Pickering emulsions as functional foods and nanoparticles as fillers and carriers of hydrophobic substances, which can enhance the functionality of edible films and hydrogels, among other applications. Subsequently, with the advent of more research focusing on remedying the stability of shellac, there is no doubt that shellac has a chance of gaining even more applicability in the food industry. On this note, some new reagents, methods, and applications need to be further developed.

Acknowledgments

This work was supported by the National Key R&D Program of China (No. 2016YFD0400203).

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