



Review

Functional Properties of Biopolymer-Based Films Modified with Surfactants: A Brief Review

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Abstract: An increase of environmental awareness recently has increased the interest of researchers in using of biopolymer-based films. The films have been prepared extensively by utilizing starch, carboxymethyl cellulose, chitosan, protein, gelatin, carrageenan, alginate, pectin, guar gum and pullulan. They are typically modified with surface-active agents (surfactants) such as glycerol monostearate, sucrose ester, sodium stearyl lactate, sodium dodecyl sulfate, ethyl lauroyl arginate HCl, Span 20 to 80, Tween-20 to 80 and soy lecithin for improving the functional properties of the films. In this brief review, two types of biopolymer-based films that prepared through casting method were categorized, specifically solution- and emulsion-based films. The four types of surfactants, namely non-ionic, anionic, cationic and amphoteric surfactants that are regularly used to modify biopolymer-based films are also described. The functional properties of the films modified with different types of surfactants are briefly reviewed. This study enhances the attraction of researchers in biopolymer-based films and the improvement of new concepts in this niche area.

Keywords: surfactant; biopolymer; film; functional properties

1. Introduction

Today—due to environmental concerns and growing of the biodegradable plastic industry—the use of biopolymer-based films in the food industry and food packaging has become a trend. The bio-based resources are also contributing to the growth of the industry since they are sustainable and renewable supplies. Biopolymers are polymeric biomolecules obtained from natural sources. Starch was commonly used for the preparation of biopolymer-based films [1,2]. Starch can be obtained from corn [3–6], potato [7–9], cassava [10,11], tapioca [12,13], sorghum [14], loquat seed [15], kudzu [16], wheat [6] and yellow pea [17]. On the other hand, carboxymethyl cellulose (CMC) [18,19], chitosan [20–24], protein [25–31], gelatin [32–36], carrageenan [37,38], pectin [39], guar gum [17,40] and pullulan [41] (chemical structures displayed in Figure 1) are also utilized in preparation of biopolymer-based films because of they are biodegradable, renewable, low-cost and abundance. The films also have moderate mechanical properties such as tensile strength and elongation at break [42]. However, they have high water vapor permeability character [43] and poor moisture barrier property [44].

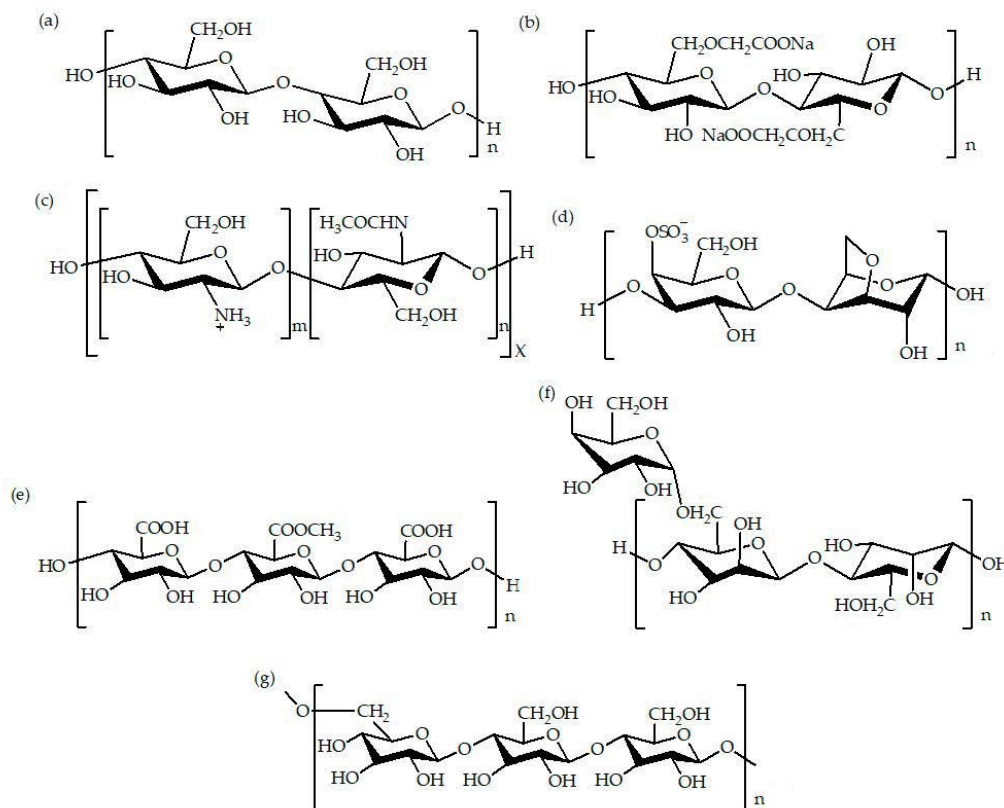


Figure 1. Chemical structures of (a) starch, (b) carboxymethyl cellulose (CMC), (c) chitosan [45], (d) carrageenan, (e) pectin, (f) guar gum and (g) pullulan.

Surface-active agents (surfactants) such as glycerol monostearate [3,5,46], sucrose ester [1,39,47], sodium stearyl lactate [39], sodium dodecyl sulfate [10], ethyl lauroyl arginate HCl [48,49], Span 20 to 80 [2,5,6,16,50,51], Tween-20 to 80 [11,15,18,20,22,37,41,52] and soy lecithin [2,27,28,31,43,53] (chemical structures indicated in Figure 2) are usually used as modifier for the biopolymer-based films. Surfactants possessed an amphiphilic character that has hydrophilic and hydrophobic properties [12]. The modification of the films by surfactants not only decreases water vapor permeability [24], but also improve the moisture barrier property of the films [51].

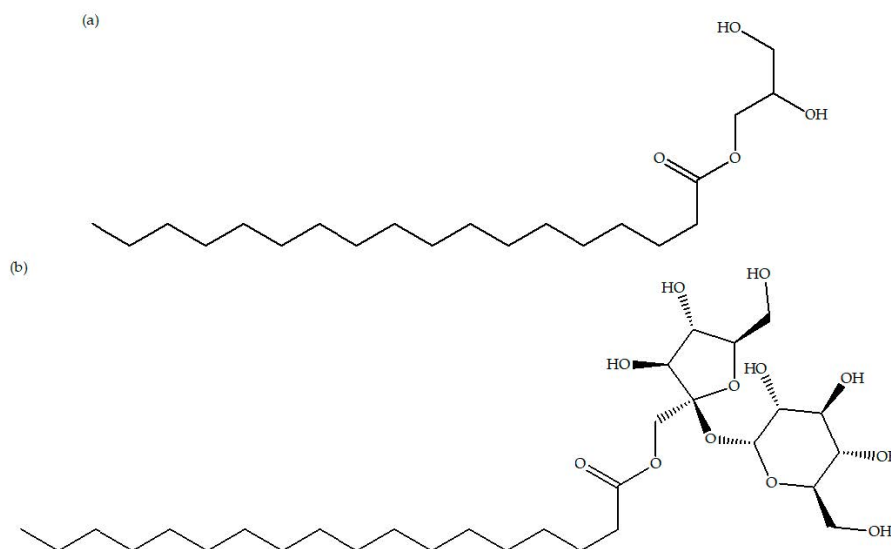


Figure 2. Cont.

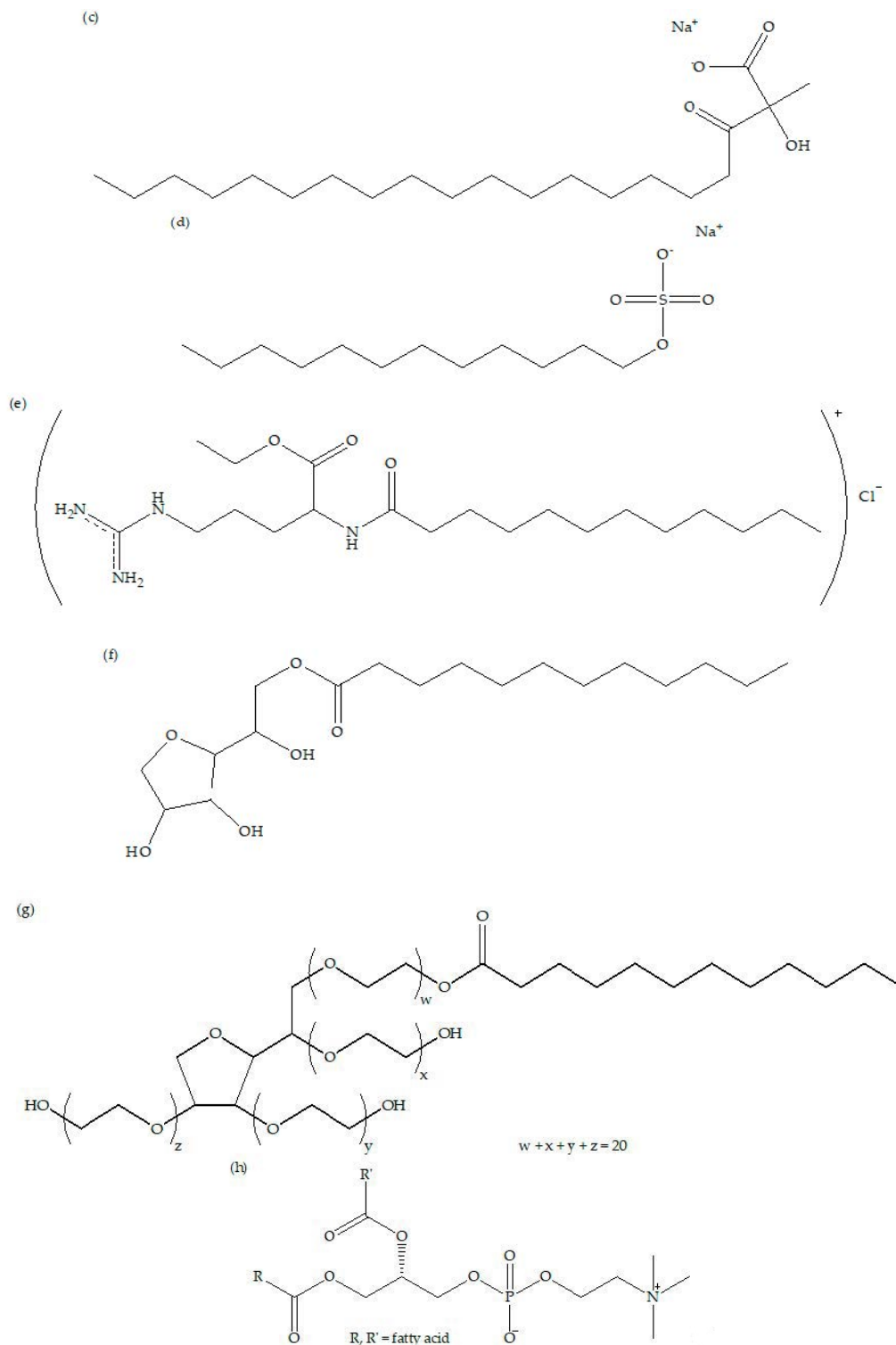


Figure 2. Chemical structures of (a) glycerol monostearate, (b) sucrose ester, (c) sodium stearoyl lactate, (d) sodium dodecyl sulfate, (e) ethyl lauroyl arginate HCl, (f) Span 20, (g) Tween-20 and (h) soy lecithin.

In the last two decades, much research has been conducted with the objective of improving the functional properties (e.g., mechanical properties (tensile strength, elongation at break), water affinities (water vapor permeability, moisture content, solubility, swelling), contact angle, thermal stability, oxygen permeability and opacity) of biopolymer-based films by utilizing surfactants. Moreover, some works have concentrated on the emulsification for the preparation of biopolymer-based

films. Nevertheless, to the best knowledge of the authors, no brief review has been created comprising the study on the preparation and modification of biopolymer-based films. This is the purpose of creating an organized review in this study.

2. Types of Biopolymer-Based Films

2.1. Solution-Based Films

Solution-based films are commonly prepared by dissolving a water-soluble biopolymer in the solvent such as water and heating at elevated temperatures with stirring to obtain a biopolymer solution. The attained solution is considered as film-forming solution (FFS) [1,6,33,35,54]. The solution casting technique is conducted by pouring the FFS into a flat-surfaced mold and drying in an oven or room temperature to acquire free-standing film [55]. The solution-based films are easy to prepare, and they are only required inexpensive tools. Table 1 shows the types of biopolymers, additional components and processing parameters of the films. The additional components such as glycerol, sorbitol and glucose (chemical structures exhibited in Figure 3) are typically added into the biopolymer solution to act as plasticizers for the prepared biopolymer-based films [4,9,28]. On the other hand, acetic acid and glucomannan function as a dissolving agent and a thickening agent for chitosan and pectin, respectively.

Table 1. Types of biopolymers, additional components and processing parameters of the solution-based films.

Biopolymer	Additional Component	Processing Parameter	References
Kudzu starch	Glycerol	3.0% (<i>w/v</i>) of kudzu starch, 0.9% (<i>w/v</i>) of glycerol, 100 mL of water, stirring at temperature of 100 °C.	[16]
CMC	Glycerol	2.5 g/L of CMC, 5 g/L of glycerol, 350 mL of water, stirring at temperature of 60 °C.	[56]
Chitosan	Acetic acid, glycerol	1 g of chitosan, 1 g of acetic acid, 20 g of glycerol, 100 mL of water, stirring at room temperature (25 ± 1 °C).	[24]
Gelatin	Glycerol	3.5 g of gelatin, 30 wt% of glycerol, 90 mL of water, stirring at temperature of 70 °C.	[57]
Pectin	Glucomannan	1% (<i>w/v</i>) of pectin, 0.75% (<i>w/v</i>) of glucomannan, 100 mL of water, stirring at room temperature (25 ± 1 °C).	[39]
Guar gum	Glycerol	0.3 g of guar gum, 25 wt% of glycerol, 100 mL of water, stirring at temperature of 40 °C.	[17]
Pullulan	Glycerol	2% (<i>w/v</i>) of pullulan, 15 wt% of glycerol, 100 mL of water, stirring at temperature of 45 °C.	[41]
Loquat seed starch	Sorbitol	3.5% (<i>w/v</i>) of loquat seed starch, 45% (<i>w/w</i>) of sorbitol, 100 mL of water, stirring at temperature of 95 °C.	[15]
Starch/chitosan	Glycerol, glucose	1.5% (<i>w/v</i>) of starch/chitosan, 1% (<i>w/v</i>) of glycerol or glucose, 100 mL of water, stirring at temperature of 90 °C.	[9]

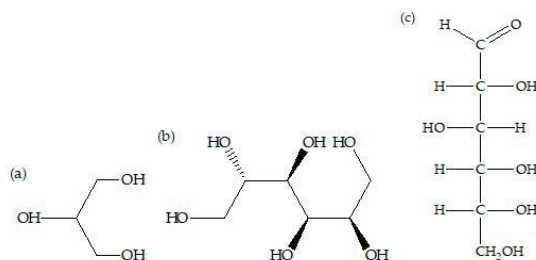


Figure 3. Chemical structures of (a) glycerol, (b) sorbitol and (c) glucose.

2.2. Emulsion-Based Films

Emulsion-based films are generally prepared by making FFS first (as described in Section 2.1). After this, oil containing surfactant is made by inserting surfactant into the oil and then it is added into the FFS, followed by homogenizing to attain film-forming emulsion (FFE) [8,35,36,43,50]. The film is obtained by casting the FFE into glass plates and drying at elevated temperature or ambient temperature [21], unless stated differently in Table 2. Table 2 indicates the types of biopolymers, plasticizers, oils, surfactants and processing parameters of the films. Unlike solution-based films, emulsion-based films require oil for example, palm oil, beeswax, candelilla wax, virgin coconut oil, cinnamon bark oil and soybean oil to form emulsions [1,21,34,44,55]. The presence of hydrophobic substance such as oil can increase the elongation at break and decrease the water vapor permeability of the films [35,36]. The surfactants such as soy lecithin, sucrose ester (sucrose stearate), sodium dodecyl sulfate, Tween and Yucca schidigera extract (YSE) frequently modified the biopolymer emulsions by stabilizing the emulsions and retain homogeneity of the films [15,19]. Moreover, the emulsion-based films have lower water vapor permeability and higher elongation at break than the solution-based films as control film [8,29,36,41,43,54].

Table 2. Types of biopolymers, plasticizers, oils, surfactants and processing parameters of the emulsion-based films.

Biopolymer	Plasticizer	Oil	Surfactant	Processing Parameter	References
Fish gelatin	Glycerol	Palm oil	Soy lecithin	3.5% (<i>w/v</i>) of fish gelatin, 10% (<i>w/w</i>) of glycerol, 100 mL of water, 25% (<i>w/w</i>) of palm oil, 12.5% (<i>w/w</i>) of soy lecithin, homogenizing at 22,000 rpm for 3 min at 70 °C, casting the FFE onto a plastic Petri dish and air-blowing for 12 h prior to drying at 25 °C and 50% ± 5% RH for 48 h in an environmental chamber.	[34]
Tapioca starch/decolorized hsian-tsao gum	Glycerol	Beeswax	Sucrose stearate	2% (<i>w/v</i>) of biopolymer blend, 10 wt% of glycerol, 100 mL of water, 10 wt% of beeswax, 10-wt% sucrose stearate, homogenizing at 10,000 rpm for 3 min at 95 °C, pouring the FFE onto a level circular Petri dish and drying in an environmental chamber at 50 °C and 58% RH for 1 h and sequentially at 25 °C for 96 h.	[1]
CMC, potato starch soy protein, pork gelatin	Sorbitol	Candelilla wax	Tween-40	5% (<i>w/w</i>) biopolymer, 3% (<i>w/w</i>) of sorbitol, 100 mL of water, 0.5% (<i>w/w</i>) of candelilla wax, 0.35% (<i>w/w</i>) of Tween-40, homogenizing at 20,000 rpm for 3 min at 90 °C, casting the FFE onto leveled polycarbonate tray and drying at 25 ± 1 °C for 24 h.	[56]
Carrageenan	Glycerol	Palm oil	Tween-20, Tween-40, Tween-80	1% (<i>w/v</i>) of carrageenan, 50% (<i>w/w</i>) of glycerol, 100 mL of water, 3% (<i>v/v</i>) of palm oil, 0.1%–0.5% (<i>v/v</i>) of surfactant, homogenizing at 13,500 rpm for 3 min at 80 °C, casting the FFE at the center of a circular glass plate and drying at 30 °C for 48 h.	[37]
Soy protein	Glycerol	Virgin coconut oil	YSE	6.5% (<i>w/w</i>) of soy protein, 20% (<i>w/w</i>) of glycerol, 100 mL of water, 0.99%–6.54% (<i>w/w</i>) of virgin coconut oil, 9%–23% (<i>w/w</i>) of YSE, homogenizing at 20,500 rpm for 2 min at 70 ± 3 °C, dispersing the FFE on acrylic plates and drying at room temperature (25 ± 3 °C) for 24 h.	[44]
Chitosan	Glycerol	Cinnamon bark oil, soybean oil	Tween-80	2% (<i>w/w</i>) of chitosan, 20% (<i>w/w</i>) of glycerol, 100 mL of water, 1%–3% (<i>w/w</i>) of cinnamon or soybean oil, 40% (<i>w/w</i>) of Tween-80, homogenizing at 7000 rpm for 2 min at 21 °C, casting the FFE onto glass plate and drying at ambient conditions (21 °C) for 24 h.	[21]

Table 2. Cont.

Biopolymer	Plasticizer	Oil	Surfactant	Processing Parameter	References
Gelatin	Glycerol	Palm oil	Tween-80	3.9 wt% of gelatin, 33 wt% of glycerol, 100 mL of water, 36 wt% of palm oil, 20 wt% of Tween-80, homogenizing at 12,000 rpm for 3 min at 60 °C, casting the FFE evenly over a rimmed acrylic plate and drying at 25 °C in a convection chamber for 48 h.	[36]
Soy protein	Glycerol	Virgin coconut oil	Soy lecithin	6.5% (w/w) of soy protein, 20% (w/w) of glycerol, 100 mL of water, 0.99%–6.54% (w/w) of virgin coconut oil, 9%–23% (w/w) of soy lecithin, homogenizing at 20,500 rpm for 2 min at 70 ± 3 °C, pouring the FFE on acrylic plate and drying at 25 ± 3 °C for 24 h.	[27]
Fish gelatin	Glycerol	Palm oil	Tween-20, soy lecithin, sodium dodecyl sulfate	3.7 wt% of fish gelatin, 30 wt% of glycerol, 90 mL of water, 50 wt% of palm oil, 25 wt% of surfactant, homogenizing at 22,000 rpm for 3 min at 70 °C, casting the FFE onto a rimmed silicone resin plate and air-blowing for 12 h at 28–30 °C prior to further drying at 25 °C and 50% ± 5% RH for 24 h in an environmental chamber.	[43]

3. Modification by Different Types of Surfactants

3.1. Types of Surfactants

Generally, surfactants are classified into four types: non-ionic, anionic, cationic and amphoteric. The classification is based on the polarity of the surfactant head group, for example, non-ionic, anionic, cationic and amphoteric or zwitterionic. There is no charge on a head group of non-ionic surfactants, while anionic and cationic surfactants have negative and positive charges on their head groups, respectively. On the other hand, there are both negative and positive charges for the amphoteric surfactants. Table 3 displays the types of surfactants and film-forming used in the preparation of biopolymer-based films. The non-ionic surfactants such as Span and Tween are widely used in the preparation of solution- or emulsion-based films [2,5,55,56]. Moreover, another non-ionic surfactant, for instance, sucrose ester has also been used for the preparation of such films [1,51]. Instead, some anionic surfactants, for example, sodium stearyl lactate and sodium dodecyl sulfate are commonly employed in the preparation of solution-based films by using gelatin and starch as polymer matrices [10,39]. The cationic and amphoteric surfactants such as ethyl lauroyl arginate HCl and soy lecithin could be utilized for both solution- and emulsion-based films [28,35,48,58].

Table 3. Types of surfactants and film-forming used in the preparation of biopolymer-based films.

Surfactant	Type of Surfactant	Film-Forming	References
Span 20 to 80	Non-ionic	Solution/Emulsion	[2,5,16,47]
Tween-20 to 80	Non-ionic	Solution/Emulsion	[20,21,37,55]
Sucrose ester	Non-ionic	Solution/Emulsion	[1,51,59]
Sodium stearyl lactate	Anionic	Solution	[39]
Sodium dodecyl sulfate	Anionic	Solution	[10,30]
Ethyl lauroyl arginate HCl	Cationic	Solution/Emulsion	[48,49,58]
Soy lecithin	Amphoteric	Solution/Emulsion	[2,28,35,43]

3.2. Modification by Non-Ionic Surfactants

Table 4 shows examples of non-ionic surfactants, hydrophilic–lipophilic balance (HLB) values, alkyl chain length and functional properties of biopolymer-based films. Most of the non-ionic surfactants such as Span, Tween and sucrose ester are used in the preparation of biopolymer-based films [1,5,6]. Span and Tween with high HLB values have slightly shorter alkyl chain-length than low HLB values,

whereas sucrose ester that have the same alkyl chain length possess different HLB values (Table 4). The HLB reveals the attraction of surfactant to water or oil. The HLB values can be calculated via Davies' method as in the equation below:

$$\text{HLB} = 7 + \sum(\text{hydrophilic group numbers}) + \sum(\text{lipophilic group numbers})$$

For example, Span 40 or sorbitan monopalmitate (chemical structure presented in Figure 4).

$$\text{HLB} = 7 + \sum(\text{group number of sorbitan}) + \sum(\text{group number of CH}_2 \text{ and CH}_3)$$

$$\text{HLB} = 7 + (6.8) + (15 \times -0.475)$$

$$\text{HLB} = 7 + (6.8) + (-7.125)$$

$$\text{HLB} = 6.675$$

$$\text{HLB} \approx 6.7$$

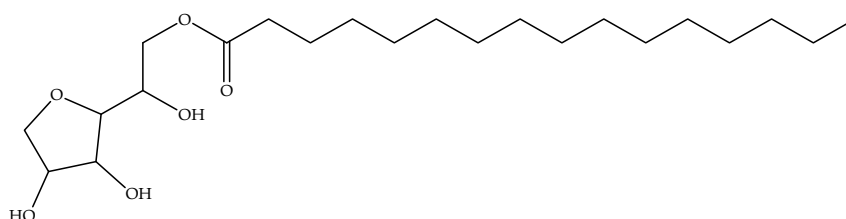


Figure 4. Chemical structure of Span 40.

Table 4. Examples of non-ionic surfactants, hydrophilic–lipophilic balance (HLB) values, alkyl chain length and functional properties of biopolymer-based films.

Non-Ionic Surfactant	HLB Value	Alkyl Chain Length	Biopolymer	Film-Forming	WVP	TS	EAB	Op	References
Span 40	6.7	C ₁₆	Corn starch	Solution	↓	↓	↓	↓	[5]
Span 80	4.3	C ₁₈	Corn/wheat starch	Emulsion	↓	↓	↑	↑	[6]
Tween-20	16.7	C ₁₂	Kudzu starch	Solution	↓	↓	↑	n/a	[16]
Tween-80	15.0	C ₁₈	Corn/wheat starch	Emulsion	↓	↓	↓	↑	[6]
Sucrose ester (S-1570)	15.0	C ₁₈	Tapioca starch/decolorized hsian-tsao gum	Solution	↓	↓	↓	↓	[1]
Sucrose ester (S-1170)	11.0	C ₁₈	Tapioca starch/decolorized hsian-tsao gum	Emulsion	↓	↓	↓	↑	[1]

WVP—water vapor permeability, TS—tensile strength, EAB—elongation at break and Op—opacity; The symbol ↑ corresponds to an increase in the properties and ↓ a decrease in the properties while “n/a” means “not available”.

The higher the HLB values are, the larger the attraction to water is and the smaller the HLB values are, the larger the attraction to oil is [20]. Span 40 has an HLB value of 6.7 which is higher compared to Span 80 which has an HLB value of 4.3, thus, Span 40 was frequently used in the preparation of solution-based films [5], whereas Span 80 is considered as hydrophobic surfactant, it was utilized in the creation of emulsion-based films [6]. On the other hand, Tween-20 (has an HLB value of 16.7) and Tween-80 (has an HLB value of 15.0) have also been used in solution- and emulsion-based films, respectively [6,16]. The same trend was observed for sucrose ester as well, whereby sucrose ester with high HLB value was utilized in the preparation of solution-based films and vice versa [1].

Table 4 also shows that Span 40 has modified the FFS of corn starch in the preparation of the corn starch-based films [5]. The modification by Span 40 improves the functional properties such as water vapor permeability (WVP) and opacity (Op) of the films. The WVP of the films decreases by up to 30% compared to the film without Span 40. This is attributed to the enhancement in the tortuosity factor for mass transport in the corn starch [5]. In addition, the transparency of the films is slightly decreased by up to 0.8% due to the existence of distributed surfactant aggregates, with a dissimilar refractive index, which increases the light scattering effect. The lower transparency suggests that the films are opaquer. Moreover, the tensile strength (TS) and elongation at break (EAB) of the films decrease by up to 57% and 12%, respectively compared to the film without Span 40. This is because of the creation of an additional anisotropic structure with decreased cohesion forces [5]. On the other hand, Span 80 modifies the FFE of corn/wheat starch containing essential oil in the preparation of the

emulsified corn/wheat starch films [6]. The modification by Span 80 improves the functional properties such as WVP, EAB and Op of the films. The WVP of the films decreases by up to 17% compared to the corn/wheat starch film. This is attributed to Span 80, which is a hydrophobic surfactant with lower HLB value and cannot interact with water or corn/wheat starch [6]. Moreover, the EAB of the films slightly increases by up to 7.3% also because of the hydrophobicity of Span 80 which can have less effect on the intermolecular hydrogen bonding within starch–starch, consequently resulting in the increase of EAB. Furthermore, the Op of the films is significantly increased by up to 186% due to the hydrophobicity of Span 80 as well, which cannot lead to uniform structure with corn/wheat starch. However, the TS of the films decreases by up to 26% compared to the corn/wheat starch film. This result proves that Span 80 plays a major impact on the reduction of TS value [6].

Tween-20 modifies the FFS of kudzu starch in the preparation of the kudzu starch-based films [16]. The modification by Tween-20 has improved the functional properties such as WVP and EAB of the films. The WVP of the films is slightly decreased by up to 7% compared to the film without Tween-20. The low decrease of WVP is attributed to the inherent hydrophilic property of Tween-20 [16]. In addition, the EAB of the films is significantly increased by up to 65% due to Tween-20 acted mechanically as plasticizer. However, the TS of the films is decreased by up to 35% compared to the film without Tween-20, this behavior is similar with the high flexibility films [16]. On the other hand, Tween-80 modifies the FFE of corn/wheat starch containing essential oil in the preparation of the emulsified corn/wheat starch films [6]. The modification by Tween-80 improves the functional properties such as WVP and Op of the films. The WVP of the films decreases by up to 11% compared to the corn/wheat starch film. The low decrease of WVP is attributed to Tween-80 is a hydrophilic surfactant with higher HLB value and can interact with water or corn/wheat starch [6]. In addition, the Op of the films has significantly increased by up to 193% due to the hydrophilicity of Tween-80 as well, which can lead to uniform structure with corn/wheat starch. However, the TS and EAB of the films are decreased by up to 28% and 12%, respectively compared to the corn/wheat starch film. This is because Tween-80 interacts with corn/wheat starch which weakens the intermolecular hydrogen bonding, subsequently resulting in the decrease of mechanical properties [6].

Sucrose ester (S-1570) or sucrose stearate modifies the FFS of tapioca starch/decolorized hsian-tsao gum in the preparation of the starch/gum-based films [1]. The modification by S-1570 has only improved the functional properties such as WVP of the films. The WVP of the films is decreased by up to 54% compared to the film without S-1570. This is attributed to the hydrogen bonding interaction between starch/gum and polar groups of S-1570, which reduces the number of polar groups free to interact with water molecules [1]. Nevertheless, the Op of the films is slightly decreased by up to 5.2% due to S-1570 can dissolve in FFS with individual molecules or micelles of the nanometers size and interact with amylose of starch and gum. Additionally, the TS and EAB of the films have decreased by up to 48% and 19%, respectively compared to the film without S-1570. This is because S-1570 has an HLB value of 15, has more hydrophilic functional groups to interact with tapioca starch and hsian-tsao gum and hindered the interaction between starch and gum chains [1]. On the other hand, sucrose ester (S-1170) modifies the FFE of tapioca starch/decolorized hsian-tsao gum containing beeswax in the preparation of the emulsified starch/gum films [1]. The modification by S-1170 has improved the functional properties such as WVP and Op of the films. The WVP of the films is decreased by up to 24% compared to the starch/gum film. This is attributed to the presence of beeswax, which also enhances the water barrier property. Moreover, the Op of the films has significantly increased by up to 155% because of the formation of beeswax globules of the micrometers size in the FFE during the drying process [1]. Nevertheless, the TS and EAB of the films have decreased by up to 55% and 30%, respectively compared to the starch/gum film, therefore the existence of beeswax did not give benefits on improving the mechanical properties.

3.3. Modification by Ionic Surfactants

There are two types of ionic surfactants specifically anionic and cationic surfactants. They are usually employed in solution-based films through the preparation of FFS. Table 5 demonstrates the examples of ionic surfactants, HLB values, alkyl chain length and functional properties of biopolymer-based films. Most of the ionic surfactants such as sodium stearyl lactate, sodium dodecyl sulfate and ethyl lauroyl arginate HCl have been used in the preparation of biopolymer-based films [10,39,49]. Table 5 also shows that the longer the alkyl chain length is, the lesser the HLB values are, in contrast to non-ionic surfactants. Sodium stearyl lactate has an HLB value of 8.3, it has longer alkyl chain length than sodium dodecyl sulfate which has an HLB value of 40. In addition, ethyl lauroyl arginate HCl also has the same alkyl chain length as sodium dodecyl sulfate and it has an HLB value of 16, which is higher compared to sodium stearyl lactate. On the other hand, sodium stearyl lactate and sodium dodecyl sulfate were frequently used in the preparation of solution-based films [10,39], whereas ethyl lauroyl arginate HCl could also be utilized in the creation of both solution- and emulsion-based films [49,58]. Therefore, the use of ionic surfactants in the preparation of biopolymer-based films can be associated with their HLB values.

Table 5. Examples of ionic surfactants, HLB values, alkyl chain length and functional properties of biopolymer-based films.

Ionic Surfactant	HLB Value	Alkyl Chain Length	Biopolymer	Film-Forming	WVP	TS	EAB	Op	References
Sodium stearyl lactate	8.3	C ₁₈	Bovine skin Gelatin	Solution	↓	↓	↑	↑	[39]
Sodium dodecyl sulfate	40	C ₁₂	Cassava starch	Solution	↓	↑	↓	↑	[10]
Ethyl lauroyl arginate HCl	16	C ₁₂	Gelatin	Solution	↓↑	↑	↑	↓	[49]
Ethyl lauroyl arginate HCl	16	C ₁₂	Chitosan	Emulsion	↑	↓	↑	n/a	[58]

WVP—water vapor permeability, TS—tensile strength, EAB—elongation at break and Op—opacity; The symbol ↑ corresponds to an increase in the properties and ↓ a decrease in the properties while “n/a” and ↓↑ mean “not available” and “unchanged”; respectively.

Table 5 also displays that sodium stearyl lactate has modified the FFS of bovine skin gelatin in the preparation of the gelatin-based films [39]. The modification by sodium stearyl lactate has improved the functional properties such as WVP, EAB and Op of the films. The WVP of the films has decreased by up to 68% compared to the film without sodium stearyl lactate. This is attributed to the negatively charged hydrophilic part is electrostatically interacted with the positively charged amino acid residues in gelatin [39]. In addition, the EAB of the films has slightly increased by up to 2.6% due to the presence of interactions between sodium stearyl lactate and gelatin chains. The Op of the films has slightly increased by up to 8.3% because sodium stearyl lactate has caused the component rearrangement in the gelatin film during the drying process. However, the TS of the films has slightly decreased by up to 6.8% compared to the film without sodium stearyl lactate, this is common behavior for films with high EAB. On the other hand, sodium dodecyl sulfate has modified the FFS of cassava starch in the preparation of the cassava starch-based films [10]. The modification by sodium dodecyl sulfate has improved the functional properties such as WVP, TS and Op of the films. The WVP of the films has decreased by up to 10% compared to the film without sodium dodecyl sulfate. This is attributed to the increased interaction between sodium dodecyl sulfate and starch which decreased the free channel for the transit of water vapor. In addition, the TS of the films has significantly increased by up to 1179% due to sodium dodecyl sulfate could form rigid complexes with amylose and amylopectin of the starch molecular chains with higher rigid and tension [10]. However, the EAB of the films has decreased by up to 39% because of the tensile strength extremely increased in starch films in the presence of sodium dodecyl sulfate. In addition, the Op of the films has increased by up to 192% compared to the film without sodium dodecyl sulfate, this is due to sodium dodecyl sulfate and starch have more interactions between molecules, which provided the starch with greater continuity and fewer empty spaces, resulting in a film that blocked more light path [10].

Ethyl lauroyl arginate HCl modifies the FFS of gelatin in the preparation of the gelatin-based films [49]. The modification by ethyl lauroyl arginate HCl has improved the functional properties such as TS and EAB of the films. The TS of the films has slightly increased by up to 4.3% compared to the film without ethyl lauroyl arginate HCl. Moreover, the EAB of the films has also slightly increased by up to 7.5%. The slight increase may be due to the low content of ethyl lauroyl arginate HCl modified the films [49]. Furthermore, the transparency of the films has slightly increased by up to 6.3% because of the good compatibility between film components. The higher transparency suggests that the films are less opaque. Nevertheless, the WVP of the films has not been influenced by ethyl lauroyl arginate HCl [49]. On the other hand, ethyl lauroyl arginate HCl modifies the FFE of chitosan containing cinnamon oil in the preparation of the emulsified chitosan films [58]. The modification by ethyl lauroyl arginate HCl has only improved the functional properties such as EAB of the films. The EAB of the films has increased by up to 169% compared to the chitosan film. This is attributed to the uniform dispersion of positively charged ethyl lauroyl arginate HCl compound in the chitosan matrix [58]. However, the TS of the films has significantly decreased by up to 53% due to the distributed small droplets of cinnamon oil, which have disrupted the chitosan matrix. Moreover, the WVP of the films has decreased by up to 39% compared to the chitosan film. This is because of the modification by ethyl lauroyl arginate HCl may split hydrogen bonding and interrupt the ordered structures of chitosan molecules, resulting in the raised WVP of the films [58].

3.4. Modification by Amphoteric Surfactant

Amphoteric or zwitterionic surfactants are also ionic surfactants, but they have both negative and positive charges. Soy lecithin or L- α -phosphatidylcholine is one of the amphoteric surfactants that has regularly been modified the biopolymer-based films [43]. Soy lecithin is a byproduct of soybean oil processing [27]. The chemical structure of soy lecithin is shown in Figure 2h, it consists of phosphate and quat groups (hydrophilic head groups) and two fatty acid groups (lipophilic tail groups) [43]. Soy lecithin has an HLB value of 4.0, and it is a predominantly hydrophobic surfactant [34]. It was frequently utilized in preparation of emulsion-based films [27,31,35,43,60,61]. Nevertheless, the soy lecithin can also be employed in the creation of solution-based films [2,28,53]. Table 6 indicates the functional properties of biopolymer-based films modified with soy lecithin. The biopolymers, for instance, soy protein and fish gelatin have been used for the preparation of emulsion-based films, whereas pig hide gelatin and potato starch utilized for the creation of solution-based films. From Table 6, it can be seen that the modification by soy lecithin has decreased the WVP of the films, this is because the soy lecithin is principally had lipophilic behavior, which could reduce the amount of water vapor to pass through the films. On the other hand, the TS of the films has also decreased with the modification by soy lecithin. However, the presence of soy lecithin has increased the EAB of the films, this is due to soy lecithin has behaved as plasticizer or lubricant in the films, causing an improvement of flexibility and the decrease of TS [27]. In addition, the films modified with soy lecithin also have high Op, especially for emulsion-based films, this is caused by the light-scattering effect of oil droplets, which is distributed throughout the films [43,61].

Table 6. Functional properties of biopolymer-based films modified with soy lecithin.

Biopolymer	Film-Forming	WVP	TS	EAB	Op	References
Soy protein	Emulsion	↓	↓	↑	↑	[27]
Fish gelatin	Emulsion	↓	↓	↑	↑	[43]
Fish gelatin	Emulsion	↓	↓	↑	↑	[61]
Pig hide gelatin	Solution	↓	↓	↑	↑	[53]
Pig hide gelatin	Solution	↓	↓	↑	↑	[28]
Potato starch	Solution	↓	↓	↑	n/a	[2]

WVP—water vapor permeability, TS—tensile strength, EAB—elongation at break and Op—opacity; The symbol ↑ corresponds to an increase in the properties and ↓ a decrease in the properties while “n/a” means “not available”.

4. Conclusions

Biopolymer raw materials, processing parameters and types of biopolymer-based films modified with surface-active agents (surfactants) have been briefly reviewed in this study. The main functional properties, for example, water vapor permeability, tensile strength, elongation at break and opacity of the films have also been identified in this brief review. Surfactants have frequently modified the biopolymer-based films because they have amphiphilic character. Surfactants employed for different types of biopolymers are mostly based on HLB value and their chemical structures. Non-ionic and amphoteric surfactants have been the two most important surfactants for the biopolymer-based films. Non-ionic surfactants provide a wide range of HLB values and various alkyl chain lengths. An amphoteric surfactant such as soy lecithin is a promising alternative to synthetic surfactants because it is a byproduct of soybean oil processing. It also has the capability to use in both solution- and emulsion-based films. The films modified with soy lecithin have great functional properties such as low water vapor permeability, high flexibility and opacity. This brief review may be useful for the commercialization of bio-based, low-cost, environmentally friendly films for numerous applications.

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