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THE POTENTIAL OF CHITOSAN-STARCH BLEND POLYMERS AS EDIBLE COATINGS FOR THE PRESERVATION OF FRUITS AND VEGETABLES: A MINI-REVIEW

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Abstract. *Fruits and vegetables are essential for human life. Maintaining the quality and safety of fruits and vegetables are critical challenges for the food packaging industry in this decade. Chitosan and starch are most widely used for food packaging as they bring unique properties for packaging applications. Chitosan has good antimicrobial properties but weak water vapor permeability. Meanwhile, starch has a better water vapor barrier but has poor mechanical properties. This paper discusses the application of chitosan and starch blends in the field of films and coatings. Several characteristics, such as mechanical, thermal, antimicrobial properties, solubility, and water vapor permeability, are also discussed. This review also discussed several applications of chitosan-starch-based films and coatings for the preservation of fruits and vegetables.*

Keywords: *Chitosan; fruit; preservation; starch; vegetable*

1. Introduction

Fruits and vegetables are important elements of the human diet as they provide the nutrition required by the human body. However, fresh fruits and vegetables are prone to damage during harvesting and postharvest handling such as sortation, storage, and transportation. Although the produce has been picked from the plant, the respiration and transpiration processes continue and cause water and nutrient loss [1]. Moreover, fresh produce is vulnerable to microbial growth, responsible for fruit and vegetable spoilage. Some freshly harvested fruits and vegetables, known as climacteric, also continue ripening after harvest, which increases the risk of microbial infection and spoilage [2]. These factors reduce the quality of the product and its safety, leading to food loss and economic losses.

One of the postharvest techniques to extend the storage period and maintain the safety of fresh produce is through edible coatings. An edible coating (EC) is a thin layer of edible material applied on the surface of fruits and vegetables. ECs act as an extra layer that coats the stomata,

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reducing respiration and transpiration rates. This reduction lowers the produces' weight and nutrient loss due to the EC's semipermeable barrier properties [1, 3]. This layer is developed with biodegradable and non-toxic material from various biopolymers, such as polysaccharides, proteins, and lipids [4].

Chitosan is one of the most used polysaccharides in fruit and vegetable coating applications. Chitosan is derived from deacetylated chitin, a major biopolymer of crustacean shells such as shrimp, crab, and crawfish [5]. This material is non-toxic, biodegradable, has good film-forming ability, and is antimicrobial [6]. Furthermore, chitosan has a positive charge that allows it to adhere to biological surfaces, resulting in stable films with good gas permeability and mechanical properties [7]. Several studies reported that chitosan coatings reduced the respiration rate and weight loss of fruits and vegetables, such as bananas [8], sweet cherries [9], pomegranates [10], tomatoes [11], and bell peppers [12]. However, chitosan has low solubility in water and organic solvents [13]. Despite its edible characteristics, its poor solubility makes chitosan coatings difficult to remove from fruits and vegetables before consumption.

On the other hand, starch is also a polysaccharide with potential edible coating applications for fruits and vegetables due to its non-toxic, filmogenic capacity, abundant availability, renewability, flexibility, and low cost [1], [14], [15]. Thakur *et al*., [16] reported that rice starch-based coating controlled the respiration and ripening of banana fruit and resulted in lower weight loss and improved firmness. A similar result was also reported by Francisco *et al*., [17], acetylated cassava starch-based film reduced the weight loss and increased the firmness of guava fruit. However, starch has major drawbacks, such as poor mechanical properties and low thermal and moisture resistance [18]–[20]. Therefore, this paper aims to review the potential of the chitosan-starch blend edible coating to preserve fruit and vegetable properties in the postharvest stage.

2. Chitosan-Starch Compatibility

The main challenge of the chitosan and starch blend is their compatibility. Each material has different characteristics due to its chemical nature. Chitosan is derived from chitin and is composed of repeating 1,4 linked 2-deoxy-2-aminoglucose units. The amino group NH_2 in this polysaccharide can be turned into NH_3^+, forming electrostatic interactions with anionic groups in an acidic environment [21]. Despite these properties, chitosan was considered a distinguished substitute for synthetic polymeric material due to its nontoxic, biodegradable, biofunctional, biocompatible, and antimicrobial characteristics [22].

Chitosan is reported to have good compatibility with starch. Xu *et al*., [21] reported that adding starch in a small volume fraction did not influence the two chitosan crystalline peaks in its

X-ray diffraction. The good interaction between chitosan and starch was also found in the Fourier Transform Infrared (FTIR) spectroscopy result. The mixing resulted in the shift of the amino peak of chitosan, which indicates the interaction of the hydroxyl groups in starch with the amino groups in chitosan. Moreover, Mathew and Abraham [23] stated that starch has a nonionic property that makes it compatible with chitosan.

3. Chitosan-Starch Blend Polymer Formulations

The most common method for developing new polymeric materials is to mix at least two polymers. Blending the chitosan with other polymers can change the functional properties of the final product [24]. Solution casting is the most used method to fabricate chitosan and starch film and coatings among all fabrication methods. The main difference of film and coating preparation is the drying process. In film fabrication, solution is casted and dried to get dried film. Meanwhile in coating fabrication, solution is applicated to obect by dipping, spraying or other methods. Table 1 shows recent work on chitosan-starch blend film. For example, Zheng *et al*., [25] dispersed chitosan in an acetic acid solution and used glycerol as the plasticizer. About 1.5% w/v chitosan was dispersed in 1% v/v acetic acid solution at 30°C for 4 to 6 h. Acorn starch at a concentration of 1.5% w/v was gelatinized in distilled water at 95°C for 20 min. Next, various fractions of gelatinized starch were mixed into 100 mL of chitosan solution. Wang *et al*., [26] also reported a similar method with the chitosan-corn starch blend. The mixture was cast in glass plate molds before being dried in the oven at 50°C. A schematic diagram of common chitosan-starch blend polymer film fabrications is shown in Figure 1.

Table 1. Recent studies on chitosan-starch blend polymers

Main Polymer	Starch	Other polymers	Final product	Reference
Chitosan	Acorn starch	-	Film	[25]
Chitosan	Corn starch	-	Film	[21], [26], [27]
Chitosan	Corn starch	-	Layered film	[28]
Chitosan	Pea starch	-	Film	[29]
Gelatin	Chitosan	Corn starch	Layered film	[30]
Chitosan	Unspecified starch	-	Film	[31]
Corn starch	Chitosan	-	Film	[32]
Purple yam starch	Chitosan	-	Film	[33]
Chitosan	Water chestnut starch	-	Coating	[34]
Chitosan	Cassava starch	-	Film	[35]
Chitosan	Sugar palm starch	-	Film	[36]
Chitosan	Banana starch	-	Coating	[37]

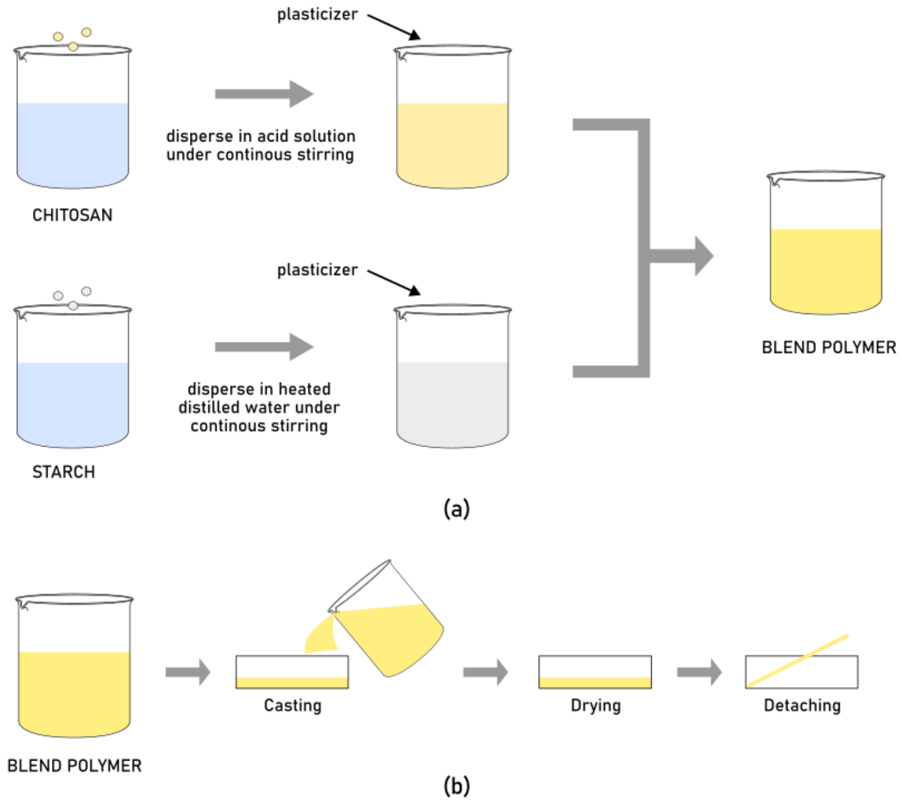

Figure 1. Schematic diagram of chitosan-starch blend polymer film fabrication, (a) blending chitosan and starch solution (b) film casting and drying

4. Chitosan-Starch Blend Polymer Properties

4.1. Mechanical properties

Mechanical properties are important parameters in film development. Several studies reported that adding gelatinized starch affects the mechanical properties of polymer blends. The addition of starch into chitosan results in a film with moderate tensile strength (TS) than pure chitosan film. This phenomenon occurs because of starch has lower crystalline structure compared to chitosan [35]. In addition, the reduction in TS was likely due to the effect of inter-molecular hydrogen bonds between the chitosan NH_3^+ group and starch OH^- [21]. However, TS can be increased by adding a plasticizer. As reported by Sun *et al*., [27], the addition of a chitosan slurry into a thermoplastic starch (starch + plasticizer) increased the TS by about 73% compared to a mixture of chitosan and starch slurry without any plasticizer. The plasticizer plays an important role as an adjuster of the molecular order of the film to change its compactness [26].

Blending starch to chitosan also affects the elongation at break (EB) of resulting films. EB indicates the flexibility of the films, which is an important characteristic in packaging applications. The presence of starch decreased the EB of the blend polymer film due to its brittleness. Zheng *et al*., [25] found that the EB of resulting films decreased with the addition of a starch solution concentration into a chitosan solution. This aligns with the increase of EB at higher chitosan concentration in the blend polymer [32]. This phenomenon can be due to the improvement of chain mobility as the effect of the interaction between the plasticizer and polymer chains [38].

4.2. Thermal properties

A previous study reported that adding corn starch into chitosan resulted in similar thermal degradation behaviors. The study also revealed that starch reduced the char residue of films [25]. The chitosan and starch blend can also increase the decomposition temperature of the resulting films. Bajer *et al*., [39] reported that the maximum rate of thermal degradation temperature shifted from 293℃ for a starch film to 298℃ for a starch-chitosan film. They also added Aloe vera gel as a modifier to the starch-chitosan film, which resulted in a higher thermal degradation temperature of 302℃ for the blend polymer with 50% Aloe vera gel w/w starch. The higher degradation temperature is caused by chitosan or chitosan and Aloe vera gel's ability to create new intermolecular hydrogen bonding that promotes the improvement of the crystalline structure of the films. The thermal stability changes from starch, starch-chitosan, and starch-chitosan-Aloe vera gel can be seen in Table 2.

Table 2. Thermal stability changes between pure starch, starch-chitosan, and starch-chitosan-Aloe vera gel films [39]

Sample formulation	Temperature of maximum rate of degradation (℃)	
	Stage I (Loss of moisture, 0-150℃)	Stage II (Sample decomposition, 150-330℃)
Starch (S)	70	293
Starch-chitosan (SC)	78	298
SC-Aloe vera gel 10% w/w starch (SCA10)	94	300
SCA20	78	301
SCA30	95	301
SCA40	90	302
SCA50	91	303

4.3. Solubility and water vapor permeability

The main drawback of starch film is its sensitivity to water molecules. Meanwhile, chitosan is insoluble in water and organic solvents. Therefore, the blend of these polymers is expected to fabricate a film with moderate solubility in water and good water vapor permeability. Garcia *et al*., [40] soaked corn starch (CS), chitosan (CH), corn starch with chitosan (CS + CH), and corn starch with glycerol and chitosan (CS + G + CH) films in distilled water to measure their solubility. They found that chitosan films have the lowest solubility among all films. The solubility of the CH film increased by about 80% by adding corn starch, from 8.6% for the CH film to 15.5% for the CS + CH film. Furthermore, having a plasticizer in the polymer blend film improves the solubility value from 8.6% (CH) to 24.2% (CS + G + CH) due to the hydrophilic characteristic of glycerol.

Additionally, chitosan is a material that is highly permeable to water vapor [41]. Blending chitosan and starch can overcome that drawback and decrease the resulting film's water vapor permeability (WVP). As reported by previous studies, the starch in chitosan-starch film effectively improved the film's permeability to water vapor [21], [25], [28], [31]. Zheng *et al*., [25] reported the reduction of WVP of about 25% for chitosan film to chitosan incorporated with starch at 0.9 w/w chitosan. This reduction may be due to the CH and AS molecular interactions, which formed a compact structure that prevents the diffusion of water molecules. Table 3 shows the effect of starch amount variation on the water vapor permeability (WVP) of films.

Table 3. The effect of various starch amount in chitosan-starch blend on water vapor permeability of films [25]

Films formulations	WVP (10^{-10} g/Pa·m·h)
CH	2.648 ± 0.136
CH-0.3AS	2.341 ± 0.103
CH-0.6AS	2.201 ± 0.165
CH-0.9AS	1.998 ± 0.211
CH-1.2AS	2.132 ± 0.156
CH-1.5AS	2.191 ± 0.231

4.4. Antimicrobial properties

The main reason for the popularity of chitosan as a food preservative is its antimicrobial properties against a wide range of food-borne filamentous fungi, yeast, and bacteria [5]. Pinzon *et al*., [37] reported that the edible coating from banana starch-chitosan decreased the fungal decay of strawberries when compared to the control (without coating). The antifungal mechanism of chitosan may be due to the activation of chitinases leading to chitin hydrolysis that prevents fungal growth. Furthermore, they found an improvement in the antifungal activity of chitosan-based coatings after adding Aloe vera gel.

Moreover, chitosan's antibacterial and antifungal properties are also present in layered film. Chen *et al*., [30] found that gelatin-chitosan-corn starch three-layer films inhibited the growth of *E. coli* colonies. They also applied the film to cherry tomatoes, and it decreased the total bacteria count at 20 days of storage compared to non- and polyethylene-covered cherry tomatoes. This finding aligns with Zhao *et al*., [28], who found that a corn starch film did not exhibit an inhibition zone. Meanwhile, the chitosan film showed an obvious inhibition zone against *E. coli, S. aureus*, and *B. cinerea*. The antimicrobial properties were noticeably increased after the treatment with tannic acid on the chitosan layer.

5. The Potential of The Chitosan-Starch Blend on Fruit and Vegetable Preservation

5.1. Apples

Martin da Costa *et al*., [33] applied a purple yam starch-chitosan edible coating to apples and found remarkable results. The coating reduced the apple's weight loss from 3.34% for the yam starch-coated apples to 2.89% for the yam starch-0.5g chitosan. The evaluation was conducted over a 4-week storage period. The authors evaluated the internal appearance of the coated apples after 30 days of controlled storage by cutting them. The apples coated with a polymer blend had a better internal visual appearance when compared to those without and the yam starch-coated apples. This finding may be due to the low water permeability rate of the blend polymer coating. Therefore, the purple yam starch-chitosan coatings coating maintained the apples' quality.

5.2. Strawberries

The application of starch-chitosan-based coating also showed a good effect on strawberry preservation. Uncoated strawberries showed fast fungal growth, with the fungal decay being 10% after 2 days of storage and 100% (full spoilage) after 8 days of storage [37]. In contrast, the starch-chitosan-coated strawberries only showed fungal decay after 3 days of storage and reached 55% decay after 14 days. The starch-chitosan coating also prevented strawberries from weight loss because the hydrophobic nature of chitosan reduced the edible coating's water permeability.

5.3. Raspberries

Cejudo *et al*., [31] applied a chitosan starch film for a 13-day preservation experiment with fresh raspberries. Raspberries packed with chitosan-starch film showed less visual spoilage than unpacked berries. The raspberries packed without the polymer blend film spoiled after 3 days. They lost weight and firmness due to the absence of a water vapor barrier. Meanwhile, the raspberries packed with the polymer blend film required a longer time before spoiling. Furthermore, the polymer blend film showed better preservation compared to pure chitosan film. This finding may be caused by the higher abundance of hydrogen bonding in the chitosan-starch film.

5.4. Cherry Tomatoes

The effect of chitosan-based polymer films also affected the storage time of cherry tomatoes. Chen *et al*., [30] reported that the three-layered film packaging from gelatin-chitosan-starch increased the storage time of cherry tomatoes. Mold was found after 20 days of storage of unpacked cherry tomatoes. Meanwhile, the tomatoes packed with gelatin-chitosan-starch film showed no noticeable shrinkage or decay on the 20th day of storage. The same trend was also seen in the firmness of the cherry tomatoes.

5.5. Mangoes

Oliveira *et al*., [42] reported that the chitosan-cassava starch coating had a significant impact on mango weight loss during its storage for 14 days. The average approximate weight loss of chitosan-starch-coated mangoes were 4.9%, while the uncoated mangoes had about 6.03% weight loss. Moreover, the chitosan-starch coating also decreased the mangoes' respiration rate during 13 days of storage. The sample with 0.5% starch and 0.25% chitosan showed a better impact on the mangoes' shelf life without significant changes in its sensorial quality attributes, such as the color of the peel, texture, and aroma of the fruit. The application of chitosan-starch coating also increased the fruit's microbial resistance.

5.6. Papayas

Escamilla-García *et al*., [43] used a mixture of chitosan and oxidized starch to coat papayas. The effect of the coating was evaluated for 15 days of storage at room temperature. They found that coated papayas had a higher firmness loss (about 92.02%) than uncoated papayas (47.36%) after 10 days of storage. The trend in firmness reduction aligns with the weight loss results where the uncoated papayas' weight loss was up to 33.3% higher than the coated fruit after 15 days of storage. In addition, the chitosan-starch coating also inhibited microbial activities.

5.7. Eggplants

The chitosan-starch blend polymer film also showed good results in eggplant preservation. Although the weight of both coated and uncoated eggplants decreased daily during storage, the weight loss of the coated eggplants was lower than the uncoated ones. Weight loss was reduced by about 48% at 12 days of storage [44]. The chitosan-starch coating also protected the eggplants from nutrient loss. The coating can slow the fruit ripening process, which correlates with the respiratory activity of the fruit. The film regulated the carbon dioxide and oxygen entry and outflow, decreasing carbohydrate content breakage.

6. Conclusion

The use of starch along with chitosan as packaging films and coatings has an excellent effect on the preservation of fruits and vegetables. According to reports by several researchers, chitosan-starch-based packaging and coatings successfully decreased the weight loss of fresh produce. These polymer blend films and coatings also maintained the produces' firmness and other sensorial properties. Moreover, adding starch to chitosan can decrease water vapor permeability and increase solubility. The antimicrobial nature of chitosan also helps the polymer blend to lengthen the produces' storage time before decay. Thus, chitosan-starch-based films and coatings provide a promising approach to increasing the shelf life of fruits and vegetables.

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