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Physical Characteristics of Biocomposite Edible Films Based on Fish Gelatin and Nanochitosan with the Addition of Beeswax: A Review

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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Review Article

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ABSTRACT

Edible films is a thin layer that can be eaten directly with the food it is coated with and is biodegradable, so it is used as an alternative to plastic packaging. The components of the edible film consist of hydrocolloids (proteins and polysaccharides), lipids (fatty acids), and composites (a combination of hydrocolloids and lipids). The physical characteristics of edible films that are commonly used are thickness, tensile strength, elongation percentage, water vapor transmission rate and solubility. Several studies have shown that the addition of beeswax to gelatin and nanochitosan edible films can produce films with good physical characteristics. The addition of a greater concentration of beeswax can increase the thickness value and decrease the value of the water vapor transmission rate and water solubility of the edible film.

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1. INTRODUCTION

Edible film is a thin layer that functions as packaging for food products that can be eaten directly with the food ingredients it is coated with. Edible film is an environmentally friendly packaging because it easily decomposes naturally or is biodegradable, so it is used as an alternative to plastic packaging [1]. Edible films are generally made from natural materials such as polysaccharides, proteins, fats or a combination of several materials (composites), with or without the addition of other materials such as plasticizers and surfactants [2]. Edible films made from only one material still have some drawbacks, so to fix them by combining hydrocolloid bolimers with lipids. Composite edible films are film which is formed from a combination of hydrocolloid biopolymers with lipids, which can increase the advantages and cover the disadvantages of each of these films [3]. The constituents of biocomposite edible films include hydrocolloids (gelatin and chitosan), and lipids (beeswax).

Gelatin is a protein derivative derived from animal bones and skin through an extraction process [4]. Fish gelatin has a high content of glycine, proline, and hydroxyproline so it is more flexible and easy to apply in food ingredients [5]. Mohammadi et al. [6], states that films made of gelatin have good optical properties, but have deficiencies in mechanical properties and water barriers, so that they can be detrimental film gelatin is used as food packaging. Gelatin films have weaknesses in physical properties such as water vapor barrier and thermal [7]. One way to improve the physical properties of fish gelatin films is by adding other biopolymers that come from renewable resources and have film-forming properties such as chitosan [8].

Chitosan is a chitin derivative which is the most abundant polysaccharide on earth after cellulose, has hydrophobic properties and can form film layers. Modification of chitosan in nano form aims to streamline the absorption of chitosan by expanding the surface of the chitosan [9]. Nanochitosan can be used as a nanofiller in making edible films, because it can improve the mechanical properties, film color and water vapor permeability of the resulting edible films compared to ordinary edible films [10]. Haghighi et al. [11] explained that edible films based on gelatin and chitosan have a good barrier against gases such as CO_2 and O_2, but still have weaknesses in mechanical properties and water vapor barrier properties. The poor water vapor barrier properties of edible films can be improved by adding lipids because they are hydrophobic, for example, like beeswax [12].

Candlebee is a type of lipid that can be used in making edible films because it has hydrophobic properties that function to inhibit water vapor that diffuses through film. The use of the right concentration of beeswax will be able to improve the physical and mechanical capabilities of edible films in protecting packaged food products [13]. Testing the physical and mechanical characteristics of the edible film can be used as a reference in adjusting it to the product to be packaged [14]. Based on this information, it can be seen that biocomposite edible films made from nanochitosan, gelatin, and beeswax can improve physical properties, especially the rate of water vapor transmission so that they have the effect of extending the shelf life and improving the quality of the products they are packaged.

2. COMPONENTS OF EDIBLE FILM

The components of the edible film consist of hydrocolloids (proteins and polysaccharides), lipids (fatty acids), and composites (a combination of hydrocolloids and lipids) [13]. Rohim et al. [15] states that hydrocolloid polymers consist of proteins (gelatin, casein, soy protein, corn protein and wheat gluten) and carbohydrates (starch, alginate, pectin, arabic gum and other carbohydrate modifications). Lipid groups commonly used in the manufacture of edible films includes waxes, beeswax, glycerol and fatty acids. Making edible films can also be made with or without the addition of plasticizers such as glycerol, sorbitol, sucrose, and others [2]. The addition of plasticizers to the manufacture of edible films is necessary to increase the elasticity and flexibility of edible films [1]. The following components make up edible film, which can be seen in Table 1.

2.1 Fish Gelatin

Gelatin is a protein derivative derived from animal bones and skin through an extraction process. Generally, gelatin is produced from materials that are high in collagen content such as those found in animal bones and skin [4].The gelatin extraction process can be carried out using an acid or alkaline process, but the gelatin extraction process is safer, faster and preferred by the industry, namely using the citric acid process. [17]. The characteristics of gelatin are bright yellow or transparent to near white, in the form of sheets, powder or flour-like, stems, like leaves, soluble in hot water, glycerol and citric acid and other organic solvents. Gelatin can expand and absorb water 5-10 times its original weight [18]. Gelatin can be used in the food and non-food industries due to the unique properties of gelatin. The characteristic properties of gelatin are that it can change reversibly from sol to gel form, expands in cold water, can form films, affects the viscosity of a material and can protect the colloidal system [19]. Gelatin in non-food products is used in the pharmaceutical and medical industries, the cosmetic industry and the photography industry. The food industry utilizes gelatin to be used as an additional ingredient, namely as an emulsifier, stabilizer, foaming agent, encapsulant and film-forming agent [20]. The following fish gelatin can be seen in Fig. 1.

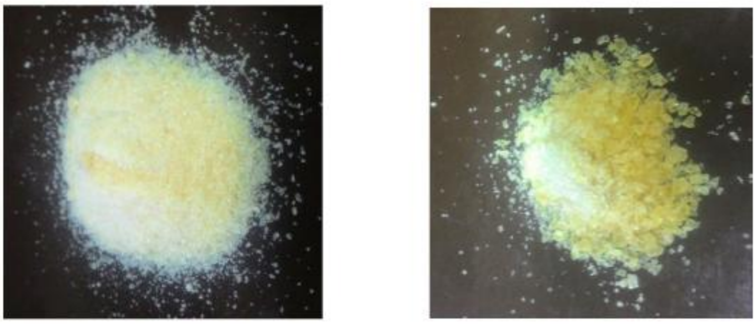
2.2 Nanochitosan

Chitosan is a natural polysaccharide resulting from the deacetylation process (removal of the -$COCH_3$ group) of chitin. Chitin is the main constituent of the exoskeleton of crustacean aquatic animals such as crabs and shrimp. Chitin is composed of several units of N-acetyl-D-glucosamine (2acetamido-2-deoxy-Dglucopyranose) which are connected linearly via ß(1→4) bonds [15]. Chitosan which has changed its size to nano aims for wider utilization with a particle size range of

Table 1. Edible film composition components

Component	Material Type	Example
The main constituent of edible films	Animal protein	Whey protein, collagen, gelatin, casein, egg white protein, fish myofibrillar protein, feather keratin.
	Plant proteins	Soy protein, corn zein, wheat gluten, pea protein, rice bran protein, cottonseed protein, peanut protein.
	Linear and neutral polysaccharides	Agar, curdlan, cereal b-glucan, methylcellulose, hydroxypropyl methylcellulose, microcrystalline cellulose, pullulan, konjac glucomannan, inulin.
	Polysaccharide liner anions	Sodium alginate, propylene glycol alginate, carrageenan, pectin, gellan gum, carboxymethylcellulose or cellulose gum.
	Cationic linear polysaccharides	Chitosan
	Fat	Waxes (beeswax, paraffin, carnauba wax, candelilla wax, rice bran wax), acetoglycerides.
	Resins	Shellac, terpene, asafoetida, benjoin, chicle, guarana, myrrh, opoponax, sandaraque, styrax
Plasticizers	Polyol	Glycerol, propylene glycol, polypropylene glycol, sorbitol, polyethylene glycol, corn syrup
	Other	Sucrose and water
Additives	Flavor	Oil based flavors, citrus, mint, volatile oils
	Antimicrobial	Organic acids (acetic, benzoic, lactic, propionic, sorbic), fatty acid esters (glyceryl monolauric); Polypeptides (lysozyme, peroxidase, lactoferrin); nitrites and sulfites, chitosan, bacteriocins (nisin, pediosin), parabens, liquid smoke, sodium chloride.
	Antioxidant	Ascorbic acid, 4-hexylresorcinol, amino acids (cysteine and glutathione), citric acid.
	Nutrition	Vitamin E, calcium, zinc, aluminium
	Emulsifier	Lecithin, mono and diglycerides, mono and diglyceride esters, Fatty sucrose esters, fatty alcohols, fatty acids
	Lipid emulsion	Edible wax, fatty acids
	probiotic organisms	Bifidobacterium (Bifidobacterium lactis Bb-12)
	Plant essential oil	Cinnamon, oregano, lemongrass, savory, sweet inula, vanillin, cloves, lemongrass

Source: Erkmen and Barazi [16]



(a) (b)

Fig. 1. Extraction of catfish skin gelatin with (a) Acid Process; (b) Base Process

Source:Nasution et al. [21]

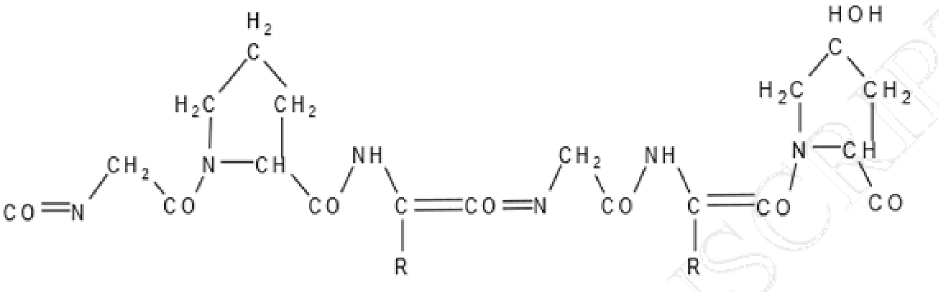


Fig. 2. Chemical structure of gelatin

Source: Hassan et al. [22]

10-1000 nm [23].Chitosan modified into a nano form is used as an additional filler in edible formulas so that it will produce nanocomposites to increase antibacterial activity. This is due to the large surface area and positive charge of the amine groups of chitosan nanoparticles [24].

Nanochitosan can be formed using various methods, but in the manufacture of stable and high quality nanochitosan usually requires a method that is quite difficult. Determining the use of the method used in the manufacture of nanochitosan depends on factors including the desired particle size, chemical and thermal stability of the particles, the kinetic profile of the particles and the toxicity of the residue associated with the final product [24]. Suptijah et al. [23], stated that the method of making nanochitosan is simpler and the process is more efficient, namely using the ionic gelation method.

The ionic gelation method is a method for making nanochitosan based on ionic interactions between amino groups in chitosan and negatively charged polyanions [25]. According to Nadia et al. [26]. The ionic gelation method is a polyelectrolyte combination between positively charged chitosan and negatively charged tripolyphosphate. The addition of tripolyphosphate (TPP) and surfactant (Tween 80) can strengthen the mechanical properties of chitosan which is easily brittle and can form ionic cross-links between chitosan molecules, while reducing the particle size with a magnetic stirrer, can distribute a more homogeneous particle size. The size of nanochitosan particles using the ionic gelation method can produce very small particles that do not polymerize and do not cause enlarged (micro) particles, which have sizes ranging from 400-450 nm by reducing the particle size using a magnetic stirrer [23].

2.3 Beeswax

Beeswax is a product of the leftover feeling of honey which is processed first from beehives to produce wax. Remnants of beehives that have been treated usually still contain as much as 30% pure beeswax [27]. The advantage of beeswax as a raw material for edible film is that it is classified as food grade, its availability throughout the year, its use is still very limited, and the price is still relatively cheap and easy to obtain [28]. The content of beeswax is composed of free fatty acids, free fatty alcohols, and mixtures [29]. Beeswax is a hydrophobic agent consisting of a mixture of esters, hydrocarbons, fatty acids, alcohols, and others [30]. The following beeswax content can be seen in Table 2.

Table 2. Beeswax content

Content	Amount %
Monoester	35
Dieter	14
Triester	3
Hydroxy monoester	4
Hydroxy polyester	8
Esters	1
Polyester acid	2
Hydrocarbons	14
Free acid	12
Alcohol	1
Etc	6
Total	100

Source: Bogdanov [27]

Fig. 3. Beeswax
Source: Bogdanov [31]

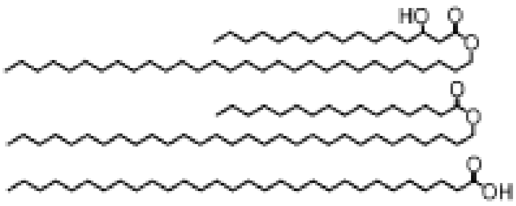


Fig. 4. Chemical structure of beeswax
Source: Floros et al. [29]

3. PHYSICAL CHARACTERISTICS OF EDIBLE FILM

Characteristics of edible film is a key success factor in making edible film as good packaging Dwimayasanti [32]. Good packaging is packaging that can control the permeability of water vapor and gas, is easy to decompose, and is able to maintain the quality or quality of food ingredients during the storage period because they are antimicrobial. Aguirre et al. [33]. Characteristics of edible films that are commonly used are physical and mechanical characteristics. Analysis of the physical and mechanical characteristics of edible films includes: thickness, tensile strength, elongation percentage, solubility, water vapor transmission rate and water content Diova et al. [34]. The physical characteristics of the edible film can determine the flexibility of the packaging, the smaller the tensile strength value and the greater the elongation value, the more applicable the edible film will be. Mechanical characteristics can determine the quality of the packaging, the smaller the water vapor transmission rate is, the better the quality of the edible film will be. [35]. Nurinda et al. [36] reported that edible films which is used as food packaging must meet the quality standards set by the Japanese Industrial Standard (1975). The following are the standard characteristics of edible films, which can be seen inTable 3.

Table 3. edible film Characteristic Standards

Characteristic Parameters	JIS standard
Thickness	Max. 0.25mm
Water Vapor Transmission Rate	Max. 7 g/m^2/ 24 hours
Tensile strength	Min. 0.3 Mpa
Elongation	Min.70%.

Source: JIS (1975) in Nurindra et al. [36]

3.1 Thickness

Film thickness is an important parameter in determining the feasibility of edible films as packaging for food products because thickness can affect the physical and mechanical properties of other edible films, such as tensile strength, elongation, solubility and water vapor permeability [1]. Factors that can affect the thickness of the edible film include the area of the printed plate and the volume of the suspension, the constituent components, and the drying and addition of glycerol [37]. The smaller the thickness of the edible film, the faster water will penetrate the edible film, but the higher the thickness, the better it will be to hold water vapor [37].

3.2 Water Vapor Transmission Rate

The water vapor transmission rate is a test carried out to find out how resistant edible film is in holding water [38]. The value of the water vapor transmission rate can determine the quality of the edible film as packaging, because the lower the value of the water vapor transmission rate, the better the quality of the edible film because it can protect the product, slow down the oxidation reaction, and can maintain the water content in the product [37]. The value of the water vapor transmission rate is influenced by several factors including the structure of the edible film constituents and the concentration of the plasticizer used as reported [39].

3.3 Solubility

Solubility is a test parameter for edible films to determine how soluble the film is when consumed and also as a determining characteristic of biodegradable films when used as food packaging [40]. Togas et al. [39] Stateds thatThe solubility percentage of an edible film can be used as an indicator to measure water resistance, film integrity and biodegradability of the edible film when used as a packaging material. The solubility of edible films is influenced by several factors, including the source of the basic ingredients for making edible films and the added plasticizers [39].

4. EDIBLE FILM BIOCOMPOSITE WITH THE ADDITION OF BEESWAX

Edible films biocomposite is an edible film composed of a combination of hydrocolloid biopolymers and lipids. The combination of these biopolymers can complement and cover the deficiencies of each of these biopolymers, thereby increasing the characteristics of the resulting edible composite film [41]. Edible films with the addition of lipids such as beeswax can improve physical properties in resisting the transmission rate of water vapor and can affect the thickness, tensile strength and elongation percentage of edible films. The hydrophobic nature of beeswax serves to increase the moisture barrier properties of the edible film, so that the edible film produced is better [42].

Edible films Lipid-based (waxes and resins) have a high efficiency of preventing moisture loss and adsorption due to their low permeability and hydrophobicity, but their appearance is generally opaque and unattractive as packaging materials [43]. The lower the value of the water vapor transmission rate, the better the quality of the edible film because it can protect the product, slow down the oxidation reaction, and can maintain the water content in the product [37]. The small value of the water vapor transmission rate indicates that the film is better when used as a packaging material because less water vapor can penetrate into the film and the moisture content of the packaged material does not increase [44].

Table 4. Biocomposite edible film with the addition of beeswax

Ingredients	The Best Beeswax Concentration	Results	Reference
Carrageenan and Beeswax	0.8%	thickness increases 0.1534 mm water vapor transmission rate decreases 25.3411 g/m^2/hour solubility 88%	Togas et al. [45]
Carrageenan and Beeswax	0.3%	thickness increases 0.0705 mm water vapor transmission rate decreases 2.1797 10g/30Min solubility 39.1547%	Diova et al. [34]
Suweg Starch and Beeswax	1%	thickness increases 0.193 mm water vapor transmission rate decreases 3.667 g/m^2/hour solubility 56.142%	Safitri et al. [14]
sago starch, Gelatin and Beeswax	1%	thickness increases 0.129 mm water vapor transmission rate decreases 0.027 g/m^2/hour	Mudaffar [13]
Cassava starch, Prolis and Beeswax	0.9%	thickness increases102.23 μm water vapor permeability decreases 7.60 × 10$^{−12}$ gm/ Pa/s solubility 21.62%	Vergara et al. [47]

The addition of beeswax as a plasticizer to chitosan-based edible films can affect the physical characteristics of chitosan edible films, such as decreased tensile strength but on the other hand can increase the thickness and elongation percent values. Chitosan-based edible films without the addition of beeswax plasticizer tend to be thin and brittle, while edible films with the addition of beeswax plasticizer are thicker in proportion to the increase in beeswax concentration [45]. Nandiwilastio et al. [12], stated that edible films chitosan-based with the addition of zinc oxide and beeswax nanoparticles can significantly affect the physical and mechanical properties of the film. Significant changes were found in the water vapor transmission rate parameter which decreased to 10.38 g/m2/hour and the film thickness increased by 90.48 µm, and has a fairly stable elongation value of more thanri 40%.

According to Togas et al. [45] that the addition of beeswax in addition to increasing thickness and decreasing the transmission rate of water vapor can reduce the solubility value of carrageenan-based composite edible films because beeswax has hydrophobic properties. Beeswax tends to be more difficult to homogenize in water, due to its hydrophobic nature [44]. According to Harumarani et al. [46], states thatThe lower the solubility of edible film, the better it is used as a packaging material and is biodegradable. Edible films that have high solubility are very good for use in ready-to-eat food products because they dissolve easily when consumed [1]. Following are some studies regarding the addition of beeswax to biocomposite edible films, which can be seen in Table 4.

5. CONCLUSION

Edible film is a thin layer and is environmentally friendly because it easily decomposes naturally or is biodegradable, so it is used as an alternative to plastic packaging. Edible films are generally made from natural ingredients such as hydrocolloids (polysaccharides, proteins) and lipids (fats) or a combination of hydrocolloid polymers and lipids (composites), so that they can be eaten directly with the food they are coated with. Edible films based on gelatin and chitosan have a good barrier against gases such as CO_2 and O_2, but still have weaknesses in mechanical properties and water vapor barrier properties. Meanwhile, edible films with the addition of lipids such as beeswax can improve their physical properties in resisting the transmission rate of water vapor and can affect the thickness, tensile strength and elongation percentage of edible films. The use of the right concentration of beeswax will be able to improve the physical mechanical capabilities of edible films such as thickness, water vapor transmission rate and tensile strength. Edible film with a lower water vapor transmission rate and lower solubility value can be used as an edible film for food packaging which is able to protect the product inside so that it can increase the shelf life of the product it is coated with.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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