Potential Biodegradable Sticker Based On Carrageenaan and Chitosan With Addition of Lauric Acid : A Mini Review

ABSTRACT

Plastic pollution is a significant problem with detrimental effects on the environment and human health. Conventional plastic stickers, widely used in various sectors, contribute to plastic waste accumulation and slow degradation. To address this issue, review has focused on developing biodegradable stickers made from natural polymers such as carrageenan and chitosan. Carrageenan, derived from red seaweed, forms gels and has potential as a biodegradable film due to its stability and high fiber content. Chitosan, derived from crustacean waste, is non-toxic and can act as a stabilizer, thickener, and protective layer in food products. However, chitosan alone has limitations in terms of brittleness and rigidity. Lauric acid, a medium-chain fatty acid found in coconut oil, can be added as a film-forming material to improve the mechanical properties and reduce water vapor permeability. Biodegradable films thickness, tensile strength & Percent elongation, and surface morphology play important roles in their quality characteristics. Biodegradable stickers made from lauric acid-based biodegradable films offer ink resistance, biodegradability, water resistance, and water solubility as key characteristics.

Keywords: Biodegradable stiker, Hydrocolloid, Lauric Acid

1. INTRODUCTION

The issue of plastic stickers is related to the impact of plastic materials used as labels on various products. Plastic stickers often cannot be recycled or composted, making them a source of pollution that harms the environment and human health [41].

Studies have been conducted to develop stickers capable of undergoing organic biodegradation, transforming into more environmentally friendly materials as a solution to address this issue. The concept of stickers naturally decomposing in a shorter timeframe and causing fewer negative impacts on the environment is quite intriguing [42]. The material used in biodegradable stickers that utilize hydrocolloid materials consists of proteins and polysaccharides. Both of these materials are capable of forming stickers with strong film-forming properties. [43]. Biodegradable stickers can offer satisfactory physical and mechanical characteristics, including effective adhesion, flexibility, and resilience against water and temperature variations. Alongside their biodegradability advantage, these qualities make them suitable for a range of applications, including food packaging, cosmetics, and commercial advertising [44].

Chitosan, as a polysaccharide, is a derivative of deacetylated chitin and is classified as a hydrocolloid compound found in crustaceans, such as crabs [1]. The advantages of chitosan include being non-toxic [2], not altering color and aroma [3], and being able to form films that are strong and not easily torn [4]. Chitosan has the potential to be developed as a material for edible film production because it can be used as a stabilizer, thickener, emulsifier, and a clear protective layer on food products. Chitosan is non-toxic, biodegradable, biocompatible, and a good film former [5]. However, the use of chitosan alone as a single raw material in the production of edible films still has some weaknesses, such as brittleness and rigidity [6].

Carrageenan is a linear sulfated polysaccharide compound composed of D-galactose and 3,6-anhydro-D-galactose, obtained from the extraction of Eucheuma cottonii, a type of red seaweed (Rhodophyceae) [7]. Carrageenan is a good alternative as a basic material for manufacturing edible film packaging to improve the shelf life and quality of packaged food [7]. Carrageenan is a water-soluble polymer and has high potential as a film former [8]. Carrageenan has the potential to be a biodegradable film due to its ability to form gels, its stability, edibility, renewability, and high fiber content [9]. By adding hydrophobic materials, the shortcomings of biodegradable stickers can be overcome. Lipids are one of the additive materials added to films containing polysaccharides to improve the hydrophobic properties of the resulting film [10].

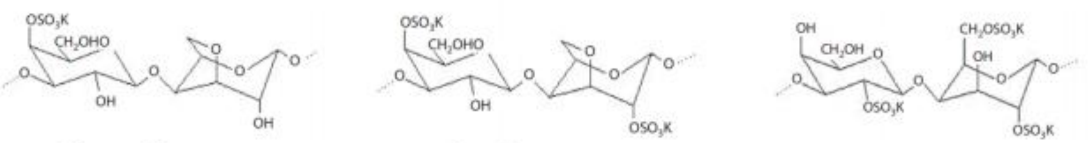
One of the fatty acids that can be used as a film-forming material is lauric acid. Lauric acid is a medium-chain saturated fatty acid composed of 12 carbon atoms and has hydrophobic properties. Lauric acid has many uses, including its role as an antiviral, antiprotozoal, and antibacterial agent in the human body. According to Santoso et al. [11], lauric acid functions as a biodegradable composite film former, belonging to the lipid group, which prevents water vapor transmission without reducing the film's elasticity. The addition of lauric acid can reduce water absorption and water vapor permeability [12].

2. BIODEGRADABLE FILM

According to Han [13], the materials used in the production of edible films are hydrocolloids, lipids, and composites. Hydrocolloids include polysaccharides and proteins. Proteins can be derived from soybeans, gelatin, milk protein, and fish protein. Polysaccharides can be obtained from cellulose and its derivatives, seaweed extracts such as alginates, agar, carrageenan, gums, chitosan, and others. Lipids can be derived from fatty acids, and composites are a combination of hydrocolloids and lipids. Plasticizers, antioxidants, vitamins, essential oils, and pigments, which are additives, are used to enhance the protective properties of biodegradable films.

2.1. CARRAGEENAN

Carrageenan belongs to the class of galactan polysaccharides found in red seaweeds of the Rhodophyta class and contains intercellular matrix components. In seaweeds, carrageenan functions as a hydrophilic structure to accommodate water pressure and waves. Besides being biodegradable, carrageenan is also used as a viscosity regulator, stabilizer, and thickening agent [16]. According to Necas and Bartosikova [17], carrageenan is divided into six classes: iota, lambda, kappa, theta, nu, and mu-carrageenan. Iota, kappa, and lambda are classes that exhibit thermoreversible properties, meaning they melt upon heating and re-gel upon cooling, whereas lambda carrageenan does not form a gel.



Kappa Karagenan　Iota Karagenan　Lambda Karagenan

Fig 1. Structure Karagenan
Source: Thakur and Takur [16]

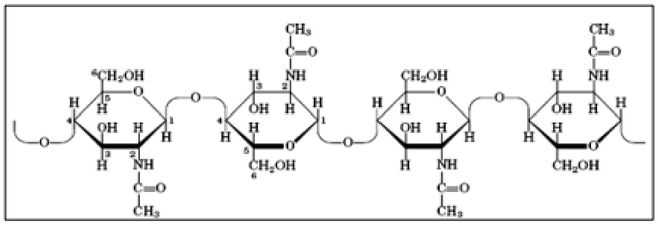
Carrageenan has the ability to form gels, making it suitable for use in edible films. Therefore, it is chosen based on its highest gel strength. According to Asikin and Kusumaningrum [18], carrageenan from seaweeds harvested at 40 days has the highest gel strength. However, the hydrophilic nature of carrageenan is a drawback as a material for forming biodegradable films with low water vapor transmission resistance.

The use of carrageenan as a film-forming material offers advantages such as rapid biodegradability, edibility, and the ability to inhibit the ingress and egress of oxygen and water vapor. However, the use of carrageenan as a film-forming material also comes with disadvantages, such as being prone to tearing or breaking during use, being hydrophilic and thus susceptible to water absorption, and having a higher production cost compared to conventional plastics [8].

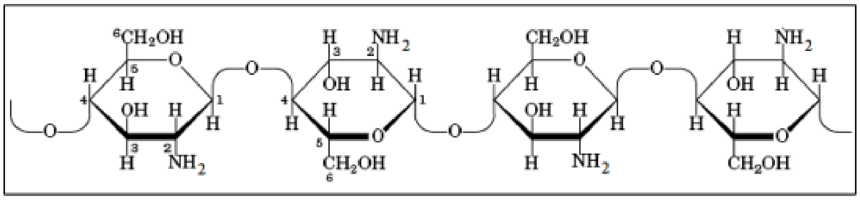
2.2. CHITOSAN

K Chitosan is a polysaccharide obtained from the deacetylation of chitin, and it is derived from the waste of crustacean animals such as crabs, shrimps, and crayfish. Chitosan carries a positive charge and is soluble at pH below due to the protonation of its amino groups. It can be designed into various forms such as films, gels, and nanoparticles. The ability of chitosan to create good films has made it one of the most extensively researched polysaccharides for edible film production [19].

According to Nurhayati and Agusman [20], the process of chitosan production involves deproteinization (removal of proteins), demineralization (removal of minerals), depigmentation (removal of colorants), and deacetylation, which eliminates the acetyl groups (-COCH¬3) using a basic solution to obtain chitosan biopolymer.



(a)



(b)

Fig 2. Structure Kitin (a) and chiitosan (b)
Source: Dompeipen [21].

Quality standard of chitosan depends on various parameters, and the quality standards for chitosan are presented in Table 1

Table 1. Standard Chitosan

Parameter	Standard	
	Dalwoo Korea	Japanese Protan Lab
Apparition	White or yellow powder	Clear solution
Particle size	25-200 mesh	Flake to powder
Water content	≤ 10 %	≤ 10 %
Ash content	≤ 0,5 %	≤ 2 %
Protein content	≤ 0,3 %	-
Distillation degree	≥ 70 %	≥ 70 %
Viscosity	50-500 cps	200-2000 cps
Insolubility	< 1%	-
Heavy metal content : As, Pb	< 100 ppm	-
pH	7-9	7-8
smell	No smell	Not smell

Source : Rochima [22]

Chitosan has the potential to be developed as a material for edible film production as it can act as a stabilizer, thickener, emulsifier, and form a transparent protective layer in food products. Chitosan is non-toxic, biodegradable, biocompatible, and an excellent film-forming agent [6].

The use of chitosan films offers advantages such as having excellent barrier properties against oxygen, water vapor, and aroma components, as well as rapid biodegradability through natural microorganisms. However, there are drawbacks to using chitosan films, including their high production cost, less favorable mechanical properties compared to conventional plastics, and their tendency to readily absorb water due to their hydrophilic nature [45].

2.3. LAURIC ACID

Lauric acid is a medium-chain fatty acid (MCFA) with 12 carbon atoms that is found in pure coconut oil or Virgin Coconut Oil (VCO). Lauric acid has various uses, such as serving as an antivirus, antiprotozoal, and antibacterial agent in the human body. In the cosmetic industry, it is used as a moisturizer and softener, and it is also utilized in shampoo and soap production [23]. Coconut oil contains approximately 51-53% lauric acid, which can be separated from the oil through hydrolysis with the assistance of water under high temperature and pressure. After hydrolysis, coconut oil is transformed into free fatty acids (including lauric acid) and glycerol [24].

Lauric acid has a melting point of 44°C and a boiling point of 225°C, so it appears as a white solid at room temperature and easily melts when heated. Its chemical formula is CH3(CH2)10COOH, and its molecular weight is 200.3 g/mol. Lauric acid is soluble in water, but the solubility decreases as the length of the fatty acid chain increases. This solubility property is utilized to separate various unsaturated fatty acids through crystallization processes [26].

According to Santoso et al. [25], lauric acid functions as a biodegradable composite film former in the lipid category, providing resistance to water vapor transmission without compromising the film's elasticity. In line with the statement by Saleh and Muhammad [33], the addition of lauric acid to biodegradable films can reduce their hydrophilic properties and excessive water vapor content.

The advantages of using lauric acid films include their ability to protect products from oxidation, unstable temperatures, moisture, and ultraviolet radiation. Lauric acid films also possess high mechanical properties. However, the disadvantages of using lauric acid films include limitations in their applications and their high production costs [46].

Table 2. Biodegradable film with addition of lauric acid

Title	Reference	Result
Effect Of Lauric Acid On The Thermal And Mechanical Properties Of Polyhydroxybutyrate (PHB)/Starch Composite Biofilms	Adorn et al [14]	The addition of lauric acid to starch-based biodegradable films enhances the mechanical properties and improves thermal stability
Effects Of Two Fatty Acids On Soy Protein Isolate/Sodium Alginate Edible Films: Structures And Properties	Chen et al [15]	The addition of lauric acid improves the mechanical properties of the film better than other fatty acids. The addition of fatty acids enhances the water barrier properties of the film.
The effect of fatty acids on the physicochemical properties of edible films composed of gelatin and gluten proteins	Fakhouri et al [12]	addition of fatty acids affects the mechanical properties, solubility, and water resistance of the film

2.4. PLASTICIZER

Plasticizer is a substance that is commonly added in the production of biodegradable films. It serves to reduce brittleness and increase flexibility and durability of the film, especially when stored at low temperatures. Glycerol is one of the commonly used plasticizers in the production of edible films and it has an impact on raw materials such as starch. Glycerol is more efficient in improving the physical and chemical properties related to tensile strength, elongation, and transparency [26].

Glycerol is suitable as a plasticizer because it is in liquid form. Its liquid form is advantageous as it is easily soluble in water and can be mixed into film solutions [27]. Glycerol is an important compound consisting of alkali trihydrate (propane-1,2,3-triol) with the chemical formula CH2OHCHOHCH2OH. This compound is widely found in animal fats.

3. CHARACTERISTIC OF BIODEGRADABLE FILM

3.1 THICKNESS

Thickness is a physical property that is influenced by the concentration of the solid solvent in the film solution and the size of the printing plate. The thickness of the film will affect the rate of water vapor and gas transmission [28]. Thickness is considered a physical property of the film that determines the quality characteristics of tensile strength, percentage elongation, and water vapor transmission rate. The thickness of biodegradable films [29]. The characteristic of thickness is the ability of biodegradable film as a packaging material. Thicker biodegradable films will result in a non-transparent appearance [30]. The purpose of measuring thickness is to understand the influence of the thickness of biodegradable films on the rate of vapor transmission and resistance to water and gas that enter the film matrix. The higher the thickness of the biodegradable film, the better its ability to inhibit vapor transmission, water resistance, and gas resistance.

3.2 TENSILE STRENGHT AND PERCENT ELONGATION

Tensile strength test is a measurement of the maximum force that a material can withstand when stretched or pulled before it breaks or tears. The higher the force applied, the greater the tensile strength. Edible films with high tensile strength will effectively protect packaged products from mechanical disturbances [40].This measurement aims to determine the force required to achieve maximum elongation in each area of the biodegradable film. Tensile strength depends on the concentration and type of materials used in the biodegradable film [31].

Percentage elongation refers to the maximum length change that occurs during stretching until the film breaks [32]. The value of percentage elongation reflects the stretchability, and generally, the addition of a plasticizer increases the flexibility of the film [33]. Decreased elasticity leads to increased film flexibility. The value of percentage elongation is related to the film's elasticity, where a higher percentage value indicates greater elasticity and better quality biodegradable film, whereas a low elongation value indicates lower film quality [34].

3.3 SURFACE MORPHOLOGY

SEM analysis is conducted to observe the compatibility of additive mixtures and to examine the surface morphology of the film. SEM analysis helps determine the morphology and structural changes of a material, such as fractures, folds, and pore formation in the film [35]. The microstructure of the film is an important element in understanding its properties. The microstructures obtained through SEM can vary from compact to porous, and from smooth to rough [36].

4. BIODEGRADABLE STICKER

The scientific term "biodegradable" refers to materials that can be degraded naturally, broken down by bacteria and fungi. In other words, you can throw items labeled biodegradable into the ground, and nature will be able to break them down and reuse their components. Therefore, the innovation of "biodegradable" stickers emerged, this term usually refers to stickers that can degrade fairly quickly and naturally, and are usually used in conjunction with packaging that is also labeled biodegradable [40].

5. CHARACTERISTICS OF POTENTIAL BIODEGRADABLE STICKERS MADE FROM LAURIC ACID-BASED BIODEGRADABLE FILM

5.1. INK RESISTANCE

Ink resistance testing is conducted to determine how well the biodegradable sticker can withstand ink and remain visible without fading or smudging during a specific storage period. The type of ink used on the biodegradable sticker can affect ink resistance. Typically, there are inks specially designed for application on biodegradable materials to minimize environmental impact and provide reasonable resistance to color fading [37].

5.2. BIODEGRADABILITY

Biodegradability describes the film's ability to degrade well in soil. Testing is performed using a soil test method by placing the biodegradable sticker on soil until the sticker undergoes complete degradation. Biodegradable sticker observations are conducted once a week. The biodegradability of the packaging due to microorganisms is influenced by additives, hydrophobic properties, polymer structure, and packaging material molecules [38].

5.3. WATER RESISTANCE

Water resistance in biodegradable stickers refers to the sticker's ability to withstand water without dissolving or undergoing significant structural changes. Good water resistance in the film is important to prevent the sticker from dissolving when exposed to moisture. The optimal water resistance in biodegradable stickers depends on factors such as the base material used, manufacturing method, and surface treatment. According to Nairfana [39], testing for water resistance is important because a good film is one that can protect the product from water, ensuring that water is absorbed by the film.

5.4. WATER SOLUBILITY

Water solubility is a parameter used to determine the ability of the biodegradable sticker to dissolve in water or resist water penetration. The percentage of solubility of the biodegradable sticker can be used as an indicator to measure water resistance, sticker integrity, and the biodegradability of the sticker when applied to a product. According to Rusli et al. [8], high solubility causes the film to dissolve easily in water and reduces its ability to withstand water. Films with high solubility are suitable for use in consumable products or packaging. High solubility is also related to the biodegradability of the sticker.

6. CONCLUSION

Biodegradable stickers offer good adhesive properties, flexibility, and resistance to water and temperature. They can be used in various applications such as food packaging, cosmetics, and commercial advertising. However, the use of chitosan alone may result in brittleness, while carrageenan may have lower water vapor transmission resistance. Lauric acid, as a fatty acid, can be added to enhance the hydrophobic properties of biodegradable films. The characteristics of biodegradable films include thickness, tensile strength, percentage elongation, surface morphology and characteristic of biodegradable sticker include ink resistance, biodegradability, water resistance, and water solubility. Overall, biodegradable stickers made from lauric acid-based biodegradable films offer a promising solution to the problem of plastic pollution.

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