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Fabrication and characterization of antioxidant biodegradable plastic from mangrove starch, chitosan, and clove essential oil

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Abstract

The development of biodegradable plastics offers a sustainable alternative to conventional ones, reducing environmental impact. This research is the fabrication and characterization of biodegradable plastic films derived from large-leafed mangrove (Bruguiera gymnorrhiza) starch, chitosan, and clove essential oil (CEO) as an antioxidant additive. Glycerol was used as a plasticizer, while carboxymethyl cellulose (CMC) and carrageenan were incorporated as thickeners. The effects of glycerol (2.7–9 ml), chitosan (4– 6% m/m), and CEO (0–8% m/m) concentrations on mechanical properties, morphology, and biodegradability were examined. Fourier-transform infrared (FTIR) spectroscopy confirmed the successful incorporation of chitosan and CEO. Scanning electron microscopy (SEM) revealed surface roughness and cavities due to incomplete chitosan dissolution. The optimized formulation (3% starch, 5 ml glycerol, 5% chitosan, and 5% CEO) exhibited superior mechanical properties, with a tensile strength of 710.98 MPa, Young's modulus of 582.65 MPa, and elongation at break of 121.4%. Soil burial tests demonstrated complete degradation within 14 days. These findings highlight the potential of mangrove starch-based bioplastics for food packaging and eco-friendly applications.

Keywords:
Bioplastic, mangrove starch, plasticizer, chitosan, CMC, glycerol, carrageenan

1 Introduction

Plastics are widely used in almost every place in the world, such as packaging, fashion accessories, kitchenware, toys, cell phones, and others. They are also widely used by large-scale industries from pharmaceuticals to automotive. It would be very hard to imagine life without plastics.

Nowadays, many people are beginning to realize the harmful effects of plastic materials obtained from petrochemicals on the environment. Southeast Asia, Indonesia is battling one of the greatest environmental challenges of our time; plastic waste generation is increasing population growth. Unfortunately, the handling of plastic waste in landfills is not quite enough.

Biodegradable plastic is an eco-friendly alternative solution to reducing the amount of plastic waste sent to landfills. Other significant classes of biodegradable plastics include starch-derived biopolymers.

Although most plastics are designed to resist degradation, biodegradable plastics are specifically formulated so that can eventually break down and return to natural compounds with minimal environmental impact [1]. The characteristic of those final products of degradation is that they are non-toxic, thus normally present in nature and living organisms, and represent food to microorganisms [2].

This research aims to manufacture eco-friendly disposable products such as thin plastic sheets for food packaging and daily use things made from natural raw material, large-leafed mangrove (Bruguiera gymnorrizha) fruit (propagule; hypocotyl) starch including additive variations: glycerol as a plasticizer, chitosan as filler, Carboxymethyl Cellulose (CMC) and carrageenan as thickeners, also Clove Essential Oil, CEO (Syzigium Aromaticum) as antioxidant agent. Therefore, we decided to complete a guide to understand and measure: its morphology and surface topography; infrared radiation absorption; physical and mechanical properties; and degradation of plastic materials decomposed by microorganisms in the soil.

Fig. 1. Bruguiera gymnorrizha (propagule or hypocotyl) fruit

Bruguiera gymnorrhiza is one of the most important and widespread mangrove species in the Pacific. As well as its botanical name, Bruguiera gymnorrhiza is known by many common, or vernacular names. These include: Madura: lindur; and Aceh: peurtut. It is a mangrove tree that belongs to the family Rhizophoraceae, and genus Bruguiera. It is found on the seaward side of mangrove swamps, often in the company of Rhizophora [3], particularly Mangrove Ecosystem Area, Silang Cadek, Baitussalam, Aceh Besar District [4].

Starch also known as amylum, is an important food product and biomaterial used worldwide for different purposes. Bruguiera gymnorrhiza fruit was found to have high starch content, yet it has not been optimally utilized. It contains about 57.73% starch, 31.56% amylose, and 26.17% amylopectin [5].

Plasticizers are added to plastics to aid flow and processing. They are low molecular weight materials that alter the properties, forming characteristics of the plastic [1], and allowing polymer chains to move past one another allowing wholesale flexibility [6].

Chitosan is a biodegradable biocompatible polymer derived from natural renewable resources with numerous applications in various fields. It has antibacterial and antifungal properties [7]. Different blends of chitosan with some carbohydrates were prepared to obtain Chitosan derivatives of certain physical and chemical properties [8].

CMC is an additive that is often used as a thickener and an emulsion stabilizer for food material. CMC can be synthesized from plant cellulose or cotton [9]. Chemically modified celluloses can be used to manufacture edible and biodegradable films [10].

Carrageenans are structural polysaccharides of the marine red algae (Rhodophyceae) [10]. Carrageenans are widely used in the food industry, for their gelling, thickening, and stabilizing properties [3].

An antioxidant is a molecule capable of inhibiting the oxidation of another molecule. Antioxidants break the free radical chain of reactions by sacrificing their electrons to feed free radicals, without becoming free radicals themselves [11].

Starch also known as amylum, is an important food product and biomaterial used worldwide for different purposes. Bruguiera gymnorrhiza fruit was found to have high starch content, yet it has not been optimally utilized. It contains about 57.73% starch, 31.56% amylose, and 26.17% amylopectin [5].

The study aimed to comprehensively analyze and measure the material's infrared radiation absorption, morphology and surface topography, physical and mechanical properties, and biodegradability in soil.

2 Materials and Methods

2.1 Materials and Apparatuses

All chemical substances purposed in this research were Bruguiera gymnorrizha fruit starch, chitosan, acetic acid (glacial) 100%, silica gel, glycerol 98%, carrageenan, CMC, CEO, and aquadest.

Hence, the apparatuses used were a beaker glass 500 ml; Erlenmeyer® flask 250 ml; laboratory bottle clear glass 250 ml; glass funnels; glass spatulas; glass jars; battery-powered water pump; electronic digital scale; magnetic stirrer with hot plate; non-contact infrared digital thermometer; electric oven; desicator; glass specimen molds (200 mm x 200 mm); SEM microscope (Carl-Zeiss® model EVO Germany); FT-IR spectrometer (ThermoScientific® iS50 Madison USA); universal testing machine (3369 Norwood® USA); 4-1 soil survey instrument; burial ground; mini gardening tools; PE nets; handscoon; and dropping pipette 5 ml.

2.2 AMC Manufacturing Procedure

The treatments include several chemical compositions that would be investigated for Bruguiera gymnorrizha fruit starch as the main raw material, filler and other additives used to manufacture the biodegradable plastics, either 'with' or 'without' antioxidant agent CEO are as follows (Table 1).

Table 1. The chemical compositions of polymer mixtures (solutions)

Starch (g)	Glycerol (ml)	Chitosan % (m/m)	CMC % (m/V)	Carrageenan % (m/V)
without antioxidant agent CEO				
3	2.7	4	1	1
3	5	5	1	1
3	9	6	1	1
with antioxidant agent CEO 5% (m/m)				
3	2.7	4	1	1
3	5	5	1	1
3	9	6	1	1
with antioxidant agent CEO 6% (m/m)				
3	2.7	4	1	1
3	5	5	1	1
3	9	6	1	1
with antioxidant agent CEO 7% (m/m				
3	2.7	4	1	1
3	5	5	1	1
3	9	6	1	1
with antioxidant agent CEO 8% (m/m)				
3	2.7	4	1	1
3	5	5	1	1
3	9	6	1	1

2.3 Sample Preparation

The preparation of a polymer mixture without antioxidant agent CEO, such as the chemical composition of 3 grams of starches; 5 milliliters of glycerol; 5% (m/m) chitosan; 1% (m/V) CMC; and 1% (m/V) carrageenan is obtained by dissolving 5% (m/m) chitosan powder in 100 ml of 1% (V/V) acetic acid solution come in a laboratory bottle, then stirred using a magnetic stirrer for 25 minutes at a stirring speed of 100 RPM. Heat the mixture on the burner, hot plate at 75℃ for 20 minutes until homogeneous. Add 3 grams of starches to the acetic acid-chitosan solution and stir the mixture at 100 RPM. Reheat the mixture on the burner at 75℃ for 30 minutes, until a homogeneous mixture is chemically created.

Next tasks, prepare the other laboratory bottle to dissolve 1% (m/V) CMC powder and 1% (m/V) carrageenan powder in 100 ml of aquadest. Then, stir the mixture at 100 RPM. Heat the mixture on the burner at 95℃ for 45 minutes until homogeneous.

Pour entire mixtures into a beaker glass along with glycerol, turn down the heat on the burner immediately around 80℃, stir the mixture at 100 RPM for 60 minutes until homogeneous, and shrink precisely at 150 ml.

There is no difference between 'without' and 'with' antioxidant agents in the preparation of polymer mixtures, such as the chemical composition having an antioxidant content of 4% (m/m) CEO. We had simply put the CEO into such the beaker glass, before pouring the plasticizer. Then, stir the mixture at 100 RPM for 45 minutes and maintain the temperature of the mixture constantly at 80℃.

Next tasks, polish the mold and release waxes flatly on the inside surface of the mold. Then, pour the polymer solution into the mold, flatten it, and let it cold for 6 hours at ambient temperature. Place the mold that contained the polymer solution in the electric oven and dry it at 65℃ for 24 hours.

After drying, the mold was removed from the oven and cooled to room temperature. Start by slowly and gently releasing the sample. Create the object into the square test sample sheets 1 inch wide by 1 inch long for studying in, FT-IR spectroscopic analysis, SEM visual inspection, and soil burial test. Besides, the specimens for the tensile test are created by referring to ASTM D882-10 standard at 1 inch wide by 100 mm long. Put all samples in a desiccator with silica gel, while waiting for further identification analyses.

As the specimen elongates the resistance of the specimen increases and is detected by a load cell [12]. This load value (force, F) is recorded by the instrument. The elongation of the specimen is continued until a rupture of the specimen is observed. The tensile strength (σ) is calculated by Eq. (1), where F is force load (N), A is cross-section area (m^2)

$$\sigma = F/A \ (N/m^2) \quad (1)$$

The relationship between stress (σ) and strain (ε) according to Hooke's Law is formulated by Eq. (2) and Eq. (3), where E is Young's Modulus (Elasticity), ΔL is Change in length (elongation) in m L= Original length (gauge length) in m.

$$E = \sigma/\varepsilon \ (N/m^2) \quad (2)$$

$$\varepsilon = \Delta L/L \quad (3)$$

The percentage elongation of the test piece at fracture is also derived [13]. This is a measure of the ductility of the material. The two halves of the broken test piece are fitted together (Fig. 2).

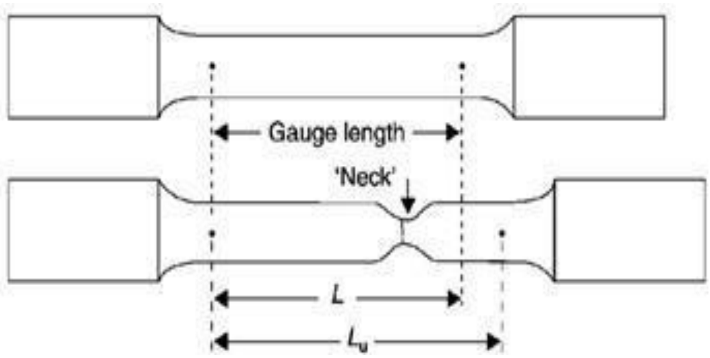

Fig. 2. The determination of the percentage elongation (Higgins, 2006)

The extended gauge length is measured (Eq. 4), where, Lu is the 'ultimate' gauge length in m.

$$\text{Elongation (\%)} = \Delta L/L \times 100 = (Lu - L)/L \times 100 \quad (4)$$

3 Results and discussion

3.1 Products

Referring to the work in the context of the biodegradability perspective, we got the averaged plastic sheets were smooth surface contours, translucent, light brown, no-crack, and flexural. They are similar to previous studies reported by [14], whereas to repair the surface structure, cellulose derivatives were used, that is CMC. Previous research also confirmed, CMC is a cellulose derivative that directly binds water and is often used as a smoothing agent in texture, as shown in Fig. 3 [15]:

Fig. 3. A physical appearance of mangrove fruit (Bruguiera gymnorrizha) starch-based biodegradable plastic

3.2 FT-IR Spectroscopic Analysis

The FT-IR spectrum below shows plastic sample has a mixture consisting of 3 grams of starch; 5 ml of glycerol; 5% (m/m) of chitosan; 1% (m/V) of CMC; and 1% (m/V) of carrageenan. Also, a plastic sample with antioxidant content, 7% (m/m) of CEO, is shown in Table 2. The FT-IR spectral profiles for plastic samples containing antioxidant agents or not as shown in Fig. 4.

Table 2. Functional group analysis

Contents	Wave Number (v, cm^{-1})	Types of Chemical Bonds /Functional Groups	Types of Compounds
3 gram of starch; 5 ml of glycerol; 5% (m/m) of chitosan; 1% (m/V) of CMC; 1% (m/V) of carrageen.	3761.19 to 3280.92	N-H Amine	Chitosan
	3761.19 to 3280.92	O-H Hydroxyl	starch; chitosan; CMC; carrageenan
	1257.59	C-O Ester	Glycerol; carrageenan
	1600.92	C=C Alkene	starch; glycerol; CMC
	1734.01	C=O Carbonyl	CMC
	3755.4 to 3263.56	N-H Amine	Chitosan
3 grams of starch; 5 ml of glycerol; 5% (m/m) of chitosan; 1% (m/V) of CMC; 1% (m/V) of carrageen. 7% (m/m) of CEO	3755.4 to 3263.56	O-H Hydroxyl	starch; chitosan; CMC; carrageenan
	1259.52	C-O Ester	Glyserol; carragernan
	1595.13	C=C Alkene	starch; glycerol; CMC
	1741.72	C=O Carbonyl	CMC
	1413.82 to 1373.32	C=H *Bending*	Antioxidant

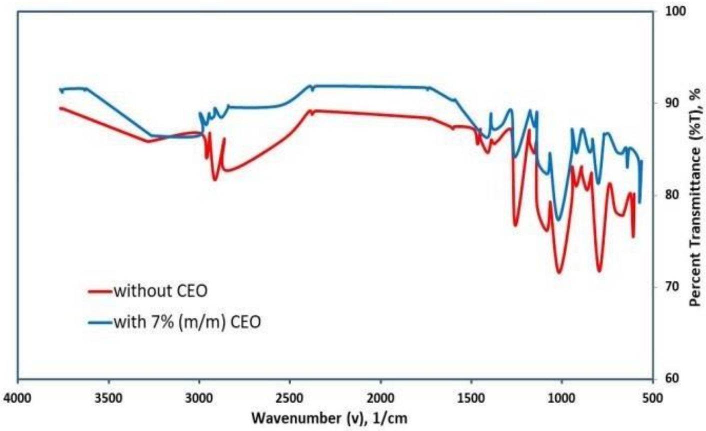

Fig. 4. The FT-IR spectral profiles for biodegradable plastic samples containing antioxidant agents or not

The plastic sample consists of 3 grams of starch: 5 ml of glycerol; 5% (m/m) of chitosan; 1% (m/V) of CMC; 1% (m/V) of CMC and 5% (m/m) of antioxidant agent, CEO has 5 functional groups consisting of N-H (amine) group in range of 3761.19 cm^{-1} to 3280.92 cm^{-1} indicating the presence of chitosan.

Within the same range, there is the -OH (hydroxyl) group, which indicates the presence of starch, chitosan, CMC, and carrageenan in the plastic sample. Then, the addition of CMC occurs C=O (carbonyl) absorption at the absorption peak of 1734.01 cm^{-1} and breaks the C-O (ester) bond in the range of 1257.59 cm^{-1} due to the addition of glycerol plasticizer and carrageenan thickener. Next is the C=C (alkene) group, which indicates the presence of starch, glycerol, and CMC in the plastic sample in the range of 1600.92 cm^{-1}.

Almost similar to the functional group analysis for biodegradable plastic samples without antioxidants, a sample with a concentration of 7% (m/V) of antioxidant agent, CEO has 6 functional groups consisting of N-H (amine) group in range of 3755.4 cm^{-1} to 3263.56 cm^{-1} indicating the presence of chitosan and within the same range, there is the -OH (hydroxyl) group, which indicates the presence of starch, chitosan, CMC and carrageenan in the plastic sample. Then, the addition of CMC causes the C=O (carbonyl) absorption in the range of 1741.72 cm^{-1} and breaks the C-O (ester) bond in the range of 1259.52 cm^{-1} due to the addition of glycerol plasticizer and carrageenan thickener. Next is the C=C (alkene) group, which indicates the presence of a mixture of starch, glycerol, and CMC in the plastic sample in the range of 1595.13 cm^{-1}. However, in the range of 1413.82 cm^{-1} to 1373.32 cm^{-1} at the absorption peak of C-H has occurred a bending vibration and sheared the bond angles into the polymer molecules (C-C, C=C, C-O, C=O), where frequencies absorbed by the polymer molecules are much lower than the infrared radiation frequencies transmitted by the FT-IR spectrometer.

3.3 Scanning Electron Microscopy (SEM)

The analysis of the surface morphology, structure, and properties of biodegradable plastic samples with the composition: 3 grams of starch; 2.7 ml of glycerol; 4% (m/m) of chitosan; 1% (m/V) of CMC; and 1% (m/V) of carrageenan, also including a biodegradable plastic sample with concentration of antioxidant agent, 8% (m/V) of CEO, as clearly shown in Fig. 5 and 6 below with 2500 times optical magnification.

Referring to SEM image (Fig. 5) provides information about the surface morphology and topography of the biodegradable plastic sample without an antioxidant agent. There are some white spots on the surface, according to [16], presumably due to the lack of homogeneity between other polymer mixtures and Bruguiera gymnorrhiza fruit starch granules. One is called, the presence of chitosan is hydrophobicity and insoluble in water, except at an acidic pH (minimum at 1% V/V) and similar to [17], chitosan is insoluble in water and in alkali and organic solvents but is soluble in most solutions of organic acids, such as acetic and formic acid, at a pH below 6.

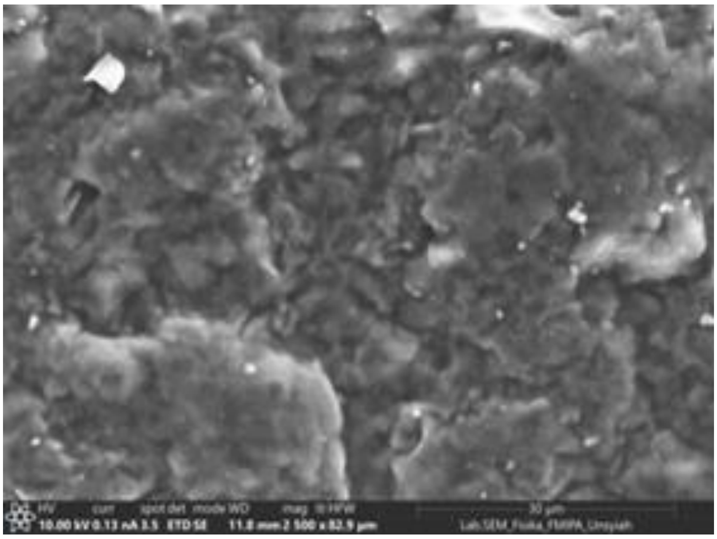

Fig. 5. A morphology and topography of a biodegradable plastic sample without an antioxidant agent at 2500x optical magnification

Besides, there is a physical interaction between the starch granules and the chitosan solution. This indicated that granules are difficult to dissolve or the rest of the chitosan incompletely dissolved during the stirring process. The plastic surface was exposed to the air. It tends to be rough, raises several cavities, and creates grooves.

Almost similar to the morphological or topographical analysis of biodegradable plastic samples without antioxidants. Fig. 6 provides information about the presence of white spots on the surface.

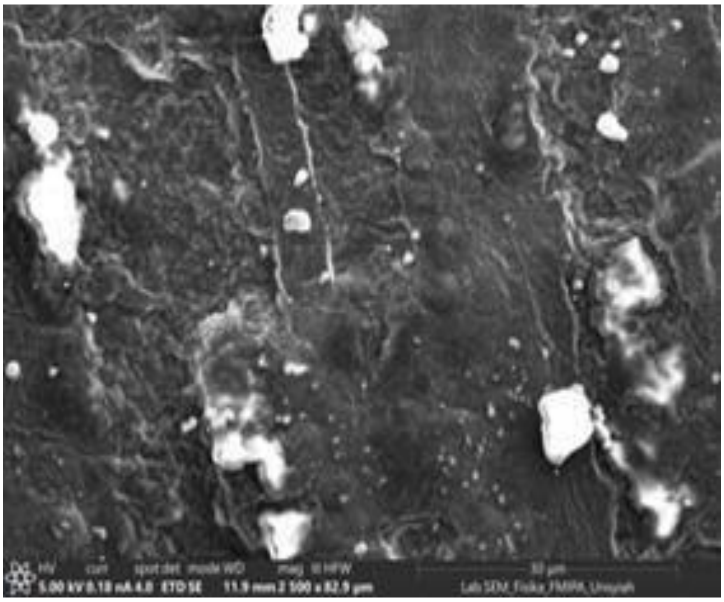

Fig. 6. A surface morphology and topography of biodegradable plastic sample with antioxidant agent: 8% (m/V) of CEO at 2500x optical magnification

However, they were more numerous than in plastic samples without antioxidants. Presumably due to the lack of homogeneity between other polymer mixtures and Bruguiera gymnorrhiza fruit starch granules.

However, there is a physical interaction between the starch granules and the chitosan solution. This indicated that granules are difficult to dissolve or the rest of the chitosan incompletely dissolved during the stirring process. The plastic surface was a little exposed to the air. It tends to be kind of smooth, arises few cavities and grooves. An aspect regarding the effect of the addition of antioxidants, CEO has raised the air bubbles on the surface; most probably occurred the formation of an incomplete emulsion between clove essential oil and water as a solvent.

3.4 Tensile test

This mechanical test briefly refers to Designation: D882–10, the standard test method for tensile properties of thin plastic sheeting, as a reference to determine the tensile strength (σ), elastic modulus (E), elongation (ΔL), and other mechanical properties of materials that depends on its chemical composition. The manufactured samples are shown in Fig. 7.

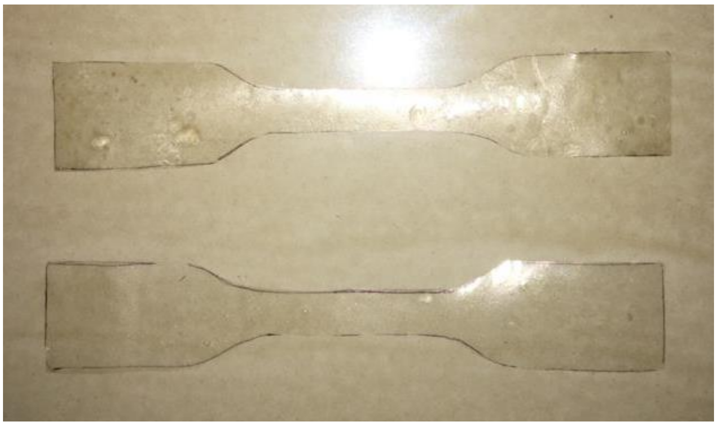

Fig. 7. The manufactured sample for tensile test

3.4.1 Tensile strength

Fig. 8 shows, the effect of glycerol plasticizer addition of 2.7 ml, 5 ml, and 9 ml to the polymer solution against the tensile strength of biodegradable plastics. According to the curve, the tensile strength tends to decrease along with the addition of glycerol. It describes an interaction between polymers and solvents that tend to reduce, form the hydrogen bonds between them, and facilitate their movement. It was similar to [17], if the plasticizer concentration is raised above the antiplasticization range, the modulus, and tensile strength will begin to fall, while the impact strength and permeability will increase (i.e., plasticization begins).

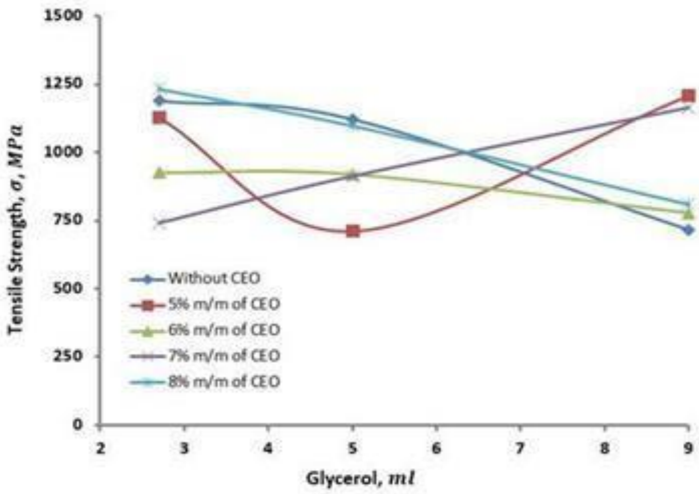

Fig. 8. The effect of the increase in glycerol concentration against the tensile strength (σ) of Bruguiera gymnorrizha fruit starch-based biodegradable plastics 'without' and 'with' antioxidants CEO

However, the plastic samples with antioxidants at a concentration of 5% m/m; and 7% m/m, show an increase in flexural strength and perhaps the non-uniformity thickness of the test specimens. It was similar to [12], the specimen thickness can affect the results. At a given span, the flexural strength increases as the specimen thickness is increased. The modulus of a material generally increases with the increasing strain rate.

Fig. 9 shows the effect of chitosan addition at concentrations of 4% m/m, 5% m/m, and 6% m/m on this plastic specimen, which seems to affect its mechanical properties.

It becomes more brittle if the chitosan concentration is increased, then its tensile strength tends to decrease, as happened to the other specimens without antioxidants and with antioxidants

CEO at concentrations of 6% m/m and 8% m/m. It was similar to [18], related to a study on starch-based bioplastics, another example is the banana weevil starch-chitosan-based edible film in which its tensile strength tends to increase along with increasing the chitosan concentration, and then it tends to decrease after passing the maximum value.

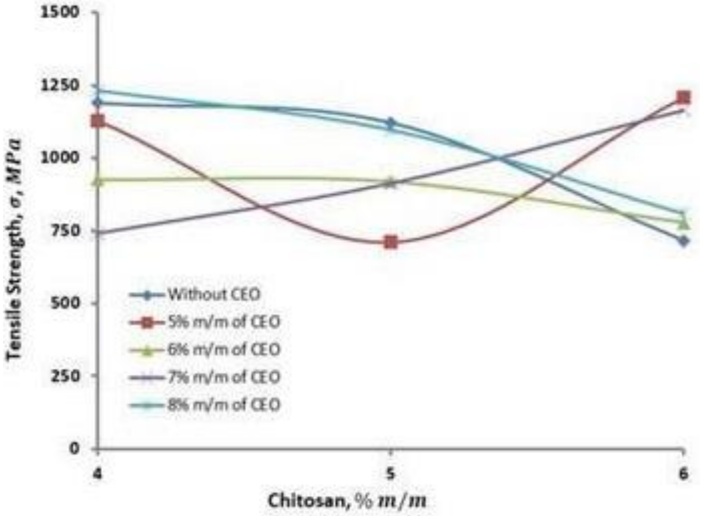

Fig. 9. The effect of the increase in chitosan concentration against the tensile strength (σ) of Bruguiera gymnorrizha fruit starch-based biodegradable plastics 'without' and 'with' antioxidants CEO

Fig. 10 describes the influence of antioxidant addition with various concentrations of 0%, 5%, 6%, 7%, and 8% (m/m) of clove essential oil against its mechanical properties that tend to decrease its tensile strength.

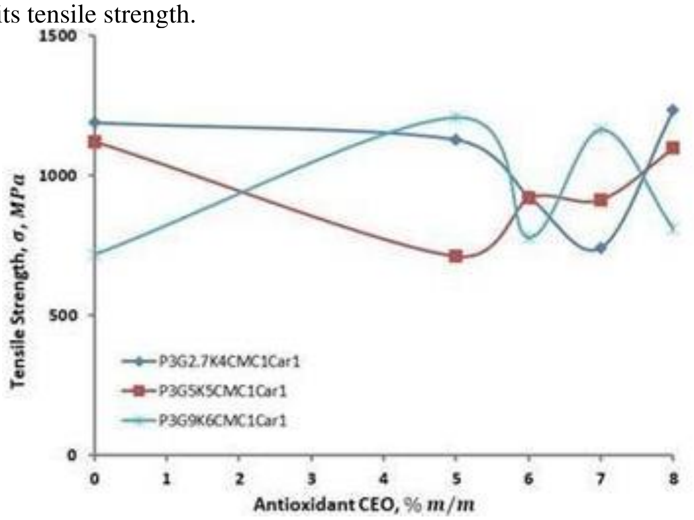

Fig. 10. Tensile strength (σ) in MPa for Bruguiera gymnorrhiza fruit starch-based biodegradable plastics at various concentrations of antioxidants, CEO

It is similar to [19], essential oils plasticize the polymer, reducing its tensile strength and increasing its elongation at break. The reason for the decrease in tensile strength is that the essential oil components weaken the interactions between the polymer molecules, increasing the flexibility of the polymer chains.

3.4.2 Young's Modulus (Elasticity)

Fig. 11 below shows the effect of glycerol addition as a plasticizer to their polymer solutions for Bruguiera gymnorrhiza fruit starch-based biodegradable plastics, against their Young's Modulus.

According to [17], if the plasticizer concentration is raised above the antiplasticization range, the modulus, and tensile strength will begin to fall, while the impact strength and permeability will increase (i.e., plasticization begins). According to [20], having a lower modulus of elasticity means that the bioplastic is highly elastic, but having a higher modulus of elasticity means that the bioplastic is a high-stiffness material.

Hereafter, the highest modulus of elasticity occurs when the plasticizer is added at a concentration of 9 ml which produces a stiffness biodegradable plastic.

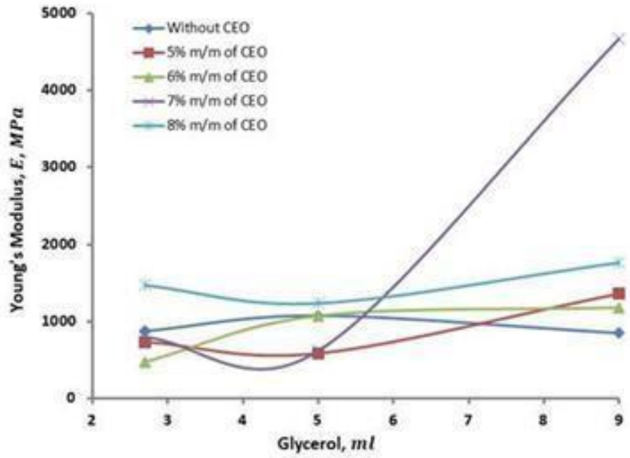

Fig. 11. The effect of the glycerol addition against the Young's Modulus (E) in MPa for Bruguiera gymnorrhiza fruit starch-based biodegradable plastics at various concentrations of antioxidants, CEO

Fig. 12 shows the addition of chitosan as filler into the biodegradable plastics has increased their mechanical properties, thereby increasing their Young's Modulus. Therefore, the highest modulus occurs at a concentration of 6% m/m which produces a stiffness biodegradable plastic. Meanwhile, Fig. 13 shows, that the addition of clove essential oil as an antioxidant can reduce the mechanical properties of biodegradable plastics which will also reduce their modulus.

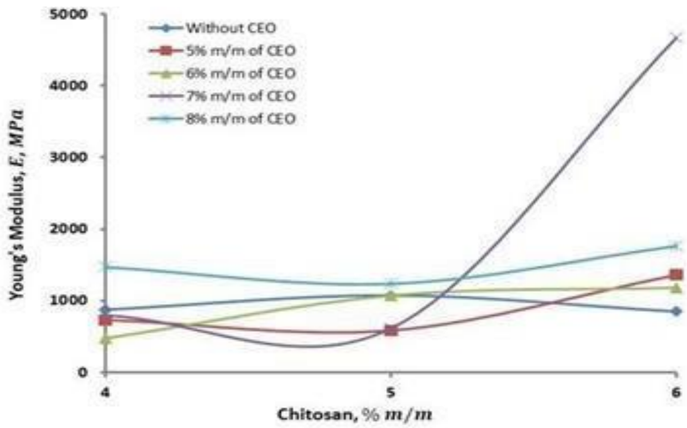

Fig. 12. The Effect of chitosan concentration on Young's Modulus (E) in MPa for Bruguiera gymnorrhiza fruit starch-based biodegradable plastics 'without' and 'with' antioxidant additives CEO

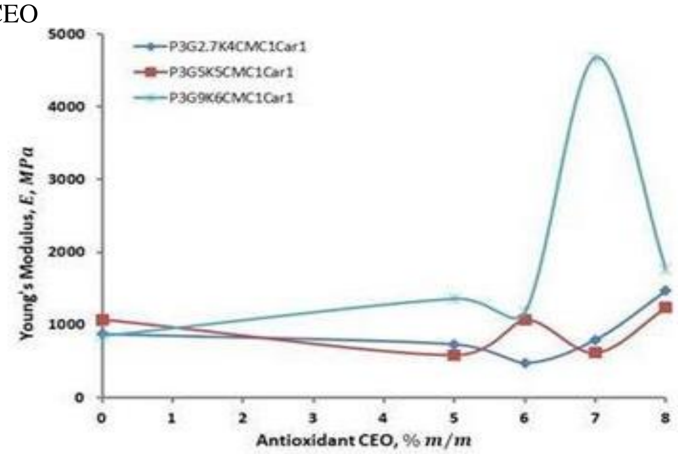

Fig. 13. Young's Modulus (E) in MPa for Bruguiera gymnorrhiza fruit starch-based biodegradable plastics at various concentrations of antioxidants, CEO.

It occurs on a specimen of plastic material with the chemical composition of 3 grams of starch; 9 ml of glycerol; 6% (m/m) of chitosan; CMC and carrageenan each have 1% (m/V) and antioxidant, CEO at concentration of 7% m/m. Its chemical composition has mechanical property tend to increase, presumably due to the non-uniformity of thickness with other test specimens, thereby it may affect the results.

3.4.3 Elongation (Change in Length)

Elongation indicates the ability of a material to undergo significant deformation before failure. The extension percentage is categorized as bad if it is less than 10% and is good if the extension percentage is more than 50% [21].

Hereafter, the change in length (elongation, ΔL) is determined by the original length (gauge length) of the specimen (L) to the 'ultimate' gauge length (LU) which is exhibited in the relationship shown by a curved graph between the addition of glycerol against the change in length (elongation) as shown in Fig. 14.

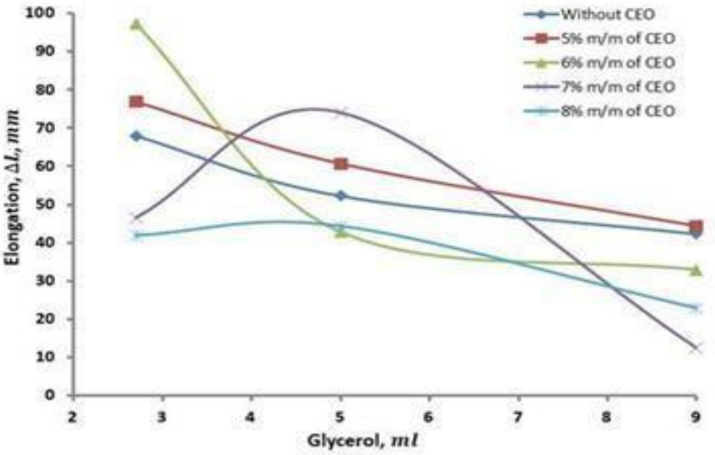

Fig. 14. Addition of glycerol in ml against change in length (ΔL) in mm for Bruguiera gymnorrhixa fruit starch-based biodegradable plastics 'without' and 'with' antioxidants, CEO

According to [22], increasing the amount of hydrogen bonds formed has created the percent elongation value smaller and produced the bioplastic become stiffer and less elastic.

According to the following statements implied in the curved graph above between the addition of glycerol against the change in length (elongation) described, an addition of glycerol plasticizer into a polymer mixture of biodegradable plastic at a concentration of 9 ml tends to produce a biodegradable plastic that is stiffer and less elastic, along as decreasing its change in length (elongation).

According to the curved graph in Fig. 15, the elongations tend to decrease along with the addition of chitosan concentration in their polymer mixture, thereby producing biodegradable plastics that are stiffer and less elastic.

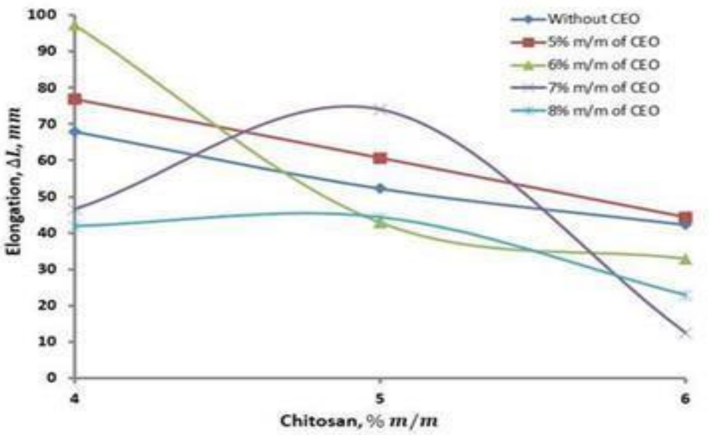

Fig. 15. Addition of chitosan against change in length (ΔL) in mm for Bruguiera gymnorrhiza fruit starch-based biodegradable plastics 'without' and 'with' antioxidants, CEO

As reported by [23], elongation is inversely proportional to tensile strength. The addition of chitosan resulted in a decreased value of elongation. It indicates that the amount of filler chitosan causes a decrease in the intermolecular bonding distance. The decreasing bond distance is due to the increasing number of hydrogen bonds formed between molecules of chitosan-amylose-amylopectin.

Based on the calculations, a biodegradable plastic with an elongation percentage of more than 50%, occurs mostly in all chemical compositions containing antioxidants or not at concentrations of 5% m/m and 6% m/m, except at concentrations of 7% m/m and 8% m/m has a chemical composition consists of 3 gram of starch; 2.7 ml of glycerol; 4% (m/m) of chitosan; CMC and carrageenan each have 1% (m/V); and another one consists of 3 gram of starch; 5 ml of glycerol; 5% (m/m) of chitosan; CMC and carrageenan each have 1% (m/V) that have a good elongation percentage, the rest less (see Fig. 16). The discrepancy in the modulus of elasticity could be attributed to the specific composition of the bioplastic, which includes starch, glycerol, chitosan, CMC, and carrageenan. Variations in the concentrations of these components, especially chitosan and glycerol, can markedly influence the mechanical properties. For instance, higher chitosan content can increase stiffness, while glycerol acts as a plasticizer, enhancing flexibility. To align the bioplastic's properties with SNI standards, consider adjusting the formulation. Reducing the chitosan concentration or increasing the glycerol content may help lower the modulus of elasticity. However, it's essential to balance these adjustments to maintain other desirable properties, such as tensile strength and biodegradability.

However, there was a mistake on the test specimen at a chitosan concentration of 7% m/m, presumably due to the non-uniformity of thickness with others, thereby it may have affected its result.

According to Indonesian National Standard (SNI) 7188.7:2016, the required elongation range for bioplastics is 21% to 220%**,** while the modulus of elasticity is specified between 117 MPa and 137 MPa. This means that your material meets the elongation requirement of SNI**.** However, its modulus of elasticity exceeds the upper limit set by SNI. This indicates that the material is stiffer than what is typically required under the standard for bioplastics.

The addition of antioxidant concentration can reduce the mechanical properties of biodegradable plastics, thus their change in lengths (elongations, ΔL). It was probably caused by the clove essential oil having hydrophobic properties which make it difficult to dissolve starch In the end will produce a biodegradable plastic that is less elastic, stiffer, and brittle.

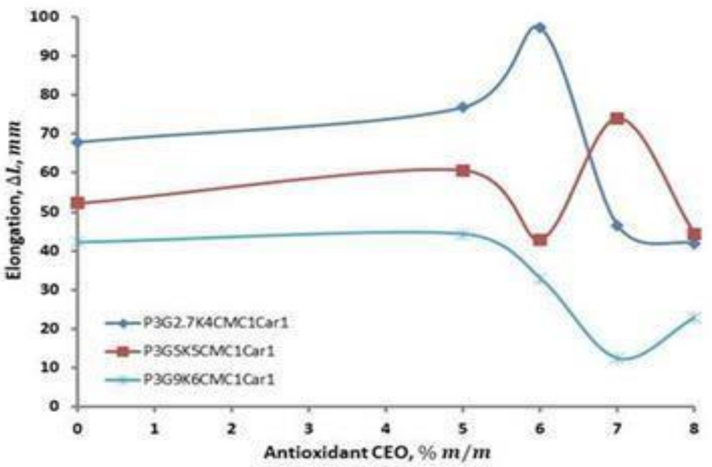

Fig. 16. Change in length (elongation, ΔL) in mm for Bruguiera gymnorrhiza fruit starch-based biodegradable plastics at various concentrations of antioxidants, CEO

3.5 Soil Burial Test

Environmental factors, such as microorganisms, temperature, humidity, and soil pH affect the degradation rate of polymeric materials which will undergo the important changes in their structure, morphology and properties. The situations in the burial process of biodegradable plastic samples in the soil by the first day are shown in Fig. 17.

(a) The digging furrow process at 15 cm deep by 1.5 m long which provided for the soil burial test. Then, measure temperature, moisture (RH) and pH at each point that represents one of the plastic samples are located in the soil;
(b) All samples were located on the first layer of polyethylene mesh;
(c) The samples are buried with soil at ±1 cm in thickness
(d) The second laying of polyethylene mesh layer;
(e) The samples are reburied by the second soil pile. Then, give a sign while waiting for the next identification process.

Fig. 17. A situation in the burial process of biodegradable plastic samples in the soil by the first day

The samples were periodically taken outdoors, and temperature, pH, and soil relative humidity (RH) were measured and recorded weekly.

A week later, the buried samples were exhumed, cleaned by the removal of dirt particles, and then dried in an oven preheated to a constant temperature of 50°C for 24 hours.

Furthermore, the samples are placed in a desiccator before undergoing the image-capturing processes. The soil burial test procedures continue to be carried out periodically by the following week period until the samples have completely degraded.

During molding the material is subjected to the first two of these so it is necessary to incorporate stabilizers and antioxidants into the plastic to maintain the properties of the material. These additives also help to delay subsequent degradation for an acceptably long time [1] and to prolong their useful lifetime [24].

After all, the samples have been completely degraded, it can be concluded, that the biodegradable plastic test sample contains 3 grams of Bruguiera gymnorrhiza fruit starch, 5 ml of glycerol plasticizer, 5% m/m chitosan filler, CMC and carrageenan thickeners each have 1% m/V, including antioxidants, CEO at a concentration of 5% m/m is the plastic sample took the longest damage and began to break down at day 14, where it began to look crack, torn and lose its weight (Fig. 18). This is because the addition of antioxidants, CEO can delay degradation, thus microorganisms in the soil need a longer time to break the long polymer chains into the shorter ones.

Chemical Composition	Representation of Soil Burial Test (SBT) (Before and After Degradation)		
	Day 1	Day 7	Day 14
3 g of strach 5 ml of gliserol 5% of chitosa 1% of CMC			
1% of carragee.	T = 28℃	T = 28℃	T = 28℃
5% CEO	pH = 6.8	pH = 6.8	pH = 6.8
	RH =wet	RH = wet	RH = wet

Fig. 18. The view of a sample of Soil Burial Test before and after degradation

4 Conclusion

This research developed biodegradable plastic films made from mangrove starch, chitosan, and clove essential oil using glycerol as a plasticizer. The results demonstrated promising mechanical and antioxidant properties. The optimized formulation (3% starch, 5 ml glycerol, 5% chitosan, 5% clove essential oil) exhibited high tensile strength, flexibility, and a controlled degradation rate, making it suitable for food packaging and sustainable applications. SEM analysis at 2500x magnification revealed some surface imperfections due to chitosan dispersion, indicating potential for further optimization in material blending techniques. These findings highlight the potential of mangrove-derived bioplastics as a viable alternative to synthetic polymers. Future research should explore additional biopolymer blends and industrial scalability for commercial applications.

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