



Research article

Effect of glycerol concentration and carboxy methyl cellulose on biodegradable film characteristics of seaweed waste

Sri Hidayati^{a,*}, Zulferiyenni^a, Ulfa Maulidia^a, Wisnu Satyajaya^a, Sutopo Hadi^{b,**}^a Department of Agroindustrial Technology, Universitas Lampung, Bandar Lampung, 35145, Indonesia^b Department of Chemistry, Universitas Lampung, Bandar Lampung, 35145, Indonesia

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ABSTRACT

Eucheuma cottonii waste seaweed has high cellulose content. Therefore, it could be potentially used as a raw material for biodegradable films to replace plastic. A plastic film is its moisture resistance, and this property allows plastic films to be used as packaging materials and biodegraded by microbes. This research aims to obtain a concentration of glycerol and Carboxy Methyl Cellulose (CMC) to obtain the best biodegradable film characteristics from *E. cottonii* seaweed waste. This study was conducted in factorial by using a complete randomized block design with two factors: glycerol concentration and CMC concentration. Each treatment has three levels and three replications (3 × 3). The first factor was glycerol concentration: 0.25% (G1), 0.5% (G2), and 0.75% (G3). The second factor included concentrations of 1% CMC (C1), 2% CMC (C2), and 3% CMC (C3). Then, Tensile Strength (TS), thickness, solubility, and elongation were observed. Functional group analysis was conducted by Fourier-transform infrared spectroscopy and biodegradability test. The results showed that the addition of glycerol concentrations of 0.5 and 0.7% and CMC from 1 to 3% produced tensile strengths of 23–39 MPa. These values are proportional to the tensile strength of Poly Tetra Fluoro ethyne (PTFE) and Poly Propylene (PP) synthetic plastics released by Dotmar Engineering Plastics. The biodegradability test showed that the produced biodegradable films decomposed after 14 days.

1. Introduction

The food industry is the largest user of plastic packaging (Corradini et al., 2013). Plastics are used as packaging to protect food before consumption (Ozcalik and Tihminlioglu, 2013). The plastic consumption of Indonesia reached 3 million tons in 2015 (Indonesian Olefin, Aromatic, and Plastic Industry Association, 2015). Plastics made from synthetic polymers are difficult to be decomposed using biotic components such as microorganisms and abiotic components (Ozcalik and Tihminlioglu, 2013). It will take approximately 100–500 years to completely decompose plastic in the soil (Akbar et al., 2013); therefore, plastics are considered as environmental pollutants. Biodegradable films are prepared from natural and easily renewable materials such as starch, cellulose, proteins, and fats (Benbettaieb et al., 2019). This compound can be used to replace plastics from synthetic polymers and be biodegraded by mi-

croorganisms (Falguera et al., 2011; Mostafa et al., 2018). Furthermore, biodegradable plastic can be a part of the solution, and it is one of the most critical challenges that must be overcome by the food packaging industries (Shen et al., 2020; Martins da Costa et al., 2020).

The resistance of petroleum-based plastics to degradation has led to accumulation of used plastics in various environments and become serious threat to wildlife ecosystems as well as human life. This problem implies that development of environment friendly packaging to replace conventional plastics is urgently needed. In this respect, fabrication of packaging material from abundantly available biopolymers such as starch, gluten, and guar gum, is environmentally advantageous. In addition to biodegradability, biopolymers are nontoxic and renewable (Abdul Khalil et al., 2017; Tongdeesoontorn et al., 2012).

Biodegradable films are mainly used to reduce the adverse effects of petroleum-based packaging and materials (Ntharanathan, 2003). The

* Corresponding author.

** Corresponding author.

E-mail addresses: sri.hidayatip@fp.unila.ac.id (S. Hidayati), sutopo.hadi@fmipa.unila.ac.id (S. Hadi).

biodegradable film can be biodegraded without any harmful effects (Haider et al., 2019). One material that can be used as a biodegradable film is seaweed. Seaweed has received considerable interest and attention in industries related to food, tissue engineering, biosensors, and application in organic transformations (Abdul Khalil et al., 2017; Dindar et al., 2015). Some types of seaweed consist of Rhodophyceae (algae red), Phaeophyceae (brown algae), and Chlorophyceae (green algae) (Jae-Lane and Carlos Brais, 2019). The types of seaweed that can be used are alginate, carrageenan, and agar. Biodegradable film from seaweed is transparent, non-toxic, water-soluble, and flexible, and has good mechanical properties as packaging material in food industries (Tavassoli et al., 2016). In addition, biodegradable films from seaweed are more environmentally friendly because they are easily integrated and are mainly obtained either by the direct production of polymers or by producing biochemical polymerization (Thakur et al., 2018). Biological resources have an essential role in producing novel and bio-based materials (Brodin et al., 2017).

The other potential and untapped materials are dregs from the seaweed industry by-products. The seaweed processing industry only

utilizes approximately 30%–35% of waste, and 65%–70% of waste has not been widely utilized. Seaweed waste is 76.15% water, 5.62% ash, 2.32% protein, 0.11% fat, 15.8% carbohydrates, 17.47% cellulose component, 21.16% hemicellulose, and 8.23% lignin. *Eucheuma cottonii* seaweed has high cellulose content, thus making it a potential raw material for biodegradable films. Cellulose film has been reported to have high water and microwave heating resistances (Sudharsan et al., 2016). Seaweed and cellulose both exhibit interesting film-forming properties (Abdul Khalil et al., 2017). Seaweed as plastic raw material is rigid and brittle; therefore, it cannot be used for packaging directly and requires the addition of a plasticizer during film processing (Deepa et al., 2016; Sadeghizadeh-Yazdi et al., 2019). The most commonly used plasticizers are glycerol and sorbitol. The widespread use of glycerol and sorbitol is potential since they are stable and non-toxic. Plasticizers can increase the flexibility and permeability of water vapor permeability (Sobral et al., 2001) and reduce stiffness. Glycerol concentration usually is related to matrix basis (starch, protein). Glycerol concentration of 10% and carrageenan 3% concentration produced an edible film with a thickness of $78.52 \pm 5.12 \mu\text{m}$, the water content of $18.84 \pm 0.18\%$, solubility $64.95 \pm$

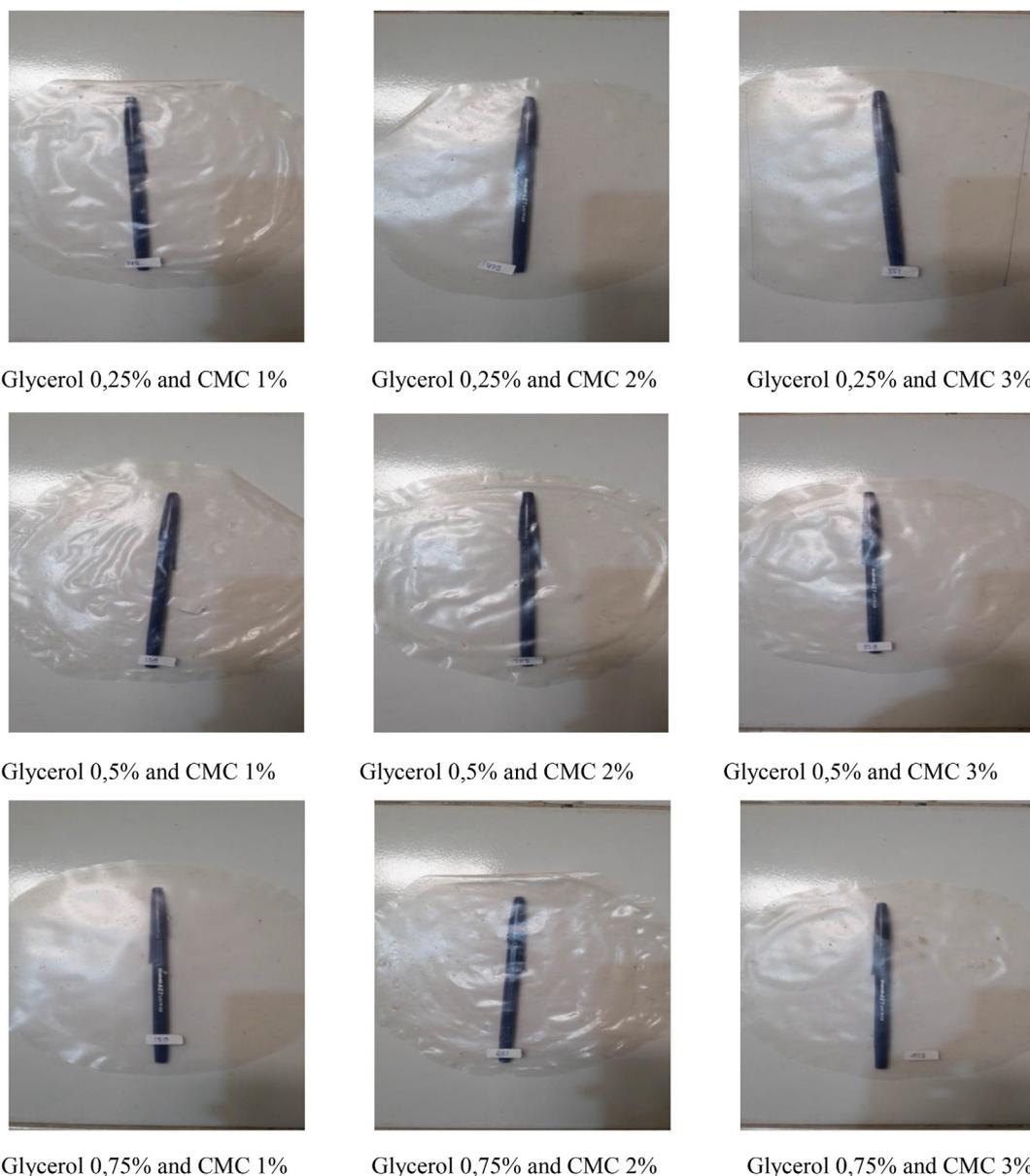


Figure 1. Visual visibility of biodegradable film from waste seaweed.

9.65%, tensile strength 4.65 ± 1.42 MPa, and elongation of $16.67 \pm 0.58\%$ (Rusli et al., 2017). Seaweed pulp cellulose content is 17.47%, although CMC is derived from cellulose, it has undergone chemical modification. CMC is derived from cellulose as an anionic linear polysaccharide (Biswal and Singh, 2004). This compound has a high viscosity, is non-toxic, is hypoallergenic, is relatively inexpensive, and is an environmentally friendly product (Criado et al., 2016). CMC can bind and absorb water because it has a hydroxyl group, which allows it to be degradable (Nie et al., 2004). CMC required to control water content, improve texture, and stability (Li et al. 2008). CMC can increase viscosity and tensile strength but decrease elongation percent (Tongdeesoontorn et al., 2012). CMC can increase ionic and chemical crosslinking to improve the mechanical properties of biodegradable film products (Ma et al., 2008; Yadav et al., 2014). CMC is a barrier against oxygen, carbon dioxide, and lipids, so it is very effective in improving the properties of the film (Hu et al., 2016). Water-soluble heteropolysaccharides with high molecular weight can produce films with the desired texture, moisture, water mobility, and product stability (Li et al., 2008).

Some researches on the process of producing biodegradable films from seaweed are available in the literature, such as using κ -carrageenan and chitosan in the presence of various organic hydroxy acids and plasticizers (Park et al., 2001), the effect of sodium alginate and κ carrageenan (Farhan and Hani, 2017), Gac pulp (*Momordica cochinchinensis*) and glycerol on film properties (Tran et al., 2020); bamboo and red seaweed (*Kappaphycus alvarezii*) (Hasan et al., 2019); seaweed (*K. alvarezii*) (Siah et al., 2015). However, not many studies have utilized solid waste from *E. cottonii* seaweed to produce a biodegradable film. The application of research results is expected to be for food packaging. Therefore, the purpose of the current study is to identify the effect of glycerol and CMC concentration on the biodegradable film characteristics of seaweed waste.

2. Experimental

2.1. Materials

The materials used were sea-soluble *Euchemacottoniisp.* from Kalianda Lampung Indonesia, glycerol as plasticizer (Rendys Chem), 100% pure vegetable glycerol, CMC food grade (Wealthy manufactured by Changsu Wealthy Science and Technology Co Ltd., China) with physical appearance as fine powder grade, aquadest, 6% tapioca, 2% H₂O₂, and soil as a decomposing medium. The tools used included a hydraulic universal

$$\text{Solubility (\%)} = \frac{\text{Initial weight (g)} - \text{dry weight (g)} - \text{cup weight (g)}}{\text{Initial sample weight (g)}} \times 100\% \quad (2)$$

testing machine (UTM) for tensile strength (TS) and thickness tests, as well as a Fourier-transform infrared (FTIR) spectroscopy.

2.2. Method

The film-making process: a total of 5 g of seaweed cellulose was fed into a 250 ml Erlenmeyer flask. After that, 6% tapioca (w/v), 15 ml ethanol, glycerol, and CMC were added according to the treatment. The mixture was dissolved with 50 ml aquadest and heated at 70 °C by using a hot plate for 30 minutes. The solution was removed from bubbles. The solution was poured on a 20 × 20 cm glass and was left to stand at room temperature for 48 hours. Tapioca addition was expected to improve the biodegradable film properties of cellulose material. In addition, starch or carbohydrates played a role in helping to obtain the right thickness for film formation. The function of adding ethanol was to simplify the process of dissolving the gel during the printing process.

2.3. Statistical analysis

The treatments were arranged in a completely randomized block design with two factors consisting of three levels and three replications (3 × 3). The first factor included glycerol concentrations of 0.25% (G1), 0.5% (G2), and 0.75% (G3). The second factor included concentrations of 1% CMC (C1), 2% CMC (C2), and 3% CMC (C3). The homogeneity of variances was tested by Bartlett's test, and the data addition was tested by Tuckey's test. The data were further processed by the least significant difference tests at the 5%. Data for biodegradability testing were presented in the form of graphics and were discussed descriptively.

2.4. Tensile strength and thickness

A UTM (Orientec Co. Ltd.) with a UCT-5T model was used for the testing. The sample sheets were cut using an ASTM D638 M-III dumbbell cutter. The test conditions were 27 °C temperature, 65% humidity in the test chamber, 1 mm/min tensile velocity, and 10% load cell scale of 50 N (American Society for Testing and Materials, 1993). The formula of the TS used as shown in Eq. (1):

$$t = \frac{F_{\max}}{A} \quad (1)$$

Where:

- t: TS (MPa)
- F_{max}: the force of TS (N)
- A: sample surface area (mm²)

The sample thickness was measured in three positions, namely, the top, middle, and bottom of the membrane (Gontard et al., 1992). The thickness value was averaged according to the thickness of the sample.

2.5. Solubility

The biodegradable plastic solubility test was performed in water by inserting a 1 cm × 5 cm plastic film sheet into a vessel containing 250 ml water (Gontard et al., 1992). Water solubility was expressed as a percentage of water-soluble film after one week of immersion (Eq. (2)).

2.6. Elongation

The percent elongation test was measured by MPY Testing Machine (PA-104-30, Ltd., Tokyo, Japan). Prior to the measurement, a 2.5 cm × 15 cm sample sheet of the film was prepared and conditioned in the laboratory with the relative humidity of 50% for 48 hours (American Society for Testing and Materials, 1993). The setup tool is the same as the testing TS. Percent lengthening was calculated at the time that the film broke or was torn. Prior to withdrawal, the length of the film was measured up to the handle (i.e., the initial length [l₀]), and the length of the film after drawing was termed after breaking (l_i). The percent elongation was calculated by the formula in Eq. (3):

$$\text{Elongation (\%)} = \frac{\text{Long after breaking} - \text{initial length}}{\text{Initial length}} \times 100\% \quad (3)$$

2.7. Functional group analysis with FTIR

The IR spectra for the biodegradable film from seaweed waste were determined using a FTIR spectrophotometer 8201PC Shimadzu. The detector used was a Deuterated Tri-Glycine Sulfate (DTGS-KBR). The background spectrum using a clean crystal cell was recorded. Films were placed onto the crystal cell and the cell was clamped into position on the FTIR spectrometer. FTIR spectra were recorded in the range of 500–4000 cm^{-1} with automatic signal gain collected in 32 scans with a resolution of 4 cm^{-1} that were rationed against a background spectrum.

2.8. Biodegradability test

A biodegradability test was performed to determine when the plastic film sample starting to degrade. The biodegradability test was performed using soil in an auxiliary degradation process or using a soil technique called the soil burial test (Subowo and Pujiastuti, 2003). A 4 cm × 1 cm sample was planted in pots filled with soil, and the pots were left exposed to open air without covered glass. The sample was observed once a week until the sample degraded completely, or until the bioplastic sheet was no longer visible, or until the bioplastic sheet fused to the ground.

3. Results and discussion

3.1. Visual visibility of biodegradable film from waste seaweed

The results showed that biodegradable films from seaweed waste could be used to produce transparent plastic (Figure 1). CMC has a hydroxyl group that binds free water from the solution, thus causing the suspension to become more viscous. This will affect the thickness of the biodegradable films and reduce transparency (Nie et al., 2017), however, in this study, the concentration and thickness of the film still did not affect the transparency properties of the film.

3.2. Tensile strength (TS)

The use of glycerol affects the Tensile Strength (TS) of biodegradable films increased the glycerol concentration caused the TS to decrease, whereas increasing the concentration of CMC, increases the TS (Table 1). The results of the analysis of variance showed that the effect of the concentration of glycerol and CMC had a significant effect on the tensile strength of the biodegradable film, but the interaction effect of the concentration of glycerol and CMC was not significantly different at the 5% and 1% levels.

Table 1 shows that increased glycerol concentration can reduce TS. High glycerol concentration decreases mechanical resistance because the plasticizer decreases the intermolecular strength of the adjacent molecule (Sobral et al., 2001). The addition of glycerol resulted in a decrease in TS and Young's modulus and increased elongation at break (Gao et al., 2017). Glycerol is a hydrophilic compound that can increase the percent

elongation and causes biodegradable films to break easily when exposed to force (Sobral et al., 2001). Several studies have reported the same phenomenon where an increase in plasticizers can reduce the strength of the mechanical properties of film biodegradable products (Bourtoom, 2008). The observed TS decreased from 26.60 to 10.66 MPa, 14.31 to 2.52 MPa, and 16.14 to 8.28 MPa when sorbitol, glycerol, and polyethylene glycol increased from 20% w/v to 60% w/v (Bourtoom, 2008). The results showed that a higher CMC concentration can increase the TS of biodegradable films. This occurs because of the intermolecular interaction between the hydroxyl groups of starch and the CMC carboxyl group, thus strengthening the mechanical properties (Li et al., 2004).

The results were reliable with corn starch films (Ghanbarzadeh et al., 2011) and biodegradable cassava starch-based films (Tongdeesontorn et al., 2012). Polysaccharide blend films with CMC can increase mechanical and barrier properties because of the chemical similarity of polysaccharides, which allows for better compatibility (Hu et al., 2016). The results of a study that used agar extract from red seaweed, namely, Gracilaria salicornia, collected from the coastal area of Malaysia indicated that the TS and percent elongation of the photobleached agar film (3.067 MPa, 3.270%) were higher than those of alkali extracted agar and film (2.431 MPa, 2.476%) (Hii et al., 2016).

3.3. Thickness

The result of the variance analysis showed that glycerol and CMC concentrations affected the biodegradable film thickness produced, and there was an interaction between the two treatments. The biodegradable film thickness ranged from 0.107 mm to 0.170 mm (Table 1). The highest thickness value was 0.170 mm in a combination of 0.75% glycerol and 3% CMC concentration, whereas the lowest thickness was 0.107 mm in a combination of 0.25% glycerol and 1% CMC concentration.

CMC is a hydrophobic compound. Glycerol can bind water easier than CMC. Higher CMC concentrations resulted in greater biodegradable film thickness because CMC can bind the water of biodegradable film materials and increase the stability of the film (Balqis et al., 2017). Higher CMC concentrations cause thicker film structures (Ntharanathan et al., 2003).

3.4. Solubility

Biodegradable film solubility testing was conducted in this study because it is an essential factor in packaging materials. The result of variance analysis showed that glycerol and CMC concentrations affected the solubility of the biodegradable film, and there was an interaction between the two treatments. The solubility value ranged from 41.5% to 64.412%. Table 1 shows that increased glycerol concentration can cause an increase in solubility because plasticizers are hydrophilic and cause increased insolubility in water (Yadav et al., 2014). An increase in the plasticizer concentration increases the film's water content because of the hygroscopic high between the macromolecules of the sharing edges. The

Table 1. The use of glycerol and CMC at various concentrations on tensile strength, thickness, percent elongation and film solubility of seaweed waste film.

No	Glycerol	CMC	Tensile Strength (Mpa)	Thickness (mm)	Solubility (%)	Elongation (%)
1	0.25	1	101.341 ^b	0.107 ^c	60.428 ^b	2.500 ^g
		2	103.952 ^b	0.130 ^{ab}	41.946 ^c	5.000 ^{fg}
		3	123.023 ^a	0.150 ^{abc}	41.750 ^a	7.500 ^f
2	0.5	1	28.996 ^{de}	0.117 ^c	63.202 ^b	59.667 ^c
		2	40.336 ^c	0.143 ^{bc}	64.412 ^b	45.000 ^e
		3	40.368 ^c	0.163 ^a	61.650 ^b	42.500 ^e
3	0.75	1	23.083 ^e	0.123 ^{de}	63.423 ^b	72.333 ^b
		2	27.143 ^e	0.140 ^{cd}	60.425 ^b	55.833 ^d
		3	39.020 ^{cd}	0.170 ^{cd}	59.905 ^b	46.500 ^a

Note: the mean value followed by the same letter means not significantly different in the 5% LSD test.

addition of plasticizers can also increase film permeability. This approach can cause water to diffuse in the film easily. Generally, an increase in the concentration of CMC can decrease solubility. The decrease in solubility occurs because there was an interaction between the molecules of cellulose, glycerol, and CMC where the hydroxyl group and CMC carboxyl group can form strong hydrogen bonds (Ghanbarzadeh et al., 2011) and ester bonds (Li et al., 2008) with the hydroxyl groups in starch, respectively. This can cause an increase in the compactness of the biopolymer matrix and reduce water solubility (Balqis et al., 2017). The solid content increased after solvent evaporation/removing which also increase the thickness. The same effect was reported on CMC-biodegradable starch films (Bourtoom, 2008), corn-CMC films (Ghanbarzadeh et al., 2011), and corn starch-CMC nanoclay films. The biodegradable film production from *Sargassum siliquosum* uses a mixture containing 2 g of extracted alginate powder and 15% w/w of sorbitol followed by treatment with 75% w/w CaCl₂ resulted in water solubility of 33.73%. The solubility of edible films produced from carrageenan ranged from $60.51 \pm 8.92\%$ to $74.20 \pm 5.33\%$. Increased solubility was found in films plasticized with 25%–30% glycerol and 30% sorbitol compared with the semirefined kappa-carrageenan film (Lim et al., 1996).

3.5. Elongation

The results of the variance analysis showed that glycerol and CMC treatment affected the percentage of elongation in the biodegradable film produced, and there was an interaction between the two treatments. The percent value of elongation obtained from this study ranged from 2.5%–76.5%. The highest elongation percentage was 72.33% in the combination of 0.75% glycerol and 3% CMC, whereas the lowest was 2.5% in the combination of 0.25% glycerol and 1% CMC (Table 1).

Table 1 shows that glycerol can decrease intermolecular bonds and increase flexibility (Nourieddini and Mendikonkuru, 1997). The increase in percent elongation occurs because the plasticizer can reduce the fragility and increase the flexibility of polymer films by disrupting hydrogen bonds between adjacent polymer molecules, thus decreases the TS of the intermolecular in the polymer chain. Furthermore, plasticizers can increase the mobility of polymer chains, which contributes to more stretchable and flexible films (McHugh and Krochta, 1994). Polyols such as sorbitol and glycerol are good plasticizers for reducing the internal hydrogen bonds for increasing the intermolecular distance. Glycerol is a hydrophilic compound that can soften the material and result in an

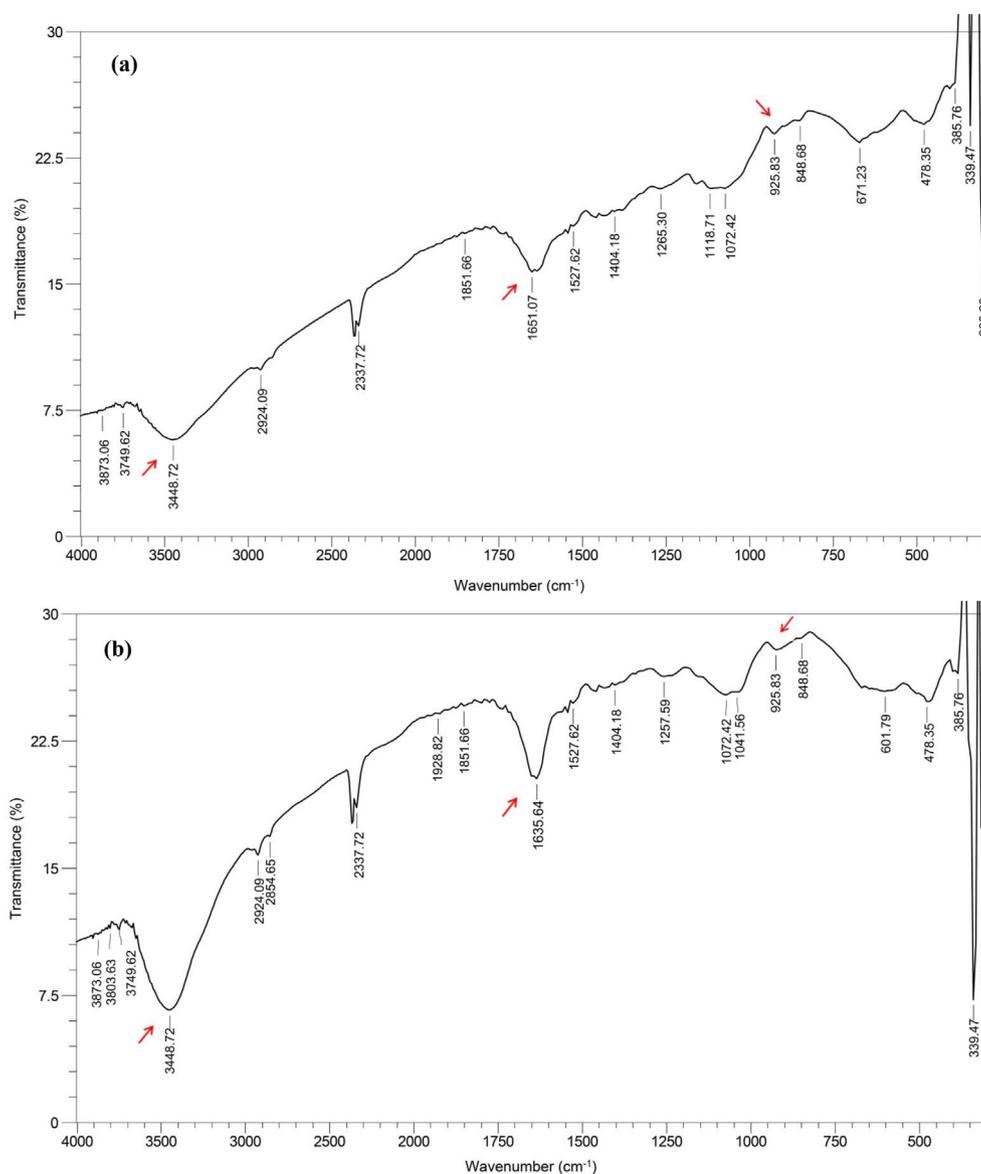


Figure 2. FTIR spectra of (a) Seaweed waste; (b) Biodegradable film.

increased percent extension. A similar effect has been reported (Chiu-marelli and Hubinger, 2014). The addition of cellulose derivatives is needed to improve the biofilm (Klangmuang and Sothornvit, 2016).

3.6. Functional group analysis with FTIR

Figure 2 shows the infrared spectra of the cellulose waste and biodegradable film of seaweed film. Tongdeesoontorn et al. (2012) reported that the hydroxyl group's bands are visible in the range $3100\text{--}3600\text{ cm}^{-1}$, and water absorption is higher for the biodegradable film than seaweed. The hydrogen bond formed by the interaction of the O-H groups at the end of the polymer chain from starch and in the

plasticator is shown in the broad absorption band at $3600\text{--}3020\text{ cm}^{-1}$. The aliphatic absorption peak C-H occurs in the absorption band of about 2950 cm^{-1} , while the bonded water molecule in starch is found at the peak of low intensity 1680 cm^{-1} .

Increased peaks in hydroxyl areas occur in biodegradable films compared to waste seaweed pulp material due to the higher solubility of biodegradable film products. A strong band at 930.3 cm^{-1} indicates the presence of 3,6-anhydro-galactose residue which is the main structure of seaweed which has the ability to form a gel (Pereira et al., 2013). The peaks between $422\text{--}600\text{ cm}^{-1}$ are dyes such as chlorophyll and carotene originally present in native seaweed and then disappear in biodegradable film products (Indriatmoko et al., 2015). The peaks in the amorphous



Figure 3. (a). Biodegradable film planted in pots filled with soil, the film sheet is clearly visible; (b). Observation of the biodegradable film test results in the first week still send the film sheets that have not been degraded; (c). Observation of the biodegradable film test results in second week, no film sheets were visible.

region show at wave number 1648 cm^{-1} . The product of biodegradable film has cellulose which is more amorphous compared to the initial raw material, namely seaweed so that a sharper peak can be seen at the region of 1648 (Abdul Khalil et al., 2018; Kizil et al., 2002). The functional groups of starch and glycerol from biodegradable seaweed waste occur in the absorption bands at 920 and 1022 cm^{-1} (Kizil et al., 2002).

3.7. Biodegradability

Biodegradability analysis showed that biodegradable films buried in soil can be completely degraded after two weeks of burial (Figure 3c). Biodegradable film having $-\text{COO}-\text{OH}$, and $-\text{COOH}$ functional groups as flexible active sites can undergo degradation with high rates since these active groups enabling the film to bind on enzyme sites faster than the case with rigid biodegradable films. In addition, polymers with shorter chain are known to degrade faster compared to the polymers with complex chemical structures which require additional enzymes or co-enzymes (Narancic and O'Connor, 2019). The biodegradable film produced was easily decomposed because the raw material used was a raw material that easily interacts with water and microorganisms and was sensitive to physicochemical influence. After biodegradable film burial in the first week, the observations showed that the biodegradable film decomposed but not completely, and remnants are still present in the soil. The results showed that biodegradable film was degraded because of the damage caused by the degradation process by fungi and bacterial decomposers.

Biodegradable films have properties that are readily biodegradable or degraded by soil compared with synthetic plastics. Biodegradable films can be biodegraded by reducing the polymer chain length by oxidation, which may be accessed by microbes. Biodegradable films made from the cellulose waste of seaweed produced in the study turned out to be easily degraded in soil both biologically and chemically.

4. Conclusions

There was an interaction between the two treatments of glycerol and CMC concentrations on the biodegradable film produced on the pellets of the thickness test, elongation percentage, solubility, and biodegradable biodegradability test of the film, except the tensile strength. The results showed that the addition of glycerol concentration of 0.5 and 0.75% with the addition of CMC from 1 to 3% produced a tensile strength of 23–39 MPa. This value is proportional to the tensile strength of Poly Tetra Fluoro ethyne (PTFE) and Poly Propylene (PP) synthetic plastics released by Dotmar Engineering Plastics. Biodegradability test shows that the produced biodegradable films decompose after 14 days.

Declarations

Author contribution statement

Sri Hidayati: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Zulferiyenni: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Ulfa Maulidia: Performed the experiments.

Wisnu Satyajaya: Performed the experiments; Contributed reagents, materials, analysis tools or data.

Sutopo Hadi: Contributed reagents, materials, analysis tools or data; Wrote the paper.

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Data availability statement

Data included in article/supplementary material/referenced in article.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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