Chapter in Book

Development of biodegradable film based on methycellulose incorporated with lemongrass extract

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Abstract: *Edible films and coating have proven their ability to preserve the food from spoilage during handling, transport, and storage. However, vast amount of carbon dioxide produced annually from fossil polymers drew a great attention from the world, and this inspires researchers to find the alternatives for this due to shortage of conventional polymers. Biopolymer film become an option as food packaging film due to their gas barrier property, production cost, safety, durability, and biodegradability. Lemongrass extract (LGE) is one of the plant extracts that has been recognised by Food and Drug Administration (FDA) to be used as additives or flavouring. The present research demonstrates that incorporation of lemongrass extract into methylcellulose is a good alternative for fossil films. To the best of our knowledge, this is the first time that lemongrass extract is added to methylcellulose film to strengthen its capabilities to preserve food. In this study, methylcellulose (MC) incorporated with lemongrass extract (MC/LGE) was prepared and the physicochemical properties of methylcellulose as a biofilm material has been studied through Fourier transform infrared (FTIR) spectroscopy, scanning electron microscopy (SEM), thermogravimetric analysis (TGA), and light transmission and transparency. There are two studies in application of MC/LCE films, namely preservation study and the biodegradable study. The absorption bands at 3329, 2912 and 1645 cm-1 showed the stretching of C-OH, C-H and C-O bonds of MC/LGE, respectively. The unstable molecular arrangement between MC and LGE has been proven by SEM results by exhibiting slightly rough surface with cracks on the film. TGA results demonstrated that MC/LGE films are thermally stable. Transparency value of MC/LGE is higher than the commercial cling film and the food could be preserved for longer period. The shelf life of cherry tomatoes has been extended after wrapped with MC/LGE films.*

Keywords: methylcellulose; lemongrass extract; biodegradable.

1. INTRODUCTION

Biopolymers are naturally derived from several organic sources like starch and plant proteins. When they are exposed to heat and moisture, they could be degraded by microorganisms (Zhang et al., 2019; Vedove et al., 2021). The capability of bio-based plastics and biopolymers in replacing oil-based packaging materials draw great attention because they provide advantages such as more sustainable and greener features and improved technical properties. The urge of incorporating biopolymers as an alternative to synthetic packaging materials is because according to the Organization for Economic Cooperation and Development (OECD) report which was published in June 2022, the global production amount of plastic waste is estimated to three-folds by 2060. Half of the production will be in landfills and less than a fifth of production will be recycled.

Besides, OECD (2022) has reported that 63 percent out of million tonnes of solid waste is derived from plastic packaging materials globally which is predicted to produce 56 gigatonnes of carbon dioxide gas until the year of 2050. The statement on recycled plastics is supported by another global statistics from OECD which stated that only 9 percent of plastic waste is recycled and 22 percent is mismanaged for the year of 2019 (Halonen et al., 2020). These statistics proves that campaigns, practices and missions on recycling to reduce plastic waste has been proved to be a failure. The increasing environmental impacts of fossil polymers (plastics) drive the urge to produce sustainable packaging materials. In recent years, biopolymers are being exploited as alternatives in food industry. Biopolymers or polymeric biomolecules are mainly synthesised from renewable materials such as starch, proteins, natural fat, oil, and sugar (Rodriguez-Turienzo et al., 2012). These materials are biocompatible, biodegradable, non-toxic, chemically inert, and cheap, depending on the composition (Mohamed et al., 2020). Furthermore, conversion of natural-based polymer waste into usable biodegradable materials is more economical (Dhall & Alam, 2020).

Lemongrass (Cymbopogon citratus) is a common spice plant in tropical countries, especially in Southeast Asia countries. The main phenolic compounds identified in C. citratus are terpenes, alcohols, ketones, aldehyde, and esters. Its leaves are mainly used as antioxidants in the fields of health and food due to the phenolic compound concentration found in the leaves (Tan et al., 2015). Antioxidants inhibit the initiation or propagation of oxidation and stop the onset of lipid oxidation effectively in food products. Lipid oxidation is a free-radical chain reaction which contributes to reduction in sensory properties and nutritional value of food (Zhang et al., 2017). Besides that, several studies proven that it possesses various pharmacological activities such as antibacterial, anti-diarrheal, anti-filarial, antifungal, and anti-inflammatory properties (Siripatrawan & Harte, 2010).

According to several studies, chitosan, gelatin and methylcellulose are mainly used biopolymers incorporated with additives to overcome the soul property of biopolymers. In this study, the incorporation of lemongrass extract into the methylcellulose biopolymer matrices is studied to enhance its capabilities to replace synthetic packaging materials, polyethylene (PE) cling film.

2. METHOD & MATERIAL

2.1 Chemicals and materials

Methylcellulose (MC) powder (viscosity: 4000 cP) was purchased from Sigma-Aldrich (USA) and lemongrass extract (LGE) was purchased from Bio-Nutricia (Malaysia). Glycerol (85%) was purchased from Merck (USA). All the chemicals were analytical grade.

2.2 Preparation of film forming dispersion

To prepare film forming dispersion (FFD), MC powder (2.0 g) was dissolved in 100mL of deionised water (80.0 °C). The solution was stirred on a magnetic stirrer continuously for two hours at 25.0 ± 2 °C. A volume of 1.0 mL of glycerol was added to the FFD and then stirred for another hour. LGE at 25% (w/w) was added to the solution and stirred at room temperature for one hour to achieve the homogeneous condition. The solution was kept refrigerated (4 °C) for 24 hours for hydration and dissolution purposes.

2.3 Preparation of films

MC/LGE biopolymer films were prepared through casting method and the method is suitable for lab-scale films production. Briefly, MC FFD (10.0 mL) was poured into a 90mm-diameter clear polystyrene petri dish. The solution was dried in an oven at 40 °C for 24 hours. The thickness of the films was measured using a digital calliper (Insize SL-1108-200). The films were sealed in a vacuum packaging for protection from humidity.

2.4 Fourier transform infrared (FTIR) spectroscopy

Functional groups of the MC/LGE film were studied using a Perkin Elmer Spectrum 3 FTIR Spectrophotometer. The film was cut into the size of 1 cm x 5 cm and placed onto a metallic slit sample holder. The film was scanned under 16 cumulative scans and bandwidth of 2 cm^{-1} in the range of 400 to 4000 cm^{-1}.

2.5 Scanning electron microscopy (SEM)

A Jeol JSM IT200 Scanning Electron Microscope was used to observe the surface morphology of MC/LGE film. Prior to analysis, a layer of gold coating was sputtered on the film to avoid charging. In this study, the magnification and acceleration voltage were set at 1000× and 3 kV, respectively.

2.6 Thermogravimetric analysis (TGA)

The thermal stability and decomposition stages of MC/LGE film was investigated by using a TGA/DSC 1 Mettler-Toledo. The film was heated at 10 °C per minute in argon atmosphere and the temperature ranged from 30 to 1000°C.

2.7 Light transmission and transparency

The light transmission of UV and visible light and transparency for MC/LGE film were determined using a Jasco V-570 UV-Visible Spectrophotometer. In this study, both properties of MC/LGE film were compared with commercial polyethylene (PE) cling film. The wavelength was set from 200 nm to 800 nm and the measurement was recorded three times. The transparency of the film was calculated using Equation (1):

$$\text{Transparency} = \frac{\text{Aborbance at wavelength 600 nm}}{\text{film thickness (mm)}} \quad (1)$$

2.8 Preservation study

Fresh cherry tomatoes (*Solanum lycopersicum* var. *cerasiforme*) were purchased from a local market in Tanjong Malim, Perak, Malaysia. The tomatoes were washed with deionised water and dried with a clean towel before one of them was wrapped using MC/LGE film. The fruits were kept at a temperature of 25 °C and 75% relative humidity for 10 days. At the end of preservation study, the appearance was observed.

2.9 Biodegradation study

Degradation study of film was studied in soil environment. Film sample of size (5 cm x 5 cm) was placed in a pot (15 cm in diameter and height of 17 cm) containing approximately 700 g of soil which was separated in two parts. The film was placed in the middle of two equal portions of soil. The pots were kept in observation for 6 months and observed monthly. The biodegradation of commercial PE cling film, representing synthetic packaging material was studied as control.

3. FINDINGS

The findings of analytical studies carried out to prove that the biocomposite film of methylcellulose incorporated with lemongrass extract is one of the promising alternatives for synthetic packaging is further discussed in the following sections.

3.1 FTIR

Figure 1 and Table 1 show the FTIR spectra and the assignment of absorption bands at various wavenumbers for MC/LGE film respectively.

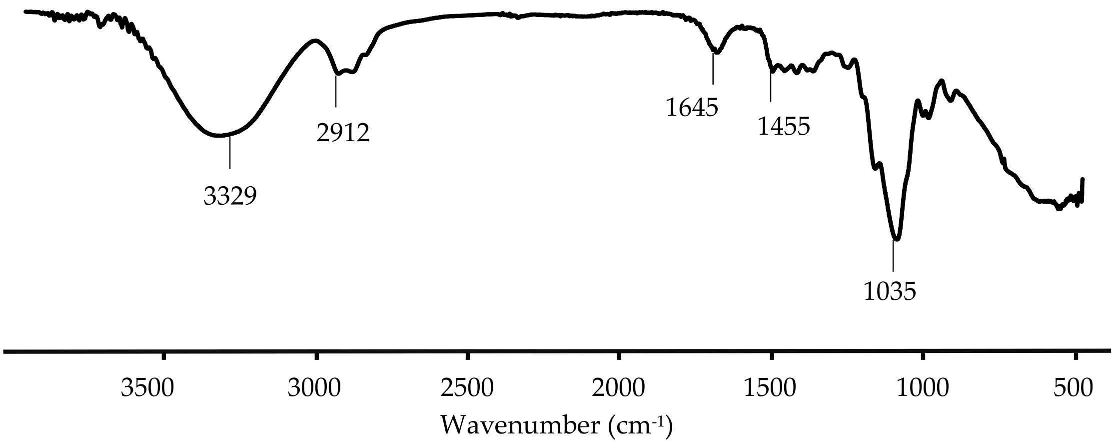


Figure 1. FTIR spectrum of methylcellulose/lemongrass extract film.

Table 1. Wavenumber and peak assignments for FTIR spectrum

Wavenumber (cm^{-1})	Assignment
3329	O–H stretching
2912	C–H stretching
1645	C–O stretching
1455	C–OH stretching
1035	- OH stretching of glycerol

3.2 SEM

Low flexibility of MC has caused the difficulty in peeling off from casting surface. The surface morphology of MC/LGE at 10,000x magnification is depicted in Figure 2. A rough surface with cracks is observed on the film.

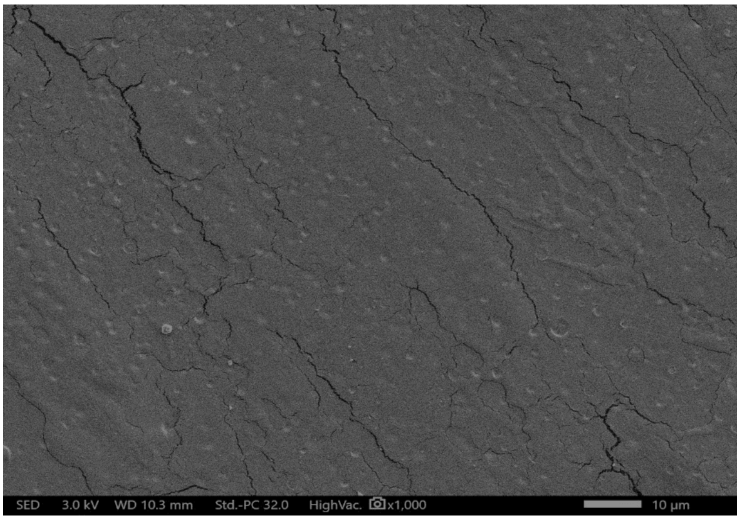


Figure 2. SEM image of MC/LGE film at 1,000× magnification.

3.3 TGA

TGA thermogram is presented in Figure 3. MC is capable to retain water inside the molecule. Hence, the first weight loss was observed approximately between 70 °C and 180 °C due to moisture loss or water evaporation. A second weight loss has been observed in the temperature range 340 °C between 383 °C and this can be ascribed to structural degradation of MC and LGE (Khan et al., 2018).

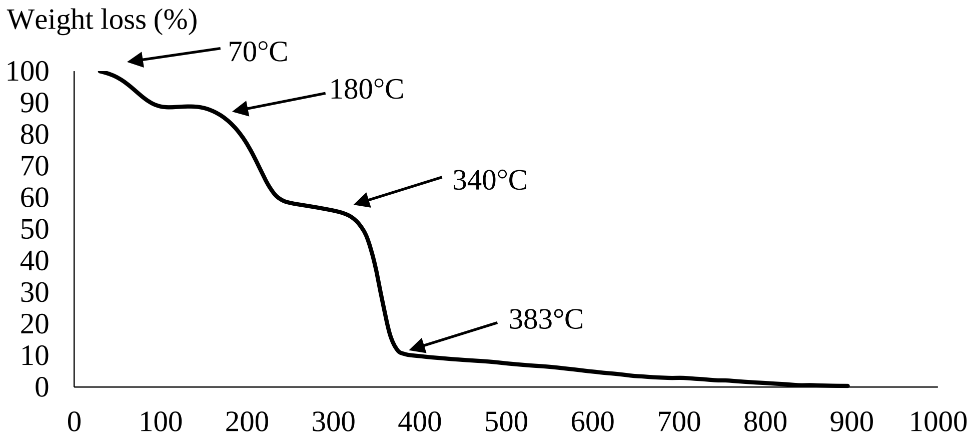


Figure 3. TGA thermogram of MC/LGE film.

3.4 Light transmission and transparency

Film formed of MC/LGE is transparent. Light transmission and transparency value of MC/LGE film is evaluated and presented in Table 2.

Table 2. Light transmission and transparency value of MC/LGE and commercial cling film.

Film	**Thickness (mm)**	**Light transmission (%T)**							**Transparency value (A/mm)**
		200	**300**	**400**	**500**	**600**	**700**	**800**	
MC/LGE	0.03	4.4	36.8	28.3	58.1	62.3	64.9	66.9	5.142
PE cling film	0.03	7.8	82.7	81.2	91.5	91.8	92.5	92.8	1.239

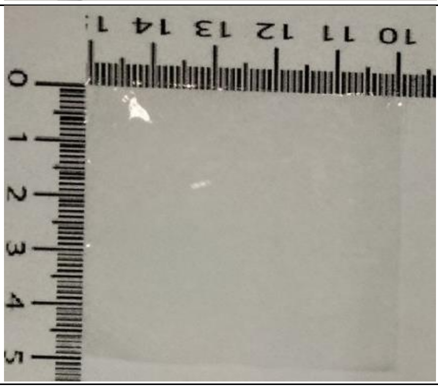
3.5 Preservation study

Table 3. Appearance and freshness of cherry tomatoes for unwrapped and wrapped with MC/LGE film for 10 days.

Title 1	Day 0	Day 10
Unwrapped		
Wrapped (MC/LGE)		

3.6 Biodegradation study

Table 4. MC/LGE film and commercial PE cling film degradation in 1 month.

Film	0 month	1 month
MC/LGE		
PE cling film		

4. DISCUSSION

The functional groups of compounds in MC/LCE were studied by FTIR. In Figure 1, FTIR spectra of MC/LGE shows certain assignments at various peaks observed as shown in Table 1. The broad peaks at 3329 cm^{-1} and 2912 cm^{-1} could be related to the presence of O-H stretching of carboxylic/phenol group and C-H stretching, respectively (Ahmed & Ikram, 2015). The peak observed at 1645 cm^{-1} may be due to the presence of aromatic C–O stretching and the peak at 1035 cm^{-1} could be due to C–OH stretching (Lagos, 2013).

In Figure 2, MC/LGE film exhibited a rough surface with few cracks indicating a non-homogeneous surface due to the presence of some large lemongrass extract molecules being less able to penetrate through the methylcellulose structure (Mohammad & Jafar, 2021). The irregular surface of film at certain spots may be due to unstable molecular rearrangement caused by crack propagation within the polymer matrix, leading to the fracture of film (Ana et al., 2015). Other than that, the volatility of extract and the effect of extract in water contributed the cracks on the films (Sajed et al., 2020). According to Khan et al. (2018), smooth surface of film and no aggregation could be arisen from the homogeneity on the surface as this indicates the well dispersion of the extract with biopolymer and the display of a miscible polymer blend.

TGA thermogram of MC/LGE film is shown in Figure 3. MC/LGE film exhibited three decomposition stages. From 50 °C and 100 °C. the first decomposition stage was observed, and the mass loss was 11.2% due to evaporation of water and volatile materials. The temperature range for second and third decomposition were between 150 °C to 250 °C and 310 °C to 350 °C respectively. The percentage of weight loss second and third decomposition were 42% and 92.5% respectively. The weight loss at second decomposition stage could be caused by decomposition of biopolymer materials (Tongnuanchan et al., 2012). The highest percentage of decomposition is at the third stage which could be due to high bond stability between functional groups of glycerol, methylcellulose biopolymer and lemongrass extract, requiring a higher temperature to break the bond (Zhang et al., 2019).

Light transmission and transparency values of MC/LGE and PE cling film are shown in Table 2. The transparency value of commercial PE cling film was 1.239 A/mm which is lower than MC/LGE film at 5.142 A/mm. It is known that film with higher transparency value indicate higher opaqueness (Tongnuanchan et al., 2012). As the transparency values increase, the opaqueness of films increases as well, where the increasing property of opacity positively relates to the improvement in light barrier properties (Wang et al., 2012). According to Kowalczyk and Biendl (2016), biopolymer films should be able to block UV light to avoid quality defects of food sample as transparent films allow light to have contact with food sample which can induce oxidation and degradation of nutritional compounds in food (Silva-Weiss et al., 2015). Hence, transparency of film is considered significant to food appearance and consumer demand (Hosseini et al., 2015).

The appearance and freshness of cherry tomatoes have been studied by wrapping them with the prepared biofilm. The unwrapped cherry tomato shows the most observable damage in terms of freshness and appearance compared to the MC/LGE wrapped cherry tomato. The effectiveness of MC/LGE biofilm to preserve cherry tomato was studied at a surrounding temperature of 20 to 25 °C for 10 days. As shown in Table 3, the MC/LGE wrapped cherry tomato could maintain a good level of appearance and freshness compared to the unwrapped sample because the biofilm could prevent the diffusion of oxygen and light to enter the food sample and therefore increase the shelf life (Atares & Chiralt, 2016).

The biodegradation study of MC/LGE biofilm has added positive feedback to implement biopolymer-additive films as a replacement to synthetic packaging materials. This could be seen in Table 4 which shows the difference in size of MC/LGE and PE commercial cling film before and after being buried in soil for degradation purposes. There was no observable size reduction in commercial PE cling film compared to MC/LGE biofilm in a month. The MC/LGE film has completely degrade in soil environment within a month. This result suggest that the prepared biopolymer film possess great potential as an alternative to petroleum-based packaging material.

5. CONCLUSION

In this study, MC/LGE films has been successfully prepared and the films exhibited outstanding physicochemical properties such as high thermal stability and high opacity in order to preserve cherry tomatoes. The preservation ability of MC/LGE films is proven by preserving cherry tomatoes for a long period at room temperature. Moreover, the MC/LGE films showed the high biodegradation properties, and this could reduce the pollution to the environment which aligns with the sustainability goal. This study is parallel to achieving UNSD goals, which is to create good health and well-being, supports industry, innovation and infrastructure, moving towards sustainable cities and communities and creating awareness towards responsible consumption and production. While keeping food fresh, other biodegradable materials should be developed more in future to be options for food wrapping films.

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