Synthesis of Methyl Cellulose from Nang Lae Pineapple Leaves and Production of Methyl Cellulose Film

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Abstract

After harvesting Nang Lae pineapple fruits, their leaves remain as agricultural waste. The Nang Lae pineapple leaves contained moisture up to 82.91±0.22% and 32.21±0.67% (w/w) cellulose. Methyl cellulose, a well-known commercial product which is widely used in agriculture, food, and pharmaceutical industries, from Nang Lae pineapple leaves (MCpl) was synthesized with various NaOH concentrations (30%, 40%, and 50%) and their properties i.e., the degree of substitution (DS), color, viscosity, water solubility, and functional groups (FTIR) were determined. The use of 40% NaOH in the methylation of cellulose gave the highest yield (136.31±26.27%) and the highest DS (1.36±0.03). However, this DS value of MCpl was lower than the DS value (1.61±0.02) of commercial MC (MCc). This synthesized MCpl using 40% NaOH was found to have better properties than the MCpl from others such as the higher lightness (L* 75.17±0.28), higher viscosity (20.40±0.25cP), higher water solubility (86.00±2.00%), and showed the similar FTIR patterns as the MCc. Thus, the synthesized MCpl using 40% NaOH was selected to be applied in the film formation. The film produced from MCpl (2% w/v) showed higher tensile strength (58.30±10.78MPa) and water solubility but lower elongation (19.63±8.15%) than MCc film. There was no significant difference between water vapor transmission rate (WVTR) of MCpl and MCc films. Hence, MCpl film can be a good alternative for using as an edible film.

Keywords: Nang Lae pineapple leaves, methyl cellulose, synthesis, edible film

1. Introduction

Methyl cellulose is a chemical compound derived from cellulose within plant or natural waste which is well known in acommercial product.It can be used as additives in adhesives or thickening agents, viscosity control agents, or protection in paint formulations. For the pharmaceutical grades, MC have been used as thickeners, binders, emulsifiers, and stabilizers in a variety of cosmetic and food products as well as it is also used as protective coatings, toiletries, as amatrix to control the release of drugs in thepharmaceutical industry [1]. Besides, cellulose- based edible films, such as methylcellulose has excellent film making characteristics with high solubility, low oxygen and lipid permeability and barriers against aroma compounds [2].

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Pineapple (*Ananas comosus*, cv. 'Nang Lae') is one of the most popular tropical fruits in Chiang Rai province, Thailand. Nang Lae pineapple is widely eatenasthe fresh cut and processed productssuch as juice concentrates, jams, and jellies. Currently, the main focus of the pineapple industry is the fruit hence, the other plant parts are considered as waste. Whilst, the highest rest (70%) is waste residue that consistss of the leaves [3] which mostly this waste is usually used as animal feed in the agriculture sector, on the other hand, this waste may be converted into more useful form that will help to eliminate variety of socio-economic problems. It can be applied to obtain a new raw material for packagingor might be biologically treated to produce food or be converted into higher value added products [4]. Nanglae pineapple leaves consist of cellulose content up to 69.17±3.24% (w/w) (from a preliminary study). Therefore, the new methylcellulose was produced from Nanglae pineapple leaves, and the properties of the produced MC such as the degree of substitution, functional group, viscosity, water solubility, and color were investigated. For application as edible film, the properties, such as mechanical properties, water solubility, water vapor transmission rate and transparency value of film from Nang Lae pineapple leaves also were examined.

2. Materials and Methods

2.1 Sample preparation

Pineapple (cv. 'Nang Lae') leaves were harvested from the local farmer in Chiang Rai province, Thailand. The leaves were cut into small pieces and weighed. Then, they were washed with running water to remove dirt and other foreign materials. The leaves were dried using a tray dryer at 80°C for 2 days. The dried pineapple leaves were turned into powder using a hammer mill and the sample was passed through a500µm sieve. The sample was kept in zipper polypropylene bags.

2.2 Extraction of cellulose

Cellulose was extracted from the pineapple leaves powder according to the method used by Rachtnapun *et al*. [5]. The pineapple leaves were soaked into 30% (w/v) NaOH at the ratio of 1:10 (w/v) with the use of autoclave at 121°C for 1 hour. The black slurry that was obtained from the extraction was filtered and washed with deionized water until the washed water becomes clear. The slurry was then bleached based on the method used by Jahan *et al*. [6]. The slurry was suspended in 20% H_2O_2 at pH 11 for 90 min and the reaction was carried out at 60°C. The pH of the slurry was adjusted to pH 7 by using 2NHCl. The slurry was filtered and the residue was dried in hot air oven at 80°C overnight. The cellulose obtained was kept in zipper polyethylene bags until it used in the MC synthesis. The % yield of cellulose was calculated by using the equation (1):

$$\text{Yield of cellulose (\%)} = \frac{\text{Weight of dried cellulose (g) x 100}}{\text{Weight of dried pineapple leaf powder (g)}} \qquad (1)$$

2.3 Synthesis of methyl cellulose (MC)

The synthesis of methyl cellulose from cellulose was based on the method used by Biswas *et al.* [7]. The synthesis was investigated by using 3 different concentrations of sodium hydroxide (NaOH) (30%, 40%, and 50%). One gram of the cellulose and 20 ml of NaOH was mixed and stirred for 1 hr. at 25°C. The sample was filtered and pressed between wipes to remove excess liquid. It was transferred to a round bottom flask containing 9 ml of acetone and stirred. Then 4 g DMS was added slowly at 25°C. The reaction mixture was stirred and heated to 50°C for 1 hr. and cooleddown to room temperature, and the sample was filtered. Acetic acid (10%) was added to neutralize the pH. The product was filtered and washed with acetone several times and dried in the hot air oven at 80°C overnight.

$$\text{Yield of MC (\%)} = \frac{\text{Weight of dried MC (g) x 100}}{\text{Weight of dried bleached cellulose (g)}} \quad (2)$$

2.4 Determination of MC properties of MC from pineapple leaf (MCpl) and commercial MC (MCc)

2.4.1 Determination of degree of substitution (DS)

Sample (0.5g) was placed into 250 ml conical flask. 50 ml distilled water was added and the pH was adjusted to 7 by using 0.02 M of HCl. 0.5 M of NaOH (25ml) was then introduced and the mixture was heated on ahot plate until a transparent solution was obtained. The excess NaOH was titrated with 0.2M of HCl in the presence of phenolphthalein as anindicator to change the pH back to 7 [8]. DS value for each sample was calculated by using the formula:

$$DS = \frac{52.5\ (M_{NaOH}V_{NaOH}) - (M_{HCl}V_{HCl})}{1000 - 42\ (M_{NaOH}V_{NaOH} - M_{HCl}V_{HCl})} \quad (3)$$

2.4.2 Determination of functional group

The functional group of MC was examined by using Fourier Transform Infra-Red (FT-IR) spectroscopy. The spectra were taken at are solution of 4 cm^{-1} with a total of 16 scans of each sample in the range of 4000-400 cm^{-1} in the transmission mode. Pellets were made from MC sample (2 mg) with KBr salt per 100 grams of dry sample [9].

2.4.3 Viscosity measurement

Two grams of MC was dissolved in 200 ml of distilled water (1% w/v) and the mixture was stirred at 25°C until dilute. The viscosity of the sample solution was then measured using a Viscometer Model Brookfield Rotational with spindle no.61 and speed of 200 rpm [10].

2.4.4 Water solubility

MC samples were weighed to the nearest 0.5 g. It was mixed with 50 ml distilled water in Erlenmeyer flask and the mixture was shaken for 24 hr. at 30°C for 160 rpm. Then, the mixture was filtered by using avacuum pump and dried filter paper. The gel collected on the filter paper was dried in hot air oven at 105°C overnight. The water solubility (%) was determined in terms of percentage of the dried gel with respect to the amount of the initial weight of the MC powder (~0.5 g) [11].

2.4.5 Color measurement

The color of MC was measured by using Colorimeter Hunter Lab (Miniscan EZ 4500L, Virgian, USA) which was calibrated by white and black tile. The result was expressed as L* value which represents lightness, a* value which represent redness (+) and greenness (-) and b* value which represents yellowness (+) and blueness (-). MC powder was placed on a glass plate and it was shot by colorimeter for four times. The average value of L*, a*, and b* was calculated. The L* value was used to determine the lightness of the MC samples.

2.5 Preparation of MC films

The film properties of the best condition of MC from pineapple leaves (MCpl) and commercial (MCc) were prepared at 2% concentration that was 2 g/100 ml solvent (solvent = 50% ethanol). Glycerol (30% w/w) was added into MC solution and stirred using magnetic stirrer bar at 40°C until the solution became homogeneous. Then, the film solution was cast into the Teflon plate and dried at room temperature (25°C) for 24 hr. MC film was stored at 25°C, 50%RH before measuring the properties.

2.6 Determination of properties of MCpl films and commercial MCc films

2.6.1 Mechanical properties

Tensile strength (TS) and elongation at break (EB) of the MC films were determined by using the ASTM Standard Method D 882-91 [12]. The film samples (20 specimens) were cut into 15x100 mm for testing. The films were equilibrated for two days at 50% RH in a climatic chamber at 25°C before the test. The initial grip separation and across-head speed of the Universal Testing Machine (UTM Instron, USA) were set at 50 mm and 50 mm/min, respectively.

$$\text{Tensile strength (MPa)} = \frac{\text{Force (N)}}{\text{Width film (m) x Thickness of film (m)}} \qquad (4)$$

$$\text{\% Elongation} = \frac{\text{Extension (mm) x 100}}{\text{Length of film (mm)}} \qquad (5)$$

2.6.2 Water solubility

The solubility of films in water was measured following the method of Dick *et al*. [13]. The films were cut into small pieces (near weight, dried 60°C for 1 day and then recorded initial weight. It was mixed with 50 ml distilled water in Erlenmeyer flask and the mixture was shaken at 30°C, 160 rpm for 24 hr. Then, the mixture was filtered by using avacuum pump and dried filter paper. The swollen film on the filter paper was dried in hot air oven at 105°C overnight and weighed. The water solubility (%) was determined in terms of percentage follow the formula as:

$$\text{Water solubility (\%)} = \left[\frac{\text{Initial dry weight} - \text{Final dry weight}}{\text{Initial dry weight}}\right] \times 100 \qquad (6)$$

2.6.3 Water vapor transmission rate (WVTR)

The water vapor transmission rate (WVTR) was investigated according to Rachtnapun *et al.* [5]. The contained cups with the dried silica gel (10 g) were covered with the film samples those were cut into circles with the diameter of 70 mm and sealed with paraffin wax. The sealed cups were weighed and stored at 25°C 75% RH in a desiccator. The cups were reweighed daily for 5days. The slope of the linear relationship between time as an independent variable and weight gain as a dependent variable was obtained from the data and it was used to calculate the WVTR based on the equation:

$$\text{WVTR} = \frac{\text{slope}}{\text{film area}} \qquad (7)$$

2.6.4 Transparency value

Transparency value was referred from the method of Han and Floros [14]. The ultraviolet and visible light barrier properties of the films were measured using a UV-Vis Spectrophotometer at 600 nm. The transparency value of the films was calculated by using the equation:

$$\text{Transparency (\%)} = (\log \text{\%T})/x \qquad (8)$$

Where %T is the transmittance at 600 nm and x is the film thickness in mm.

3. Results and Discussion

The Nang Lae pineapple leaves that were used in this study contained 82.91±0.22% of moisture. The pineapple leaves sample were pre-treated with hydrogen peroxide. During the pre-treatment process, most of the hemicellulose was solubilized, degraded to monomeric sugars (such as xylose, arabinose, and galactose) and washed away [7]. The amount of cellulose obtained from the purified sample was 32.21±0.67% (w/w).However, this Nang Lae pineapple leaves showed higheryield of cellulose than the otheragricultural wastes such as wheat straw (32.00%) [7], rice straw (32.1%) [15] and banana wastes (13.2%) [16].

In addition, the pre-treated cellulose (bleached) from pineapple leaves was converted into methyl cellulose (MCpl) where the different concentrations of NaOH was used for the alkaline treatment. The experiment that was carried out by using 30% NaOH (methylcellulose A), 40% NaOH (methylcellulose B) and 50% NaOH (methylcellulose C) are summarized in Table 1. This methylation reaction involved the removal of thehydroxyl group from cellulose and the substitution of methoxy group as seen in the reaction equation:

$$\text{Cell-OH+NaOH} \rightarrow \text{Cell-ONa+}H_2O,$$
$$\text{Cell-ONa+}CH_3O\ SO_2\ OCH_3 \rightarrow \text{Cell-}(OCH_3)_3 + Na_2SO_4$$

The molecular weight of methoxy group is higher than ahydroxyl group. Thus, the yield (% dried weight) was found to be more than 100% which percent yield of the MC synthesized showed between 126 to 138 g per 100 g of cellulose.

Table 1. Methylation reaction for pineapple leaves.

Methylcellulose sample	Concentration of NaOH (%)	Yield (% dried weight)
A	30	132.61±28.67[a]
B	40	136.31±26.27[a]
C	50	126.41±11.98[a]

Each value is expressed as mean±SD. Means in arow followed by different letters are significantly different (*P*<0.05), based on Duncan's test.

Note. Methylcellulose A, B, and C showed the extraction by using 30% NaOH, 40% NaOH and 50% NaOH, respectively

3.1 Properties of MC from pineapple leaf (MCpl) and commercial MC (MCc)

3.1.1 Degree of substitution (DS)

Methylation was performed at 50°C for 3 hr. with the use of acetone as solvent and DMS as amethylating agent [17]. For the methylation reaction to occur, the cellulose must be in thealkaline state thus, the cellulose was first undergone alkali treatment using NaOH. Acetone was selected as the solvent as it produced methyl cellulose with higher DS value [7,17,18] which may be due to the effective interaction of this solvent with the hydroxyl group of cellulose, decreasing the amount of inter-chain hydrogen bonds if compared to other solvent such as toluene. This interaction is predominant on the original cellulose and on low DS methylcellulose at the initial steps of the synthesis thus, the use of acetone since the start of the synthesis is favorable to increase the DS of the methyl cellulose [19].

The effect of the use of different NaOH concentrations(30-50%) at the beginning of the reaction on DS was investigated and the results are as seen in Table 2. It was observed that increasing the concentration of NaOH solution from 30% to 40% caused the DS to increased from 1.30 to 1.36. This may be due to better and more uniform accessibility of the cellulose chains within the fiber at such a higher concentration. However, the increase further in the concentration of alkali up to 50% caused a decrease in DS from 1.36 to 1.33 which may be due to the depolymerization of the cellulose [20]. These DS values of methyl cellulose from pineapple leaf is higher compared to methyl cellulose from sugarcane (1.20) [21]. As methylcellulose B showed higher DS value than methyl cellulose A and C which this indicated that methylcellulose B had the highest level of substitution. On the other hand, the DS value for commercial MC was 1.61 ±0.02 which was significantlyhigher thanthose of MC_{pl}. However, these DS values (for MCcand MCpl) were in agreement with the range of average DS values (1.3 to 2.6) found from the literature [22].

Table 2. Degree of substitution (DS) values of methylcellulose.

Methylcellulose sample	DS
Commercial	1.61 ± 0.02^{a}
A	1.30 ± 0.02^{c}
B	1.36 ± 0.03^{b}
C	1.33 ± 0.01^{b}

Each value is expressed as mean±S.D. Means in arow followed by different letters (a to c) are significantly different ($P<0.05$), based on Duncan's test.
Note Methyl cellulose A, B, and C showed the extraction by using 30% NaOH, 40% NaOH and 50% NaOH, respectively.

3.1.2 Functional group

The Functional group of methylcellulose synthesized was represented in Figure 1 and from the absorption bands of commercial methylcellulose as stated in Table 3.The absorption of 3450 cm^{-1} is due to stretching of O-H groups and that one at 2913 cm^{-1} is due to the C-H stretching. The band at 1638 cm^{-1} corresponds to the bending mode of the absorbed water. The spectrum shows a peak at 1456 cm^{-1}, which is attributed to the CH_2 bending and that at 1315 cm^{-1} to the O-H bending. The absorption band at 1150 cm^{-1} relates to C-O antisymmetric bridge stretching. All the methylcellulose synthesized from pineapple leaves cellulose with the use of 30% NaOH (methylcellulose A), 40% NaOH (methylcellulose B) and 50% NaOH (methylcellulose C) similarly show all absorption bands in IR spectra as the commercial methylcellulose.

In order to see the methylation process, the main evidence falls on the absorbance intensities of the bands within the region 3600 to 2700 cm^{-1} which were due to OH stretching (around 3500 cm^{-1}) and CH stretching (around 2900 cm^{-1}). As seen in Figure 2, the ratio of absorbance intensities at area 3500 cm^{-1} and 2900 cm^{-1} of both methylcellulose A and B werelower than those of methylcellulose C. These indicated that methylcellulose A and B had undergone a complete methylation process than methylcellulose C as more of CH stretching was formed (due to the substitution by methoxy group) proportional to the decrease in the intensity of OH stretching (due to the loss of OH group). Based on the comparison of the absorbance intensities of methylcellulose A, B, and C with the commercial methylcellulose, The result showed that methylcellulose B had the most similar absorbance intensities in this particular region as the commercial methylcellulose. There were no lignin-associated absorbances at 1730 and 1512 cm^{-1} observed in the spectra of all methylcellulose tested. Thus, it can be said that the purification of cellulose during the cellulose extraction to remove lignin and other impurities were a success.

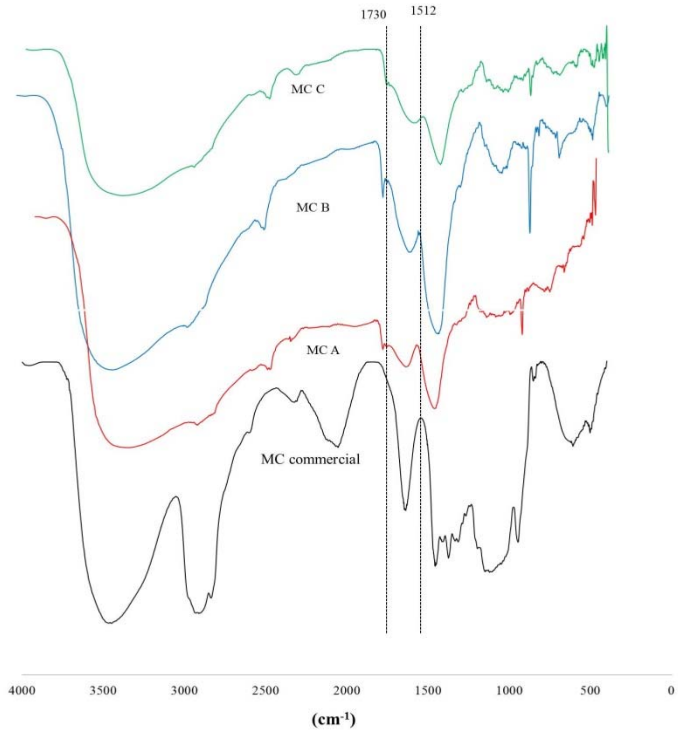

Figure 1. FTIR spectra of methylcellulose by determination of (a) commercial methylcellulose,(b) methylcellulose A (synthesized with 30% NaOH), (c) methylcellulose B (synthesized with 40% NaOH) and (d) methylcellulose C (synthesized with 50% NaOH).

Table 3. Assignment of main absorption bands in the methylcellulose.

Wavenumber (cm^{-1})				**Assignment**
MCcom	MC A	MC B	MC C	
3450	3399	3416	3393	OH stretching
2913	2952	2958	2958	CH stretching
1638	1617	1611	1594	H-O-H bending of absorbed water
1456	1441	1441	1438	CH_2 ending
1315	1295	1298	1295	OH in plane bending
1264	1270	1272	1269	C-O stretching of ether linkage
1150	1136	1155	1150	C-O-C antisymmetric bridge stretching

Note Methyl cellulose A, B, and C showed the extraction by using 30% NaOH, 40% NaOH and 50% NaOH, respectively

3.1.3 Viscosity, water solubility, and color measurement

The viscosity, water solubility and color of the methylcellulose synthesized were also determined in the experiment. Based on Sigma-Aldrich product information, the approximate methylcellulose viscosity is between 15 to 25 cP. The results showed that the commercial methylcellulose (MCc) was the most viscous methylcellulose, followed by methylcellulose B (20.40±0.25cP), while methyl cellulose A has the lowest viscosity value (19.48±0.12 cP).

The methylcellulose was also added in water which was stirred for 24 hr in order to see its solubility. Some of the reaction product was not soluble in water. The methylcellulose synthesized from pineapple leaf cellulose (MCpl) was found to be more soluble than commercial methylcellulose. The solubility of methyl cellulose A (synthesized using 30% NaOH), methyl cellulose B (synthesized using 40% NaOH) and methyl cellulose C (synthesized using 50% NaOH) were 81.00±6.08%, 86.00±2.00%,and87.33±2.31%,respectively while the commercial methyl cellulose was 73.67±1.53% soluble. The insoluble portion of MC from pineapple leaf was in the form of small precipitates while for commercial MC, it tends to form theswelling residue which did not pass through when it was filtered. The high viscosity of commercial methylcellulose may cause it to require alonger period of time in order to solubilize.

For color determination of the methylcellulose, the L* value which refer to lightness was used for comparison. Lightness is the representation of thevariation in the perception of a color space's brightness. A substance can have lightness value between 0 (the darkest color) to 100 (the lightest color). Figure 2 shows the different in color of the methylcellulose. From the experiment, the degree of lightness increase in the order of MC C < MC A < MC B < Commercial MC. Thus, this indicates that the commercial cellulose has the lightest color while methylcellulose C has the darkest color.

Figure 2. (A) Commercial methylcellulose, (B) methylcellulose A synthesized using 30% NaOH, (C) methylcellulose B synthesized using 40% NaOH and (D) methylcellulose C synthesized using 50% NaOH.

When the properties of pineapple leave methylcellulose (MC_{pl}) were compared between one another, methylcellulose B which was synthesized by the use of 40% NaOH was found to have the best properties compared to the two other methylcellulose synthesized using 30 and 50% NaOH. Methylcellulose B exhibited the highest DS, the most similar FTIR spectra as the commercial methylcellulose, highest viscosity, good solubility and the lightest color when compared with methylcellulose A and C. Therefore, methylcellulose B was selected for film applicationby the film properties such as mechanical property, water solubility, water vapor transmission rate (WVTR) and transparency value of MCpl and MCc were investigated.

3.2 Properties of MCpl films and commercial MCc films

3.2.1 Mechanical properties

The mechanical properties of MC_{pl} and MC_c films showed in Table 4. The MCpl film was found to be able to withstand ahigher level of stress as it had a greater tensile strength value than the MCc film. However, the stronger MCpl film cannot extend for long distance. It tends to break at ashorter distance compared to the MCc film as the MCpl film had lower flexibility as represented by the value of % elongation. The MCpl has lower DS value than MCc which it means that the extent of methylation in MCpl was less than MCc. Lower extent of methylation indicated that there was amore hydroxyl (OH) groups presence in MCpl and more methoxy (OCH_3) groups in MCc. The high amount of OH groups in MCpl allows the formation of hydrogen bonds between the methylcellulose chain thus result in higher TS for MCpl. However, the presence of these bonds also causes low flexibility of other film based on MC [23].

3.2.2 Water solubility

Water solubility is an important property of edible films in enhancing product integrity and water resistance. The water solubility of the both of MCc and MCpl films showed in Table 4. The result displayed that the MCc film was lower water solubility than MCpl film. Whilst this effect was reported by Linden and Lorient [24] that the solubility of MC depends on its DS (DS 1.61 for MCc and DS 1.36 for MCpl). The dissolution of methyl cellulose which is a hydrophilic polymer involves the penetration of water to the polymer bulk and swelling. Then, it is followed by the disruption of hydrogen and Van der Walls forces between polymer chains. The chemical degradation which occurs through hydrolyses leads to the creation of oligomers and monomers [25].

Table 4. Mechanical property, water solubility, water vapor transmission rate (WVTR) and transparency value of methylcellulose films.

Film properties	Commercial MC	Pineapple leaf MC
Tensile strength (MPa)	41.58±11.05^{b}	58.30±10.78^{a}
Elongation (%)	35.85±8.77^{a}	19.63±8.15^{b}
Solubility (%)	86.33±1.53^{a}	79.00±2.65^{b}
WVTR (g/m^{2}day)	31.98±2.12^{a}	36.71±2.75^{a}
Transparency (%)	71.08±1.53^{a}	67.57±1.50^{b}

Each value is expressed as mean±SD. Means in colum followed by different letters (a to b) are significantly different (P<0.05), based on Duncan's test.

3.2.3 Water vapor transmission rate (WVTR)

Water vapor transmission rate (WVTR) is very important determination in food packaging because it is directly related to the ability of the film to control moisture transfer between the food and the environment. The result of WVTR of MCc and MCpl films showed in Table 4. From the result in Table 4, the MC filmfrom pineapple leaf (MCpl) was higher water vapor transmission rate (36.71g/m^{2}day) than the commercial methylcellulose (MCc). This effect may be due to the difference in hydrophilicity between MCc and MCpl film, including the thickness of MCpl film was less than MCc which makes the water to easily pass through the MCpl film when compared with MCc film. However, The both MC film from commercial and pineapple leaves showed higher WVTR than other films such as PE film (7.69g/m^{2}day) [26] and LDPE films (a range of 16 to 23 g/m^{2}day) [27]. Whilst MC films are better water vapor barriers than hydrophilic films based on starch and wheat gluten [28, 29].

3.2.4 Transparency value

Transparency of the film is a relevant property as it has a direct impact on the appearance of the packaged product. The transparency values of the films measured by UV-Vis spectrophotometer were shown in Table 4. The MCc film production was reasonably clear while the MCpl film appeared as slightly cloudy, opaque, and yellowish. The transparency value of the MCc film (71.08±1.53%) was significantly higher than MCpl film (67.57±1.50%). The MCpl film solution was slightly immiscible where there was the presence of small particles in the film. As the MCpl film contained immiscible components, these components affect the extent of light scattering where there was more light scattered for the MC_{pl} film and hence, result in lower transparency value. However, the values of transparency for both MCpl and MCc films were higher than the methylcellulose film (61.70%) produced by Noronha *et al*. [30].

4. Conclusions

The properties of methylcellulose synthesized from pineapple leaves and the properties of the film produced from it were successfully investigated and compared with the commercial methylcellulose. Methylation process done with the use of 40% NaOH was found to be the best condition that produced methylcellulose with the best properties and DS value of 1.36±0.03. Methylcellulose from pineapple leaves is a good alternative source for the industry and it can be used to make edible films with good properties. The pineapple leaves methylcellulose film is thin, having high tensile strength (58.30±10.78MPa), and high solubility (79.00±2.65%). However, this film has low flexibility (19.63±8.15%elongation) and transparency (67.57±1.50%) if compared to the commercial methylcellulose film. For further study, it is recommended to find some ways to make the methylcellulose synthesized to have alighter color. Besides, lignin and fat removal may be applied on the pineapple leaves sample before the cellulose extraction process. As the presence of fat may affect the effectiveness of the methylation process hence, it removal may result in the formation of methylcellulose with higher DS value. Other than that, the absence of fat may help to increase the solubility of methylcellulose in water. When methylcellulose is more soluble, there will be no or less formation of small particles on the film hence, the transparency may be increased.

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