Synthesis and Characterization of Carboxymethyl Cellulose Powder and Films from *Mimosa pigra*

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ABSTRACT: *Mimosa pigra* peel was sun-dried for 2 days and then ground before being boiled with 30%w/v sodium hydroxide (NaOH) at 100°C for 3 h, washed and then dried at 55°C to constant weight. The cellulose was then synthesized using different NaOH concentrations and monochloroacetic acid (MCA) in isopropyl alcohol (IPA). Effects of various NaOH concentrations on degree of substitution (DS), viscosity and thermal of carboxymethyl cellulose from *Mimosa pigra* peel (CMC_m) were investigated. The increasing of NaOH concentration resulted in increasing DS and viscosity. However, viscosity of CMC_m decreased as temperature increased. Thermal properties were studied using differential scanning calorimetry (DSC). The melting point of the samples decreased as %NaOH increased. The effects of various NaOH concentrations in CMC_m synthesis on the mechanical properties and water vapor permeability (WVP) of the CMC_m films were investigated as well. With increasing NaOH concentrations (30–50%) were also found to result in improved mechanical properties. However, when the level of NaOH concentration was 60%, the mechanical properties of the CMC films decreased. This result indicates that the highest mechanical properties were found for 50% NaOH-synthesized CMC_m films. The WVP of the CMC_m films increased as %NaOH increased. In addition, the CMC_m films were tested to determine the effect of glycerol as a plasticizer on the mechanical properties. Increasing the amount of glycerol showed an increase in elongation at break but also led to a decrease in tensile strength. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 122: 3218–3226, 2011

Key words: biopolymers; films; structure–property relations; carboxymethyl cellulose; *Mimosa pigra*

INTRODUCTION

Mimosa pigra is a prickly mimosoid native to Mexico, Central and South America. It is also an invasive weed, especially in parts of South East Asia and Australia. In Thailand, *Mimosa pigra* became a serious problem in the early 1980s,[1] by blocking irrigation systems that supply rice fields and reducing crop yields. However, in other locales such as tropical Africa it has also been used as a tonic for diarrhea, gonorrhea and blood poisoning and as animal feedstock.[1] Moreover, *Mimosa pigra* can serve as a pollen source for bees as cited by beekeepers in Thailand.[2] *Mimosa pigra* has also reportedly been used as a cellulose source.[3]

Cellulose, which consists of a linear chain of glucose units, is the most common organic compound on earth and is found in the cell wall of all plants.

Cellulose does not dissolve in water. However, some cellulose derivatives can be dissolved in water such as carboxymethyl cellulose (CMC). CMC is one of the most widely used cellulose derivatives because the CMC production process is simple and low cost. So, it has been used in numerous industrial applications including food,[4] pharmaceutics,[5] and nonfoods.[6–13] The degree of substitution (DS) of CMC is dependent upon the concentration of monochloroacetic acid, temperature and reaction time,[14] where the solubility of CMC increases with increasing DS.[15]

Although cellulose is usually obtained from wood, many researchers have studied the production of CMC from agricultural waste as a cellulose source such as sugar beet pulp cellulose,[16] cavendish banana pseudo stem (*Musa cavendishii* LAMBERT),[17] sago waste,[18] papaya peel,[19] mulberry paper waste.[20] Very little work has been done on production and characterization of CMC film from agricultural wastes.[19–22] However, the production and characterization of CMC film from *Mimosa pigra* has not previously been reported. Therefore, the objectives of this work were: (1) to determine the effect of various NaOH concentrations in the CMC synthesis on DS of CMC (2) to study the effect of various NaOH

concentrations on the mechanical properties and water vapor permeability (WVP) of CMC films, and (3) to investigate the effect of the amount of glycerol as a plasticizer on the mechanical properties of CMC films.

METHODOLOGY

Materials

Mimosa pigra peel was obtained from Muang District, Chiang Rai Province, Thailand. Sodium hydroxide (artificial grade) and glacial acetic acid were purchased from Lab-scan; isopropyl alcohol (IPA), ethanol and absolute methanol from Northern Chemical; hydrochloric acid and sodium chloride from Merck, Germany and chloroacetic acid from Sigma-Aldrich, USA.

Material preparation

Mimosa pigra trees naturally grow in various sizes in Chiang Rai Province, Thailand. In this study, trees were separated into five size categories (1.5, 2.0, 2.5, 3.0, and 3.5 cm) by trunk diameter. The moisture content of the *Mimosa pigra* trunk was determined and the peel was sun dried about 2 days. The *Mimosa pigra* peel was weighed to calculate the moisture content and percent dryness of *Mimosa pigra* peel. The dried peel was cut into small pieces and then ground with a hammer mill (Armfield, England). The resulting powder was then screened to a size below 1 mm. The percent dryness of *Mimosa pigra* powder was measured. Two grams of *Mimosa pigra* powder were weighed and dried in an oven at 105°C for 4 h. and the percent dryness calculated. The dried *Mimosa pigra* peel powder was kept in polypropylene bags at room temperature for further study.

Cellulose extraction from dried *Mimosa pigra* peel powder

The dried *Mimosa pigra* peel powder was extracted with 30% (w/v) sodium hydroxide (NaOH) solution at a ratio of cellulose to solvent 1 : 20 (w/v) at 100°C for 3 h. The obtained black slurry was then filtered and washed with cold water until the washed water was neutral. The residue was dried in an oven at 55°C to constant weight.[19] The dried cellulose pulp from *Mimosa pigra* peel was ground, passed through 200 mesh sieve and kept in polyethylene bags to use for modification of CMC_m in the next step. The percent yield of cellulose pulp was calculated using eq. (1):

$$\text{Yield of cellulose pulp (\%)} = \frac{\text{Weight of dried cellulose pulp (g)} \times 100}{\text{Weight of Mimosa pigra powder (g)}} \quad (1)$$

Synthesis of carboxymethyl cellulose

Ten grams of cellulose powder, 100 mL for various NaOH concentrations (30, 40, 50, and 60% (w/v)) and 500 mL of isopropanol were mixed in a beaker for 1.5 h. The carboxymethylation reaction was started by adding 15 g of chloroacetic acid and continuously stirring for 1.5 h. The beaker containing the mixture was covered with aluminum foil and placed in oven at 55°C for 3.5 h. After being heated, the solution separated into two phases. The liquid phase was removed and the solid phase was suspended in 66.67 mL of absolute methanol, neutralized with acetic acid (90% v/v) and then filtered using a Buchner funnel. The final product was washed 5 times by soaking in 200 mL of ethanol (70% v/v) for 10 min to remove undesirable byproducts, and was then washed again with 200 mL of absolute methanol. The obtained CMC_m was dried in an oven at 55°C for 12 h[19].

Degree of substitution (DS)

The degree of substitution (DS) of CMC is the average number of hydroxyl group in the cellulose structure which was substituted by carboxymethyl and sodium carboxymethyl groups at C2, 3, and 6. DS was determined by the USP XXIII method described for Crosscarmellose sodium. The methods include two steps—titration and residue on ignition.[23] The DS of CMC_m can be calculated using eq. (2):

$$\text{DS} = A + S \quad (2)$$

where A is the degree of acid carboxymethyl substitution which can be calculated using eq. (3):

$$A = \frac{1150M}{(7120 - 412M - 80C)} \quad (3)$$

S is the degree of sodium carboxymethyl substitution which can be calculated using eq. (4):

$$S = \frac{(162 + 58A)C}{(7120 - 80C)} \quad (4)$$

where M is the net milliequivalent of base required for the neutralization of 1 g CMC_m as determined in titration testing; C is the percentage of residue on ignition of CMC_m as determined in residue on ignition testing.

Titration testing

About 1 g of CMC_m, accurately weighed, was added into a 500 mL Erlenmyer flask and followed by 300 mL of sodium chloride solution (10% w/v). The

Erlenmyer flask was closed with a stopper and allowed to stand for 5 min with intermittent shaking. Five drops of *m*-cresol purple and 15 mL of hydrochloric solution (0.1*N*) were added and the mixture was shaken. If the solution was still violet, hydrochloric solution (0.1*N*) was added until the solution became yellow. In the next step, the solution was back titrated with sodium hydroxide (0.1*N*) until the solution turned to violet at the endpoint. The net amount of milliequivalent base required for neutralization of 1 g CMC_m (*M*) was calculated using eq. (5)[23]:

$$M \text{ (mEq)} = \text{mmole} \times \text{Valence} \qquad (5)$$

where *m* is 10^{-3}; mole is mass in grams per molecular weight of NaOH and the valence of NaOH is 1.

Residue on ignition testing

A crucible was placed in the oven at 100°C for 1 h and kept in a desiccator until the weight reached a constant value. CMC_m 1000 g was added into a crucible. To obtain the black residue, the crucible containing CMC_m was ignited at 400°C for about 1–1.5 h and placed into the desiccator. Sufficient sulfuric acid was used to moisten the entire residue and heated until the volatization of white fumes was completed. The crucible containing residue was ignited at 800°C $\pm$ 25°C to obtain the white residue and placed in the desiccators to achieve an accurate weight. All treatments were made in triplicate. The percentage of residue on ignition was calculated using eq. (6)[23]:

$$C = \frac{(\text{Weight of residue})}{\text{Weight of CMC}} \times 100 \qquad (6)$$

Infrared spectroscopy (IR)

The functional groups of the cellulose and CMC_m samples were determined by using infrared spectrophotometer (Bruker, Tensor 27, Germany). Pellets were made from cellulose and CMC_m sample ($\sim$ 2 mg) with KBr.[22]

Viscosity

The viscosity of cellulose and CMC_m was measured using a Rapid Visco Analyzer (Model: RVA-4, Australia). The sample solution was prepared by dissolving 3 g of CMC_m in 25 mL of water (12% w/v) by stirring at 80°C for 10 min. Viscosity was performed in two steps. In the first step, the speed was set at 960 rpm for 10 s. For the second step the temperature was varied from 30, 40, 50, and 60°C at 5 min

intervals with a speed of 160 rpm. All measurements were performed in triplicate.

Thermal properties

The thermal property according to the melting point of CMC_m was determined using differential scanning calorimetry (DSC) DSC823 Mettler Toledo Schwerzenbach instrument (Mettler Toledo, Switzerland). Ten milligrams of each sample contained in aluminum pans were heated from -50 to 350°C with a heating rate of 10°C min^{-1} under N_2 gas at a flow rate of 50 mL min^{-1}. Melting point was determined from the thermogram. All samples were run in triplicate.

Film preparation

The film-forming solutions were prepared by dissolving 3.0 g of CMC_m in a constantly stirred 100 mL of distilled water at 80°C for 10 min. To obtain film-forming solutions with plasticizer, the solutions were added with various amount of glycerol (0.1, 0.2, and 0.3 mL). The solutions were cooled to around 20–25°C and cast onto cellophane plates (30 cm $\times$ 15 cm). CMC_m film was obtained by drying at room temperature for 36 h. Then, the film was peeled off the plates. Thickness of film was controlled by the volume of solution (60 mL) on each plate.

Mechanical properties

The film samples were cut for mechanical properties testing. Specimens of $1.5 \times 14 \text{ cm}^2$ rectangular strips were used for tensile strength and folding endurance. CMC_m films were preconditioned at 27°C $\pm$ 2°C with 65% $\pm$ 2% relative humidity (RH) for 24 h (Thai Industrial Standard; TIS 949-2533). The tensile strength (TS) and percentage elongation at break (EB) were measured using an Instron Universal Testing Machine Model 1000 (H1K-S, UK) according to ASTM Method (ASTM, D882-80a, 1995a). The initial grip separation and cross-head speed were set at 100 and 20 mm min^{-1}, respectively. The TS value was calculated by dividing the maximum load with the initial cross-sectional area of the specimen. The EB value was calculated as the percentage of change of the initial gauge length of a specimen (100 mm) at the point of a sample failure. The folding endurance was measured using MIT Folding Endurance Tester (GT-6014A, UK) according to ASTM Method (ASTM, D2176-1993). The load tension of 0.5 kg and a uniform rate of 175 $\pm$ 25 double folds/min were used in testing. All mechanical tests were performed in 10 replications.

Water vapor permeability (WVP)

Water vapor permeability (WVP) of the CMC_m films was measured using the ASTM method (ASTM, E96-93, 1993). The CMC_m film specimen was cut into circle (7-cm diameter). Circular aluminum cups with a diameter of 8 cm and depth of 2 cm were used. Each cup containing 10 g of dried silica gel was covered with a circular film (diameter of 7 cm) and sealed with paraffin wax. The sealed cups were weighed and kept in a desiccator with saturated solution of sodium chloride (NaCl) for providing 25°C, 75% RH. Then, the cups were weighed every day for 2 weeks to provide the slope of weight gain (Y axis) and time (X axis). The water vapor transmission rate (WVTR) of the films was measured from the weight gain of the cups and calculated by dividing the slope of the film area following eq. (7):

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

where the film area was 28.27 cm^2.

The water vapor permeability (WVP) (g m^{-2} mmHg^{-1} day^{-1}) was calculated as eq. (8):

$$WVP = \frac{WVTR \times L}{\Delta P} \quad (8)$$

where L is the mean film thickness (mm) and ΔP is the partial water vapor pressure difference (mmHg) across the two sides of the film specimen (the vapor pressure of pure water at 25°C is 23.73 mmHg).

Statistical analysis

Data were analyzed by Duncan's Multiple Range Test ($P \leq 0.05$) using the SPSS software program.

RESULTS AND DISCUSSION

Material acquirement

The effect of various dimensions of *Mimosa pigra* tree [1.0–1.5 (Group 1), 1.5–2.0 (Group 2), 2.0–2.5 (Group 3), 2.5–3.0 (Group 4), and 3.0–3.5 (Group 5) cm] on percent dryness of *Mimosa pigra* peel was investigated. The peels for various tree dimensions provided an appreciably similar percent dryness value between Groups 1 and 4 (40–43% dryness value), with Group 5 having a slightly higher value (55%). The majority of trees were of an average size of 2.0–2.5 cm (Group 3) with *Mimosa pigra* trees in Groups 1 and 5 rarely being found. The dried peel was then ground in a hammer mill into small sizes of about 1-mm dimensions and the percent dryness of *Mimosa pigra* powder was measured again. Two grams of *Mimosa pigra* powder were weighed and dried in an oven for 4 h and the percent dryness calculated. The values of percent dryness for each tree were not significantly different showing about 88% dryness for all samples.

Cellulose extraction and CMCm synthesis from dried *Mimosa pigra* peel powder

The delignification using NaOH solution due to NaOH pretreatment resulted in the highest level of cellulose.[24] In this study, the percent yield of cellulose pulp from *Mimosa pigra* powder was delignified with various NaOH concentrations (10–60%). The percentage yield of cellulose pulp increased as NaOH concentrations (10–30%) increased because the delignification was improved at higher NaOH concentration.[25] Nevertheless, it slightly decreased after increasing of NaOH concentration above 30% due to the alkali-catalyzed degradation of cellulose.[26] Therefore, 30% (w/v) NaOH was chosen to extract cellulose in this experiment.

The *Mimosa pigra* powder was cooked by 30% (w/v) NaOH at ratio of cellulose to solvent 1 : 20 (w/v) at 100°C for 3 h to obtain cellulose. For all sized *Mimosa pigra* trees, the percent yield of extracted cellulose was ∼ 25% (data not shown). The percent yield of cellulose is not different in *Mimosa pigra* trees from Groups 1 to 4. The *Mimosa pigra* trees in Group 5 provided the highest percentage yield of cellulose. However, Group 5 does not occur abundantly in the wild so samples are hard to collect as discussed previously. The cellulose was then synthesized using different NaOH concentrations (30, 40, 50, and 60%w/v) and monochloroacetic acid (MCA) in isopropyl alcohol (IPA). The effect of sodium hydroxide on properties of CMC_m and CMC_m film was discussed in the next section.

Effect of NaOH on degree of substitution of CMC_m

The effect of various NaOH concentrations CMC synthesis on the DS of CMC_m is shown in Figure 1. As NaOH concentration increased from 30 to 50%, the DS of CMC_m increased. However, the DS decreased at 60% NaOH concentration. This phenomenon can be explained by the carboxymethylation process as described in Barai et al.[27] and Pushpamalar et al.[18] The two competitive reactions took place simultaneously. The first reaction was a cellulose hydroxyl reaction with sodium monochloroacetate (NaMCA) to obtain CMC_m as shown in eq. (9).

$$Cell - OH + ClCH_2COONa \rightarrow$$
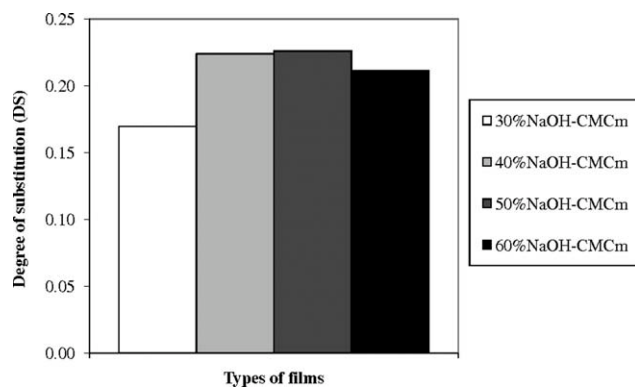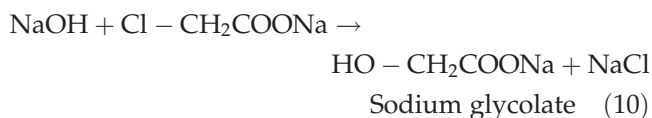$$Cell - O - CH_2COO^-Na^+ + NaCl + H_2O \quad (9)$$
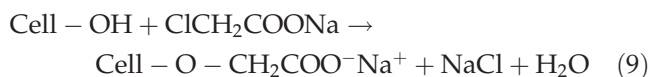
Carboxymethyl cellulose

Figure 1 Effect of various NaOH concentrations in the alkalization reaction on DS of CMC synthesized from *Mimosa pigra* peel (CMC$_m$).

The second reaction with NaOH and NaMCA to form sodium glucolate as byproduct follows eq. (10):

$$NaOH + Cl - CH_2COONa \rightarrow$$
$$HO - CH_2COONa + NaCl$$
$$Sodium\ glycolate \quad (10)$$

The second reaction overcomes the first at stronger alkaline concentration. If the alkaline level is too high, a side reaction predominates with the formation of sodium glycolate as a by product, thus lowering of DS. Moreover, the decline of DS at the higher 60% NaOH concentration could also be due to chain degradation of CMC polymers,[27] which is similar to the results found in other studies.[17,18]

Infrared spectroscopic analysis

Infrared spectroscopy spectra of cellulose from *Mimosa pigra* and CMC$_m$ synthesized with 30% NaOH concentration are shown in Figure 2. The results of CMC$_m$ with other NaOH concentrations were similar to 30% NaOH; therefore, the data are not shown. Cellulose [Fig. 2(a)] and CMC$_m$ [Fig. 2(b)] provided the same functional groups such as hydroxyl groups (OH), hydrocarbon groups (CH$_2$), carbonyl groups (C=O), and ether groups (—O—). The broad band at 3200–3600 cm^{-1} is due to O—H stretching. The bands at 1450 and 1000–1200 cm^{-1} are due to —CH$_2$ scissoring and —O— stretching, respectively. The band at 3000 cm^{-1} is due to C—H stretching vibration. The band around 1600 cm^{-1} is due to C=O stretching.[22] In the CMC$_m$ sample [Fig. 2(b)], the carbonyl group (C=O), methyl group (—CH$_2$) and ether group (—O—) group outstandingly increased, but the band of hydroxyl group (—OH) decreased comparing with cellulose [Fig. 2(a)]. This result confirmed that carboxymethylation took place on the cellulose molecules.[22] Similar observations have been reported previously.[17–19,22]

Effect of various NaOH concentrations on viscosity of CMC$_m$

It is known that the viscosity of CMC is affected by the CMC concentration,[28] NaOH concentration[29] and sodium monochloracetate (NaMCA) concentration.[17] Adinugraha et al. (2005)[17] found that the DS of CMC increased with increasing NaMCA concentration. DS of CMC at low NaOH concentrations increased and would level off as NaOH concentration above 20% (with 3–5 g NaMCA). However, in our study, viscosity CMC$_m$ increased with increasing of NaOH concentration (Fig. 3) due to more carboxymethyl groups acting as hydrophilic groups substituted the hydroxyl groups of cellulose polymers. Therefore, the increase in the DS of CMC improved the ability of CMC to immobilize water in a system. Even though the DS slightly decreased at 60% NaOH, the viscosity of CMC$_m$ increased most likely caused by the aggregation of CMC$_m$. This behavior should be investigated further.

In addition, the effect of temperature on the viscosity of CMC$_m$ was also investigated. The viscosity of CMC$_m$ decreased as temperature increased. The effect of increasing the temperature of a CMC solution is to reduce the cohesive forces while simultaneously increasing the rate of molecular interchange. The former effect tends to cause a decrease of shear stress, while the latter causes it to increase. The net result is that liquids show a reduction in viscosity with increasing temperature.[30]

Effect of various NaOH concentrations on thermal properties of CMC$_m$ powder

The effect of various NaOH concentrations on the thermal property of cellulose from *Mimosa pigra* peel and CMC$_m$ powder was determined by differential scanning calorimetry (DSC). The DSC thermograms of cellulose and CMC$_m$ are shown in Figure 4. The melting temperature (T_m) of cellulose (*Mimosa pigra* pulp) was 107.3°C, and CMC$_m$ synthesized with 30, 40, 50, and 60% NaOH was 114.5, 106.7, 98.3, and 98.1°C, respectively, as shown in Figure 5. The melting temperature of CMC$_m$ slightly decreased as the level of NaOH increased. While the %NaOH in the alkalization reaction increased, the substitution of carboxymethyl group also increased. This change in melting temperature results from the interference with crystallinity caused by the presence of random irregularities produced by the relatively bulky side groups from carboxymethyl group.[31]

Effect of various NaOH concentrations on mechanical properties of CMC$_m$ films

The tensile strength (TS) of CMC$_m$ films with various NaOH concentrations in CMC synthesis are
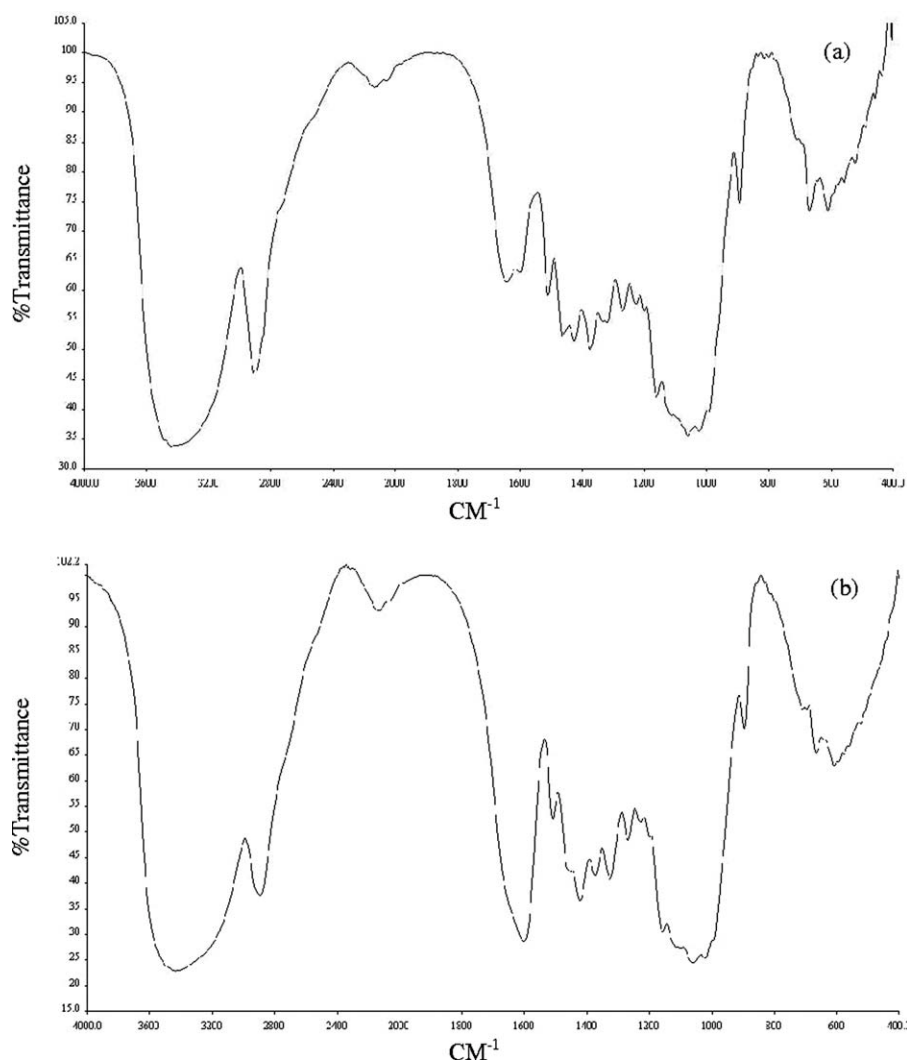
Figure 2 FTIR spectra of (a) cellulose from *Mimosa pigra* peel and (b) CMC_m synthesized with 30%NaOH concentrations.

shown in Figure 6 where the TS of the films increased with increasing concentrations of NaOH (30–50%). The increase in the TS value was correlated with an increasing DS value due to the substituent of carboxymethyl groups which affected an increase in the ionic character and intermolecular force between the polymer chains.[17] Nevertheless, the TS declined at high NaOH concentrations due to the sodium glycolate formation as a byproduct in the synthesis of CMC and polymer degradation. Barai et al. (1997)[27] reported that at high NaOH concentrations, the formation of sodium glycolate increased providing a decreasing in the CMC content thus lowering the intermolecular forces.

The percent elongation at break (EB) of CMC_m films with various NaOH concentrations in CMC synthesis are shown in Figure 7. The EB of these CMC_m films increased with increasing NaOH concentrations (30–50%) although it declined at a 60% NaOH concentration. This can be explained since at higher NaOH concentrations cellulose swells more

which decreased the crystallinity of the cellulose structure with a flexibility increase. However, at high concentrations of NaOH, CMC_m films obtained
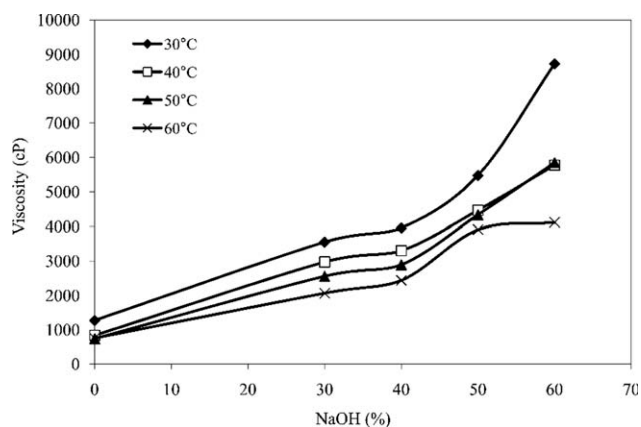

Figure 3 Effect of various NaOH concentrations on viscosity of CMC_m synthesized from *Mimosa pigra* peel at different temperatures.
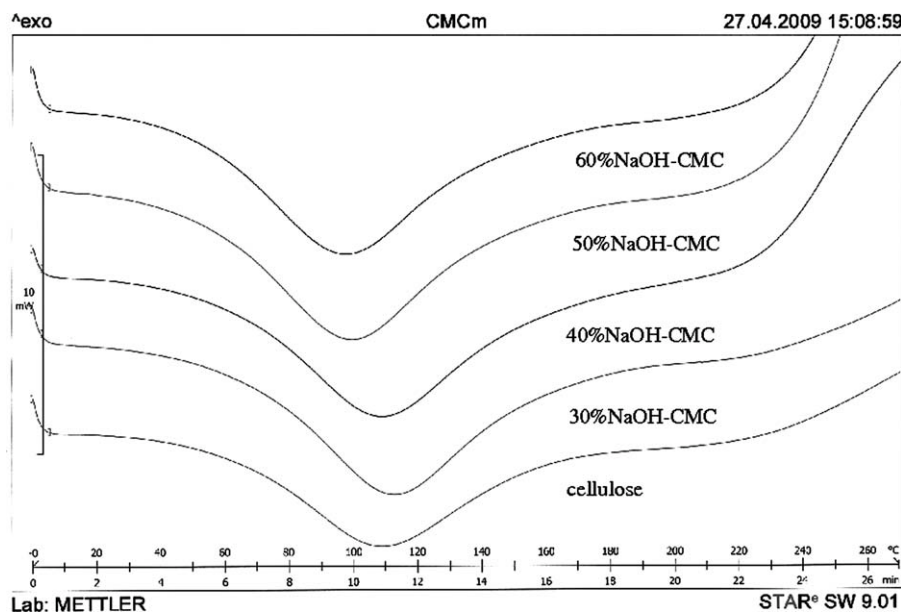
Figure 4   DSC thermograms of cellulose and CMC from *Mimosa pigra* peel with various NaOH concentrations.

the lower flexibility due to hydrolysis reaction in cellulose chain occurred.[32]

Determining of folding endurance is normally employed for paper and plastic materials. Fold endurance measures the durability of materials when repeatedly folded under constant load. Folding endurance or strength is measured and reported in the number of double folds. The test is primarily used for testing paper or plastic films that are frequency handled such as currency papers and wrapping paper or film. In this research, we tried to synthesize CMC from a nonvaluable weed, *Mimosa pigra*, to replace or decrease use of synthetic polymer. Thus, the folding endurance is an important value to indicate the potential of CMC_m film as a packaging material. The folding endurance of CMC_m films formed using various NaOH concentrations in CMC synthesis are shown in Figure 8. The folding endurance of the CMC films increased with increasing NaOH concentrations for (30–50%) along with the flexibility of

the film formed. Nonetheless, the folding endurance of CMC_m films synthesized with a 60% NaOH concentration decreased because of the lower flexibility of the film (lower EB value) (Fig. 7). This phenomenon was possibly due to the shorter polymer chain length generated in the hydrolysis reaction at high alkaline concentrations.[20,33]

Effect of various NaOH concentrations on water vapor permeability (WVP) of CMC_m films

The weight gain of CMC_m films synthesized with various NaOH concentrations increased with increasing time where the slope can be used to determine the water vapor transmission rate (WVTR). The water vapor transmission rate (WVTR) and water vapor permeability (WVP) for CMC_m films synthesized with various NaOH concentration are shown in Table I. The WVP of the CMC_m films
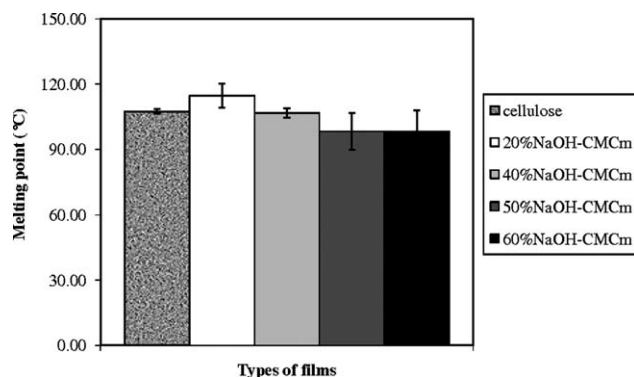

Figure 5   Melting points of cellulose and CMC from *Mimosa pigra* peel with various NaOH concentrations.
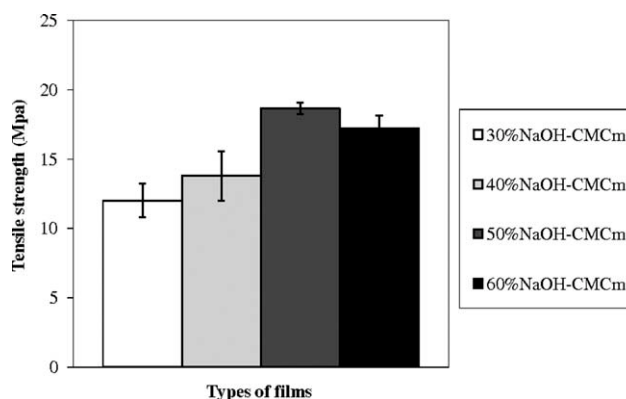

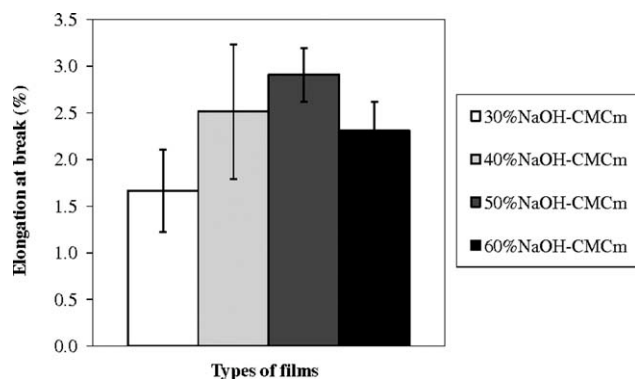**Figure 6**   Tensile strength of CMC films synthesized with various NaOH concentrations (30, 40, 50, and 60%).

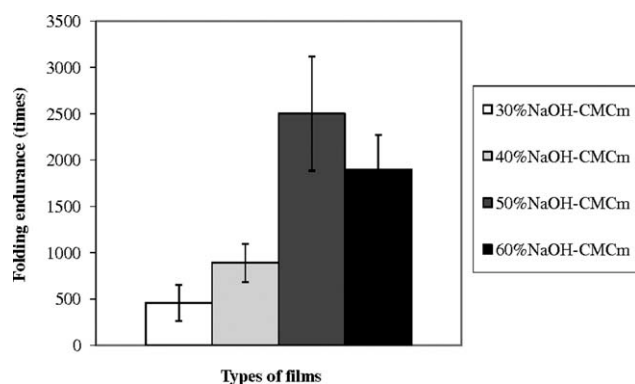**Figure 7** Percent elongation at break of CMC films synthesized with various NaOH concentrations (30, 40, 50, and 60%).

Figure 8 Folding endurance of CMC_m films synthesized with various NaOH concentrations (30, 40, 50, and 60%).

Figure 9 Tensile strength of CMC films with various amounts of glycerol (0.0, 0.1, 0.2, and 0.3 mL) used as a plasticizer.

Figure 10 Percent elongation at break of CMC films with various amounts of glycerol (0.0 (control), 0.1, 0.2, and 0.3 mL) used as a plasticizer.

increased when NaOH concentrations increased because the material changed from cellulose to CMC_m which causes a higher polarity[18] [see eq. (9)]. The results were in agreement with Rachtanapun et al.[34] They also reported that WVP of carboxymethyl rice starch film increased with increasing of NaOH concentrations. In addition, Li et al.[35] found that carboxymethylation affected to reduction crystallinity, change in granule morphology, and increase of polarity.

The results from the experiment indicate that the CMC_m film synthesized with 50% NaOH concentrations present the best mechanical properties since WVP of CMC_m films slightly increased by NaOH
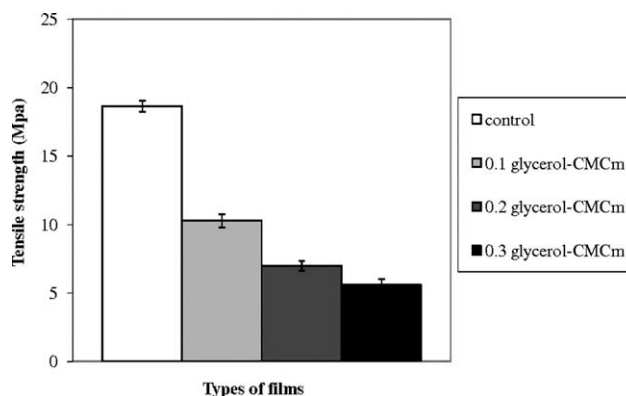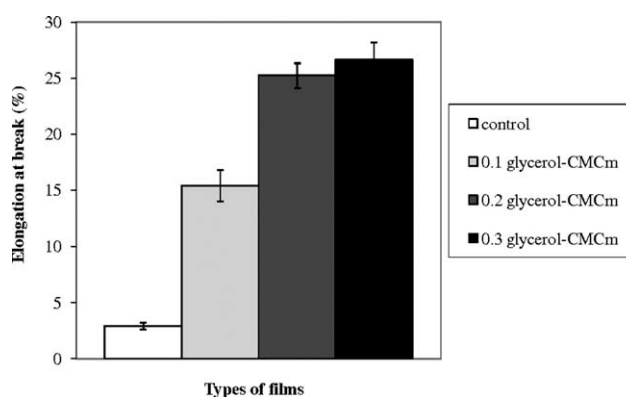
concentration. However, the CMC_m film was not suitable for utilization due to its brittleness. Therefore, this method was chosen to improve the properties to overcome the brittleness by adding glycerol as plasticizer in the next experiment.

Effect of amount of glycerol on mechanical properties of CMC_m films

To improve the properties of CMC_m films, a plasticizer (glycerol) was added to the film-forming solutions before casting. The effect of the amount of

TABLE 1
Water Vapor Transmission Rate and Water Vapor Permeability for CMC_m Films Synthesized with Various NaOH Concentrations (30%, 40%, 50%, and 60%)

Type of films	Thickness (mm)	Water vapor transmission rate (WVTR) (g/day.m^2)	Water vapor permeability (WVP) (g.m/m^2.mmHg.day)
30%NaOH-CMC	0.041 ± 0.002^b	65.641 ± 0.174^a	1.512 ± 0.004^f
40%NaOH-CMC	0.048 ± 0.002^c	65.723 ± 0.644^a	1.773 ± 0.002^g
50%NaOH-CMC	0.054 ± 0.003^d	66.207 ± 1.772^a	2.009 ± 0.005^h
60%NaOH-CMC	0.061 ± 0.003^e	66.466 ± 1.827^a	2.278 ± 0.006^i

Different letters in the same column indicate significant differences between the means obtained in Duncan's test ($p < 0.05$).

glycerol on mechanical properties of CMC_m films was also studied. The TS of CMC_m films decreased with increasing amounts of glycerol (Fig. 9) but EB of CMC_m films increased with increasing amount of glycerol (Fig. 10). This is because the glycerol increased the flexibility of the films due to its ability to reduce internal H-bonding between polymer chains with increasing space.[36] The increasing in flexibility with increasing plasticizer content in hydrophilic films has been reported previously.[19,37–40]

CONCLUSIONS

The results obtained in this study showed that *Mimosa pigra* peel could be successfully used as a raw material, to produce carboxymethyl cellulose (CMC_m) using various NaOH concentrations. This shows that NaOH content is the main parameter which correlates with the properties of CMC_m. The DS of CMC_m increased with increasing of NaOH concentration (30–50%) in CMC_m synthesis and declined at 60% NaOH concentration. The mechanical properties of CMC_m film had a strong relationship with the DS. To improve the mechanical properties, CMC_m had to consist of a high DS. The most optimized mechanical properties were found for 50%w/v NaOH-synthesized CMC_m films.

CMC is used in a wide variety of food, pharmaceutical and nonfood industries which could find the results presented here to be useful. Understanding methods to modify cellulose such as varying the NaOH levels examined in this article provide insight into new and potentially beneficial uses for cellulose from *Mimosa pigra* peel.

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