

Preparation and Characterization of Palm-based Sodium Carboxymethyl Cellulose for Application in Food Additive

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Abstract: In this study, α -Cellulose extracted from oil palm empty fruit bunches (OPEFB) was used as the raw material for producing different grade carboxymethyl cellulose (CMC). For the conversion into carboxymethyl cellulose, the α -cellulose was subjected to an etherification process, using sodium hydroxide and monochloroacetic acid (MCAA), with isopropanol as a supportive medium. The calculated CMC yield from cellulose ranged from 115.43% to 149.35%. The results indicated that the concentration of NaOH did affect the properties of produced CMC. At higher concentrations of alkali, NaOH reacts with sodium monochloroacetate to form sodium glycolate, which leads to low purity of CMC produced. CMC's purity was increase with increasing the alkali concentration from 25 to 30% of NaOH and then decreased slightly. At a low concentration of 25 % of NaOH solutions, the DS value is higher, while the DS value decreases when the NaOH concentration increases to 35 %. The produced CMC was having a wide range of viscosity depend on temperature and CMC concentration. These optimization factors allowed CMC high purity, providing plenty of opportunities for its multi-application and could be exploited as food additives.

Keywords: carboxymethyl cellulose; empty fruit bunch; food additive.

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1. Introduction

Agricultural wastes or by-products from agricultural activities are blamed for causing massive litter to landfill and leading to pollution [1]. The increasing environmental concerns have forced researchers to utilize material from agricultural waste to obtain useful industrial materials. Agriculture waste consists of carbohydrate polymers (polysaccharides), which contains various fine chemicals through chemical and biochemical modifications. The conversion of agricultural waste into fine chemicals would lighten a variety of socio-economic problems.

The main component of these agricultural wastes is cellulose from plant cell walls. Cellulose can be chemically altered to be converted to useful chemical feedstock [2]-[4]. As cellulose is a renewable natural resource, it has economic gains over other synthetic polymers and can be utilized in various industries such as paper and packaging materials. However, due to inter- and intra- molecular hydrogen bonds in its structure, cellulose does not melt nor dissolve in most common solvents [5]. To utilize cellulose in the food industry as a thickening, gelling, or stabilizing agent, cellulose must be converted to its derivatives.

Among all these modified cellulosic products, CMC has been produced largely due to its wide commercial applications. Carboxymethylation of cellulose is a versatile modification since it provides access to water-soluble materials. It can also be intermediate with various valuable features [6]. CMC has been accepted in a broad circle of applications within different industries, including food ingredients, pharmaceuticals, cosmetics, paper products, ceramics, detergents, adhesives, lithography, and materials [7]-[9]. Large quantities of CMC are produced in crude commercial grades, without any further refining, used as a component in detergents, cloth, pesticide, adhesives, ceramics, lubricants, oil drilling mud, cement, and in the paper and coating industry [10]. Only high-purity grades CMC will be used in the food and pharmaceutical industries.

Sodium carboxymethyl cellulose is man modified water-soluble cellulose derivatives produced by converting alkali cellulose swollen in a certain concentration of sodium hydroxide (NaOH) and organic solvent with monochloroacetic acid or its sodium salt under heterogeneous reaction conditions [11,12]. There are 3 hydroxyl groups (-OH) in cellulose structure and usually can be replaced by carboxymethyl groups in the order of C2>C6>C3 [13]-[15].

Most of the cellulose sources used to produce CMC commercially are cotton and wood, but many other resources could also be used. Some papers have reported the synthesis of CMC, from different agro-cellulosic sources, such as paddy straw [16],[17], kenaf [18], sugar cane bagasse [19], pulp beet [20],[21], cavendish banana pseudostem [22], bagasse of sago [23], pod husk cacao [24], asparagus stalk [25], tea stalk [26] etc.

To date, there is a limited amount of published work on the synthesis of CMC from oil palm biomass, especially oil palm empty fruit bunches (OPEFB). OPEFB is generated during the extraction of palm oil from fresh fruit bunches, and the major component in OPEFB is 40-50% of cellulose. This study aims to prepare high purity CMC from OPEFB.

2. Materials and Methods

2.1. Materials.

Shredded fibers OPEFB was collected from Hulu Langat Palm Oil Mill. After drying, OPEFB fibers were kept in a plastic bag and stored at room temperature. Analytical grade sodium hydroxide, monochloroacetic acid (MCAA), and glacial acetic acid were purchased from Merck Sdn Bhd. (Malaysia). Isopropanol (IPA), ethanol, and methanol were obtained from the local market (System). Sodium chlorite (NaClO₂) used in this project was 80% pure and purchased from Acros (Belgium).

2.2. Method.

2.2.1. Preparation of holocellulose from OPEFB fibers.

In this study, holocellulose was extracted from OPEFB fibers according to American Standard Test Method (ASTM), No. D 1104-56 [27], which is sodium chlorite-acetic acid or chlorous acid method. 4.0 grams of EFB was mixed with 160 ml distilled water, 0.5 ml glacial acetic acid, and 6.0 grams of sodium chlorite. Then, the sample was heated to 70-80 °C with occasional swirling for 1 hour. Under the same conditions and amount of water, acetic acid and sodium chloride were added at hourly intervals for a period of 4 hours. Then, the mixture was filtered, and the solid residue, holocellulose was washed with hot distilled water and acetone to remove the odor of acetic acid. Finally, the holocellulose was placed in an oven at 100-105

°C for 2 hours and cooled in a desiccator before weighing it. The yield (%) of holocellulose was measured based on the dry weight basis and calculated using the following equation:

$$\text{Yield (\%)} = \frac{\text{Weight of dried holocellulose}}{\text{Weight of dried OPEFB}} \times 100 \quad (1)$$

2.2.2. Preparation of cellulose from holocellulose.

The α -cellulose was extracted from holocellulose according to ASTM No. D 1103-60 [28]. 20 ml of 17.5 % sodium hydroxide (NaOH) was added to the beaker containing 2.0 gm of holocellulose. At intervals of 5 minutes, for a period of 15 minutes, 10 ml of 17.5 % NaOH was added. After that, 67 ml of distilled water was added to the sample, and the mixture was incubated for 45 minutes. The α -cellulose was filtered and washed with 50 ml of 8.3 % NaOH, 50 ml of glacial acetic acid, and distilled water. Finally, the α -cellulose was oven-dried at 100-105 °C for 2 hours and cooled in a desiccator before weighing it. Product yield (%) was measured based on the dry weight basis and calculated using the following equation:

$$\text{Yield (\%)} = \frac{\text{Weight of dried cellulose}}{\text{Weight of dried holocellulose}} \times 100 \quad (2)$$

2.2.3. Synthesis of carboxymethyl cellulose (CMC).

The CMC can be synthesized from cellulose via its reaction with alkali and monochloroacetic acid [29-31]. The steps involved in the CMC production is shown in Figure 1.

CMC was produced by etherifying the hydroxyl groups with monochloroacetic acid (MCAA) in the presence of aqueous alkali, NaOH. Cellulose was dispersed in 200 ml of isopropanol (IPA) and 30 ml of NaOH (25~35 % v/v) under constant stirring for 1 hour 30 minutes. After that, an MCAA amount was added, and the mixture was heated in the oven (at 30~70°C) for 4 hours. After cooling, the mixture was neutralized with acetic acid until pH 6-8 was achieved and precipitated with 200 ml of methanol. The solid fibers in the solution were filtered off and washed with several portions of ethanol to obtain the final product, CMC, which later was oven-dried at 100-105 °C for 2 hours and cooled in a desiccator before weighed. The produced CMC in 3 different concentrations of NaOH, namely CMC A (in 25% of NaOH), CMC B (in 30% of NaOH) and CMC C (in 35% of NaOH).

2.3. Characterization of Carboxymethyl cellulose, CMC.

2.3.1. Yield measurement.

The produced CMC yield (%) was measured based on the dry weight basis and calculated using the following equation:

$$\text{Yield (\%)} = \frac{\text{Weight of dried CMC}}{\text{Weight of dried cellulose}} \times 100 \quad (3)$$

2.3.2. Fourier transform infrared spectroscopy (FTIR).

The FTIR spectra of test samples were collected using Perkin Elmer TG-IR Hyphenation consist of FT-IR Frontier with scanning range between 650~4000 cm^{-1} at a resolution of 8 cm^{-1} .

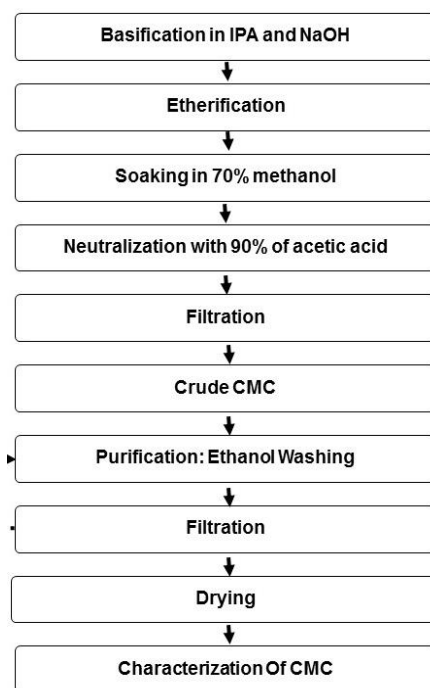


Figure 1. Process flow of CMC synthesized from cellulose.

2.3.3. Purification of CMC.

A purification of materials was carried out according to ASTM No. D 1439-03. 0.5 gm of dried CMC was weighed, and 75 ml of 80 % hot ethanol (60 °C) was added. The beaker containing the mixture was then placed in constant-temperature water at 60 °C for 10 minutes with continuous stirring. The mixture was filtered, and the same amounts of ethanol (60 °C) were added again and placed under the same temperature for 10 minutes. After that, the mixture was filtered, and 150 ml of ethanol (60 °C) was added and filtered. Finally, the mixture was washed with 30 ml of 95% ethanol before oven-drying it at 105 °C for 2 hours. The dried materials were cooled in a desiccator before weighing them. The purity of CMC was calculated as follows:

$$\text{Purity (\%)} = (A \times 10000) / [B (100-C)] \quad (4)$$

where A= weight of dried residu; B=weight of specimen used; C=moisture in the specimen, %.

2.3.4. Determination of degree of substitution, DS.

The morphology of synthesized CMC was examined by Hitachi S-3400N Scanning Electron Microscope (SEM) equipped with Bruker XFlash 6I10 Energy Dispersive X-Ray (EDX) analysis. The samples were analyzed without coating. The DS values were determined by wt %Na values using the following formula [23], [32]-[33].

$$\text{Degree of substitution} = \frac{162 \times \% \text{ Sodium}}{2300 - (80 \times \% \text{ Sodium})} \quad (5)$$

where, 162 = MW of the anhydrous glucose unit; 80 = net increment in the anhydrous glucose unit for every substituted sodium carboxymethyl group; 23 = molecular weight of sodium.

2.3.5. Determination of viscosity.

The viscosity of CMC was determined using the method documented by Aqualon [34]. The solution of 1%, 2%, and 3% CMC in distilled water were prepared. The measurements

were carried out at room temperature using a Brookfield Viscometer DV II model equipped with a number 7 spindle operating at 20-200 rpm.

3. Results and Discussion

The holocellulose was obtained in yellowish fibrous form. The holocellulose fibers were used for cellulose extraction without any purification and analysis. White color cellulose fibers were with cotton-like texture. Figure 2 demonstrates the appearance of OPEFB, holocellulose, and cellulose. The calculated CMC yield from cellulose ranged between 115.43~149.35% depending on NaOH concentration, reaction temperature, and amount of MCAA used. The color of the CMC produced was white to yellowish fibers (Figure 2(iv)). Table 1 showed the range of yields for holocellulose, cellulose, and CMC produced.

Table 1. Range of yields for holocellulose, cellulose, and CMC.

Product	Yield %
Holocellulose	66.41-77.19
Cellulose	46.96 - 50.44
CMC	115.43 - 149.35

All FTIR spectra of produced CMC are found comparable to standard CMC (*Sigma-Aldrich*). Figure 3 showed an overlay FTIR spectrum of one of the CMC produced with cellulose from EFB and standard CMC from Sigma-Aldrich. The frequency of the absorption bands of the representative spectra of cellulose and CMC obtained were similar, indicating that cellulose and CMC have a similar backbone and functional groups.

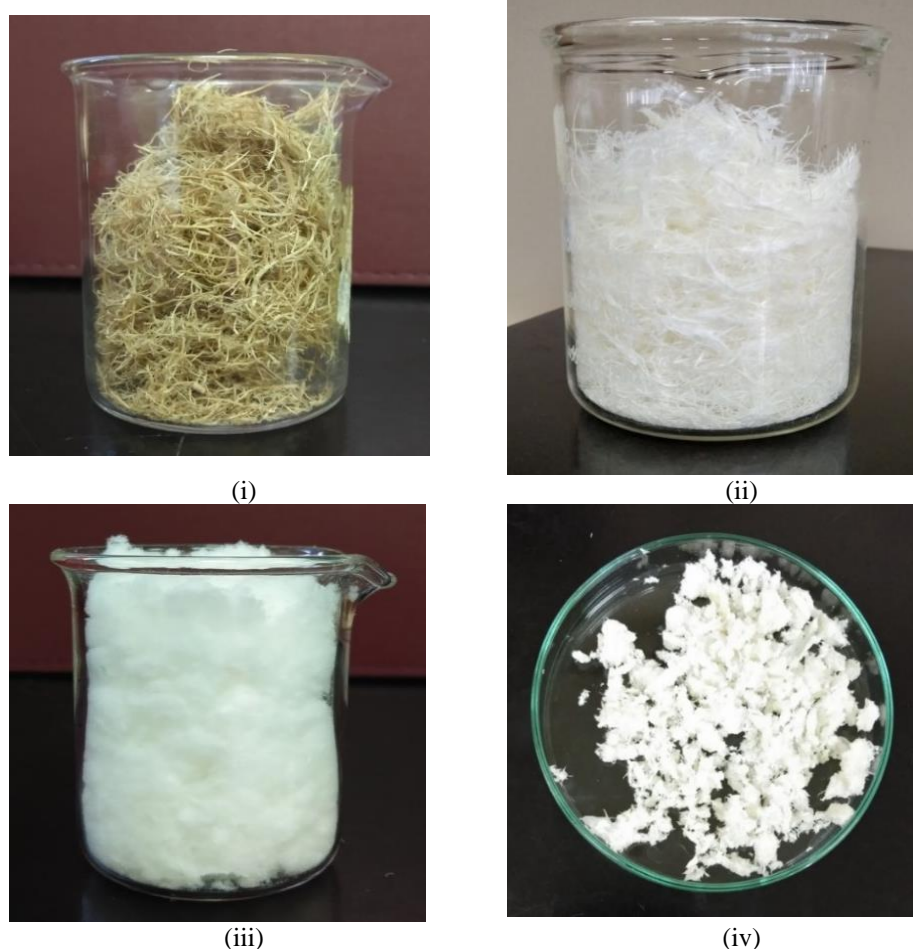


Figure 2. Physical form of oil palm empty fruit bunch, OPEFB (i), holocellulose (ii), cellulose (iii) and carboxymethyl cellulose (iv).

The broadband at 3200~3600 cm^{-1} is due to O-H stretching. The bands at 1450 and 1000~1200 cm^{-1} are due to $-\text{CH}_2$ scissoring and $-\text{O}-$ stretching, respectively. The band at 3000 cm^{-1} is due to a C-H stretching vibration, while the band around 1600 cm^{-1} is due to CO stretching. The presence of peak intensity at 1060 cm^{-1} demonstrated the presence of $-\text{CH}-\text{O}-\text{CH}_2$ (carboxymethyl group) stretching in the CMC [35].

The absorbent bands in the CMC- spectra were similar regardless of reaction parameters, but they differed from those of cellulose. The presence of a new and strong absorption band in CMC No 1 around 1580~1600 cm^{-1} confirms the stretching vibration of the carboxyl group (COO^-) and 1415~1419 cm^{-1} is assigned to carboxyl groups as it salts [16], [36]. According to reference [16], the carboxyl groups (COO^-) and their salts have a wave number of about 1600~1640 cm^{-1} and 1400~1450 cm^{-1} . A similar observation for the absorption bands has previously been reported [36], [37]. The spectrum of CMC results from different parameters was found similar.

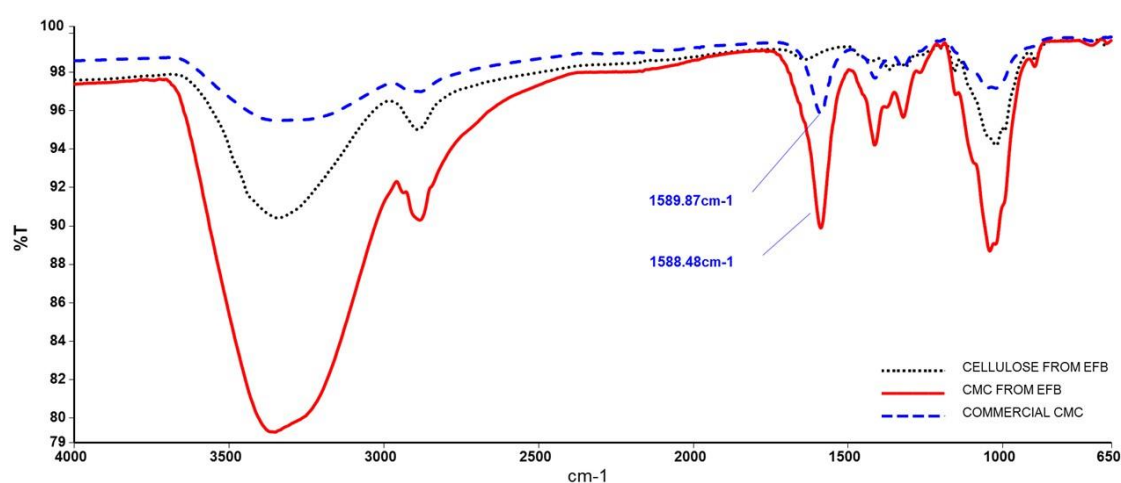


Figure 3. An overlay FTIR spectrum of CMC A with standard CMC and cellulose from EFB.

3.1. Properties of synthesized CMC.

3.1.1. Purity.

Purity refers to the percentage of pure CMC sodium salt in oven-dry products. There are four types or grades of CMC: high purity, mainly purified, semi-purified, and technical. Reference [31] points out that high purified CMC can be used in the food and pharmaceutical industry. CMC must be purified to a level of 99% for human consumption. Table 2 showed the purity of the CMC produced.

Table 2. Percentage purity of palm based carboxymethyl cellulose.

Properties	CMC A	CMC B	CMC C
Purity	99.05%	99.80%	99.62%

The purity of CMC produced using a low concentration of NaOH has lower purity than CMC produced in 25% and 35% of NaOH. This is due to the imperfection in alkalization and carboxymethylation processes that lead to NaCl and sodium glycolate formation.

The imperfection in alkalization and carboxymethylation processes will cause more NaOH and MCAA consumption for the formation of NaCl and sodium glycolate (Figure 4). The basic difference between highly pure and technical grade CMC is that the salts, chloride,

and sodium glycolate are washed out using large amounts of ethanol from the technical grade CMC, resulting in the formation of purified grade CMC.

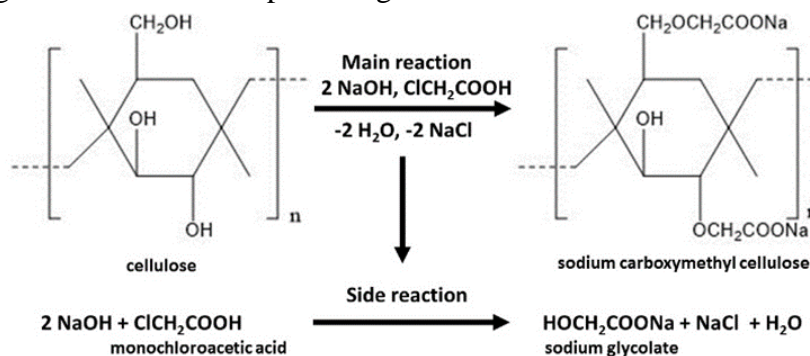


Figure 4. Side reaction, hydrolysis of MCAA occurs, where sodium glycolate and sodium chloride are formed.

3.1.2. Degree of substitution, DS.

The degree of substitution, DS of CMC can be determined based on the amount of carboxyl group or amount of sodium (Na) in CMC.

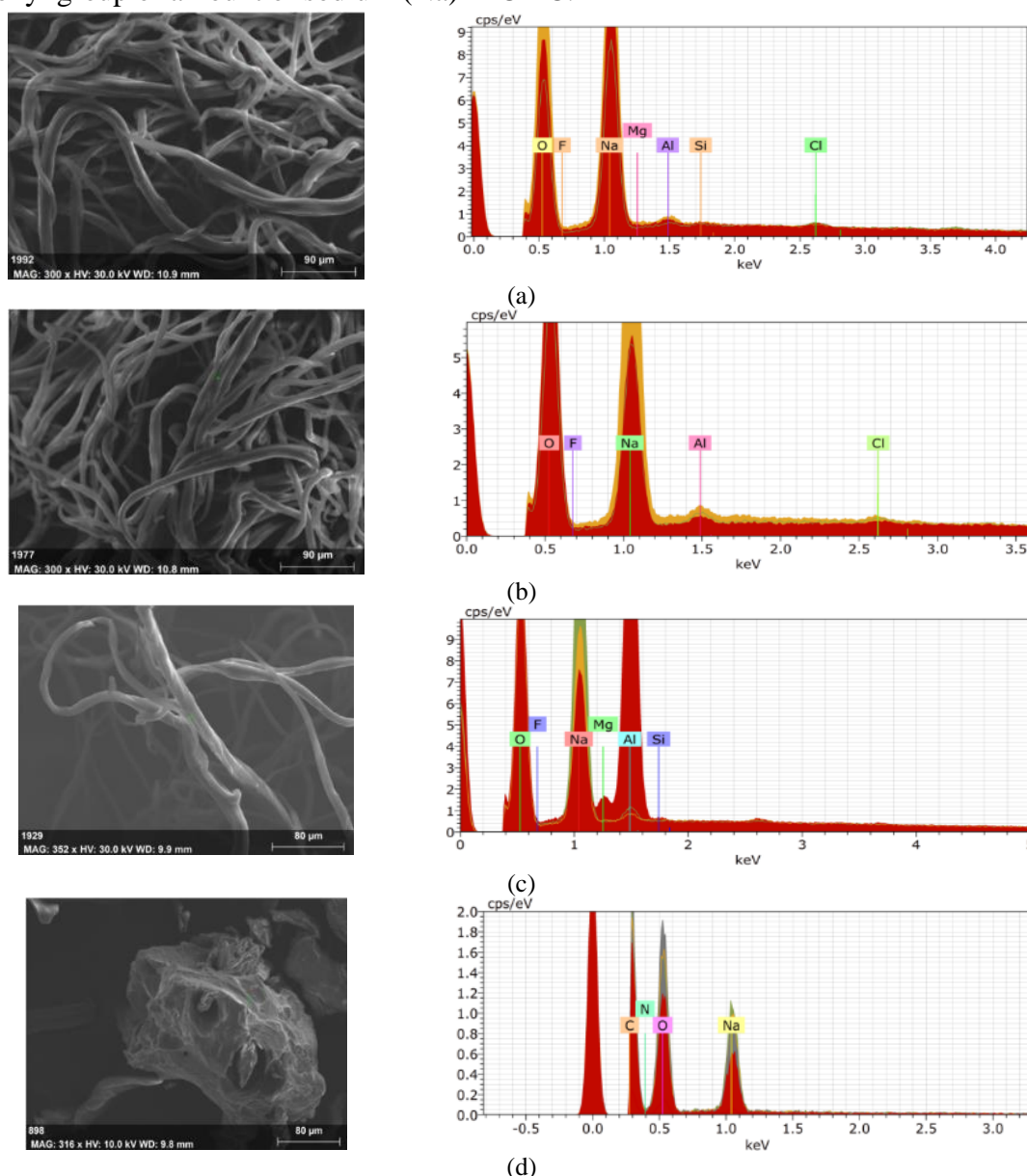


Figure 5. SEM images with EDX of (a) CMC A, (b) CMC B, (c) CMC C and (d) standard commercial CMC (300-352x magnification).

The DS value of CMC can be determined from both the IR spectra, Energy Dispersive X-Ray (EDX) [33], and potentiometric titration [32,38]. Researchers determined DS value using the calculation of sodium quantity from potentiometric titration, which has limitations to DS values below 0.85 and low-grade CMC. The DS value also can be determined using FTIR spectra based on the intensity of the peak of the carboxyl group, but the values obtained by FTIR spectra are the measurements of relative values to the cellulose.

In this work, DS was determined using Energy Dispersive X-Ray (EDX) analysis of scanning electron microscopy (SEM) images based on the percentage of sodium (Na) in the product. The micrograph of CMC A, CMC B, CMC C, and standard commercial CMC is shown in Figure 5. The calculated DS value is tabulated in Table 3.

DS is an important CMC parameter. DS gives improved compatibility with other soluble components such as salts and non-solvents. The palm-based CMC A recorded the highest calculated DS value of 1.3, followed by CMC B with 0.70 and 0.68 for CMC C.

Table 3: Degree of Substitution of Palm Based Carboxymethyl Cellulose by EDX Analysis.

Properties	Average % of Na (from EDX Analysis)	Calculated DS	DS value from CoA*
CMC A	11.24	1.30	-
CMC B	7.38	0.70	-
CMC C	7.23	0.68	-
Standard commercial CMC	8.91	0.91	0.82-0.95

*CoA = certificate of analysis

3.1.3. Viscosity.

The viscosity of CMC is an important parameter for industrial use. It provides information for flow characteristics of the fluid flow involved in processing operations and products using different CMC concentrations. The synthesized CMC are transparently soluble in distilled water solutions with up to 3 % dilution at 25°C with a spindle speed range from 20~200 rpm.

The results show that 1% dilution in distilled water, CMC B has the highest viscosity, 129~234 cP compared to CMC A, 127~225 cP, and the lowest, CMC C with 47~68 cP. However, at varying the produced CMC dilution in distilled water and temperature, the result shows that the synthesized CMC are produced in a wide range of viscosities (Table 4). This may be due to DS value as it gives improved compatibility and solubility in water.

It is well known that the viscosity of CMC is influenced by many factors, including temperature and CMC concentration [37], [39]-[41]. Since this CMC produced from OPEFB can reach high viscosity, which is higher than 1000 cP, this palm-based CMC has the potential to be used as a food thickener. Bono and co-workers [24] reported that the CMC produced from palm kernel cake is in the category of low viscosity, which is only 66.6 cP at 3% dilution in distilled water.

Table 4. Viscosity of palm-based carboxymethyl cellulose

Solution of CMC in distilled water	Unit	CMC A	CMC B	CMC C
at 25°C				
1% dilution	cP	127-225	129-234	47-68
2% dilution	cP	555-950	1019-1687	156-376
at 20°C				
1% dilution	cP	142-237	159-256	54-84
2% dilution	cP	915-1280	1124-1788	169-396

4. Conclusions

High-grade carboxymethyl cellulose was successfully synthesized using carboxymethylation of cellulose in various NaOH concentrations, reaction time, and MCA levels. The chemical configuration of cellulose and CMC were confirmed by the Fourier-transform infrared spectroscopy (FTIR) technique. CMC with high purity (CMC A, CMC B, and CMC C) can be used as an additive for the pharmaceutical and food industries. This palm-based CMC has a huge potential for future green-chemical demand since it is being produced from a renewable resource and sustainable.

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Conflicts of Interest

The authors declare no conflict of interest.

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