

Article

Sustainable Production of Carboxymethyl Cellulose: A Biopolymer Alternative from Sugarcane (*Saccharum officinarum* L.) Leaves

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Abstract: This study explores the potential of sugarcane (*Saccharum officinarum* L.) leaves (SCLs), a significant agricultural waste, for the sustainable production of carboxymethyl cellulose (CMC) utilizing an innovative approach of carboxymethylation with monochloroacetic acid and varying sodium hydroxide (NaOH) concentrations (ranging from 20 to 60 g/100 mL). The optimal carboxymethylation condition was identified as 40 g/100 mL NaOH, which yielded the highest degree of substitution (DS = 0.86). Furthermore, a higher ash content in the obtained CMC indicated significant carboxymethyl substitution within the structure. The chemical structure of cellulose and the resulting polymers were characterized using Fourier transform infrared spectroscopy (FTIR). The FTIR spectrum exhibited characteristic peaks of carboxymethyl groups and their salts at wavenumbers of 1588–1591 cm^{−1} and 1413–1415 cm^{−1}, respectively. The analyses from X-ray diffraction (XRD) and field emission scanning electron microscopy (FE-SEM) revealed detailed insights into the crystallinity and morphology of carboxymethyl cellulose (CMC). The levels of heavy metals, including arsenic, lead, cadmium, and mercury, in the purified CMC were assessed using inductively coupled plasma mass spectrometry (ICP-MS) and were found to be within the recommended WHO/FAO limits, except for lead, which exceeded permissible levels. The prepared CMC demonstrated high purity at 99.34%, surpassing the 98.00% purity requirement for commercial-grade CMC. This product exhibits strong potential for diverse industrial applications, including paper coating, textile sizing and printing, ceramic glazing, and various uses in the oil industry.

Keywords: agricultural waste; alpha cellulose; carboxymethyl cellulose; sugarcane leaves



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1. Introduction

The proliferation of undesired agricultural waste products and byproducts resulting from agricultural activities and agro-based processing poses a critical environmental challenge, thereby leading to the genesis of pollution. A considerable portion of these agricultural residues primarily comprises cellulose originating from the intricate composition of plant cell walls [1]. Cellulose is the most abundant polymer on earth, making it the most common organic compound [2,3]. The photosynthetic process yields considerable cellulose production, with an estimated annual output of approximately 830 million metric tons. Notably, cellulose constitutes 40% of the dry weight of crops, thus accounting for an annual production of around 200 million tons. Within the plant kingdom, cellulose content varies, with plants typically containing approximately 33%, wood comprising around 50%, and cotton boasting an impressively high cellulose content of 90% [2,4].

Cellulose, a linear and high-molecular-weight polymer comprising glucose monosaccharide units interconnected through beta acetal linkages, exhibits unique characteristics due to the presence of hydrogen bonding in its molecular structure. So, cellulose displays

limited solubility and resistance to melting in common solvents. These attributes render it less relevant for numerous industrial applications, with some notable exceptions including its usage in the construction of wood-based structures; the production of various paper products; as raw material for fabricating cotton, linen, and rayon textiles; incorporation in nitrocellulose explosives, and the synthesis of cellulose acetate films [5]; as well as use in food industries [6]. Cellulose's inherent susceptibility to chemical derivatization enables its transformation into valuable chemical feedstock. Notably, etherification stands out as a pivotal pathway in the cellulose derivatization process [5,7,8]. The conversion of cellulose into high-value cellulose esters and ethers has garnered substantial attention within the realms of green and sustainable chemistry, thereby fostering the advancement of environmentally friendly technologies [9].

Carboxymethyl cellulose (CMC) is an anionic, water soluble derivative of cellulose, which is a linear polysaccharide composed of an anhydro glucose unit linked by β -1,4-glycosidic bonds. At the molecular level, the major difference between CMC and cellulose is only some anionic carboxymethyl groups (i.e., $-\text{CH}_2\text{COOH}$) in the CMC structure that replace the hydrogen atoms from some hydroxyl groups present in the pristine cellulose infrastructure [10]. CMC, a derivative of cellulose, is environmentally friendly due to its nontoxicity, biodegradability, and diverse uses. As a result, it has been used in numerous industrial applications, including paper, textile, pharmaceutical, food exploration, paint industries, biomedical, construction, plastics, cosmetics, and oil industries [5,10].

In the customary production of CMC, a two-step reaction process is employed involving alkalization and etherification. Initially, cellulose is activated by NaOH in an organic solvent, such as isopropanol (IPA), ethanol, or methanol, in a reaction depicted as shown in Equation (1). Subsequently, cellulose is subjected to etherification at hydroxyl groups using monochloroacetic acid (MCA) to yield CMC, as depicted in Equation (2) and Figure 1. The tandem of these two reactions is collectively referred to as the carboxymethylation reaction. Nevertheless, an additional side reaction, as depicted in Equation (3), may occur between the surplus NaOH and MCA [1–3,10,11].

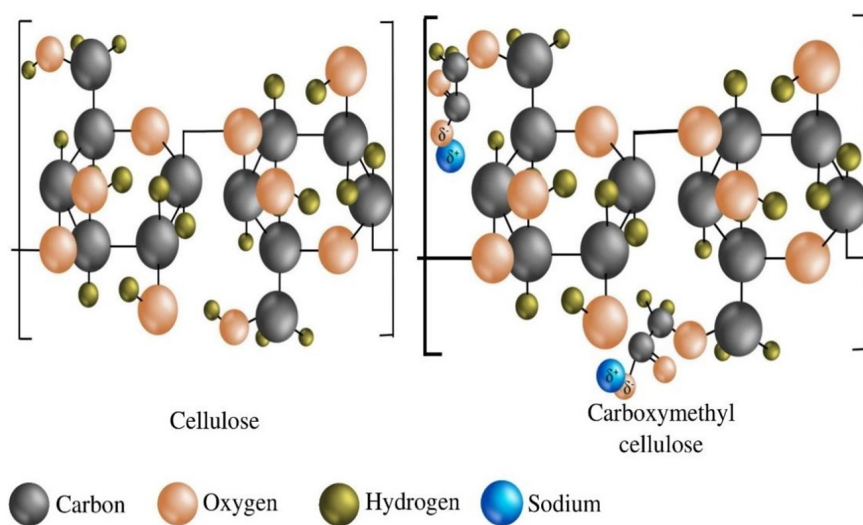
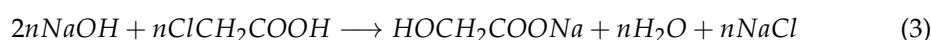
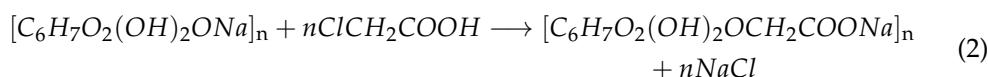
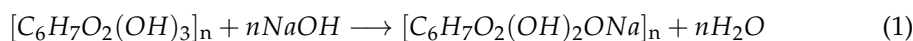


Figure 1. The fundamental structural difference between cellulose and carboxymethyl cellulose (CMC) [10].

The properties of cellulose derivatives are primarily evaluated based on their DS value. The DS value is technically defined as the average number of substitutes of the reactive group by other active molecules in the polymer chain. During the synthesis of CMC from cellulose, the value of DS is determined by the number of substituent groups (carboxymethyl) attached to each anhydro glucose unit [10,12]. A maximum DS of three can be achieved [13,14]. In commercial CMC, the most common DS obtained is usually lower which is from 0.4 to 1.4 [12]. The DS of CMC is dependent upon the reagent's concentration, reaction time, solvent, and temperature [12,15].

Many CMC derivatives have been prepared through carboxymethylation reactions using diverse substances (i.e., corn husk [1,16], mimosa pigra [3], corn cob [4], pistachio shells [17], sugarcane straw [9], cashew tree [12], brewer's spent grain [7], sugar beet pulp [8], durian rind [18,19], cavendish banana pseudostem [20], lantana camara [21], sago palm [22], palm bunch [23,24], bagasse [23], palm kernel cake [25], and papaya peel [26,27]), as well as some waste materials (i.e., office wastepaper [2,11], carbon foam [28], and domestic matchstick waste [29]). These uses have attracted research interest for CMC production. However, to date and to the best of our knowledge, there has been no prior research on the production of CMC from sugarcane (*Saccharum officinarum* L.) leaves (SCLs).

Thailand, a predominantly agricultural nation with nearly half of its land area dedicated to cultivation, generates substantial agrarian residues or wastes. This encompassing array of agricultural byproducts comprises rice straws, rice husks, cane leaves and tops, bagasse, corn husks and stalks, cassava roots, palm trunks, palm leaves and branches, cashew nut shells, and other materials. Over the years, extensive efforts have been made to repurpose these waste materials, including their conversion into animal feeds, fertilizers, or bio-based energy sources [30]. Nevertheless, certain agricultural wastes remain challenging to employ in these conventional ways, thus leading to their disposal in landfills or incineration and resulting in deleterious environmental consequences, particularly air pollution. In 2018, an excess of 159.90 million tons of agricultural wastes remained unutilized even after secondary attempts at recycling [31]. SCLs represent a surplus biomass that remains in the fields following the sugar cane harvesting process, with an excess of 45.19 million tons remaining unutilized [31].

SCLs have been recognized for their substantial cellulose content in previous reports [32–34]. The isolation of cellulose materials from sugarcane leaves (SCLs) facilitates the synthesis of carboxymethyl cellulose (CMC) through etherification by utilizing an alkaline medium and an etherifying agent. This research aims to investigate the impact of varying sodium hydroxide (NaOH) concentrations on the synthesis process. The concentration of NaOH is crucial in etherification, thus significantly influencing the degree of substitution (DS), yield, purity, and the physicochemical properties of the resulting CMC. The study focuses on evaluating the properties of CMC derived from SCLs, including assessments of DS, yield percentage, purity, sodium chloride content, ash content, chemical structure, and crystallinity.

2. Materials and Methods

2.1. Materials

Sugarcane leaves (SCLs) were procured from sugarcane fields situated in the Satuek district (15.11° N, 103.33° E) of Buriram, Thailand for the purpose of this study. Currently, Thailand has a total sugarcane cultivation area of 17,635,756,800 m². In the province of Buriram, the entire cultivated area for sugarcane amounts to 208,504,000 m², which represents 1.18% of the nation's total sugarcane cultivation area. Within Buriram province, Satuek district is recognized as an area with a sugarcane cultivation area of 29,123,200 m², thus accounting for 13.97% of the entire sugarcane cultivation area in the province of Buriram [35]. The SCLs, amounting to an excess of 0.074 million tons, are left in the fields after the sugarcane harvesting process in Satuek district, Buriram, Thailand, without being utilized. The chemicals utilized in the experimental procedures included isopropyl alcohol, methyl alcohol, acetic acid, ethyl alcohol, monochloroacetic acid, sulfuric acid, hexyl alcohol,

ammonium oxalate, sodium metabisulphite, sodium hydroxide, and sodium chlorite, all of which were acquired from Italmar, Thailand.

2.2. Preparation of Sample

The collected SCL samples underwent a rigorous washing process with water and were subsequently air-dried under ambient conditions over a period of seven consecutive days. Following this, the samples were sectioned into small pieces and subjected to further drying at a temperature of 105 °C for a duration of 6 h. The resulting dried material was then finely powdered in a pin mill and subsequently sieved through a 70-mesh screen (size, 0.210 mm) to ensure uniform particle size distribution [18]. The finely powdered SCL sample was carefully stored in an airtight zip lock polybag at room temperature, minimizing moisture absorption, and preserved for subsequent characterization and experimental investigations.

2.3. Isolation of α -Cellulose

A measured quantity of powdered SCL sample was treated with an 18% NaOH solution for a duration of 2 h, with intermittent stirring to maintain a solid-to-liquor ratio of 1:100 at room temperature [16]. Following the reaction, the cellulose residue was separated via filtration and subsequently subjected to thorough washing with 2% acetic acid, followed by rinsing with hot distilled water. Thereafter, the cellulose was stirred with n-hexane for 1 h at room temperature, followed by additional washing steps employing 95% ethanol. Following the initial filtration, the obtained cellulose was subjected to heating with a 0.7% NaClO₂ solution buffered at pH 4 in a solid-to-liquor ratio of 1:50 at a temperature range of 90–95 °C for a duration of 90 min. Subsequently, the α -cellulose was separated via filtration and underwent a series of washing steps with ethanol and distilled water. The isolated α -cellulose was then treated with a 0.2% sodium metabisulphite solution for 15 min, followed by additional filtration and thorough washing with distilled water. Finally, the α -cellulose was dried at 60 °C until reaching a constant weight [36]. The percentage yield of the alpha cellulose products was calculated using Equation (4).

$$\text{Yield of alpha cellulose (\%)} = \frac{W_1}{W_0} \times 100\% \quad (4)$$

where

W_1 is the weight of alpha cellulose.

W_0 is the weight of powdered SCLs.

2.4. Synthesis of Carboxymethyl Cellulose

In this study, carboxymethyl cellulose (CMC) was synthesized using cellulose derived from SCLs and various sodium hydroxide (NaOH) concentrations (20%, 30%, 40%, 50%, and 60% *w/v* of NaOH). The initial step in the CMC synthesis involved transferring 15.0 g of cellulose powder to 50 mL of sodium hydroxide at different concentrations (20%, 30%, 40%, 50%, and 60% *w/v* of NaOH) and stirring for 30 min. In the subsequent step, the solution was covered with aluminum foil and heated to a reaction temperature of 55 °C in an oven for 3.5 h. After heating, the solution separated into two phases: a liquid phase, which was removed, and a solid phase, which was suspended in 100 mL of methanol (70% *v/v*). The mixture was then neutralized with glacial acetic acid and filtered using a Buchner funnel. To eliminate undesirable byproducts, the solid was washed five times with ethanol (70% *v/v*) by suspending it in 300 mL for 10 min. Finally, it was washed with 300 mL of absolute methanol and dried in an oven at 55 °C overnight [18,23]. The percentage yield of the CMC products was calculated using Equation (5).

$$\text{Yield of CMC (\%)} = \frac{W_1}{W_0} \times 100\% \quad (5)$$

where

W_1 is the weight of CMC.

W_0 is the weight of alpha cellulose.

2.5. Characterization

2.5.1. Degree of Substitution (DS)

The DS for CMC was determined following the D1439-03 Standard Test Methods for Sodium Carboxymethyl Cellulose. Initially, 5 g of CMC was introduced into a 200 mL solution of HNO₃–methanol (1:1 *v/v*). This solution was agitated and left undisturbed for 3 h. Subsequently, the solution was rinsed with a 70% methanol solution (*v/v*) and dried in an oven until a constant weight was achieved. Next, 2 g of the dried sample was dissolved in 200 mL of distilled water and 30 mL of 1 N NaOH. The solution was then titrated with excess NaOH using 1 N HCl to a phenolphthalein endpoint. Titration was considered complete when the color transitioned from dark pink to colorless [8,9,37]. The DS was determined using Equations (6) and (7).

$$\text{Degree of substitution} = \frac{0.162A}{(1 - 0.0584A)} \quad (6)$$

$$A = \frac{(BC - DE)}{F} \quad (7)$$

where

A is milliequivalents of acid consumed per gram of sample.

B is NaOH solution added (mL).

C is normality of the NaOH solution (N).

D is HCl required for titration of the excess NaOH (mL).

E is normality of the HCl (N).

F is acid carboxymethyl cellulose used (g).

The value 162 is gram molecular mass of the anhydro glucose unit of cellulose.

The value 584 is net increase in molecular mass of anhydro glucose unit for each carboxymethyl group substituted.

2.5.2. Purity

A quantity of 1.5 g of CMC was introduced into 100 mL of 80% methanol (*v/v*). This solution was subjected to stirring and allowed to stand for 10 min. Subsequently, the solution was filtered and rinsed with 100 mL of 80% methanol (*v/v*). To achieve a state of pure CMC, the sample was then dried at 100 °C until a consistent weight was attained [8,9]. The percentage of CMC purity was calculated using Equation (8).

$$\text{Percentage of CMC purity (\%)} = \frac{W_1}{W_0} \times 100\% \quad (8)$$

where

W₁ is the final weight.

W₀ is the initial weight.

2.5.3. Sodium Chloride Content

A quantity of 2.0 g of CMC was placed into 250 mL of a 65% methanol solution (*v/v*). This solution was allowed to stand for 5 h, after which it separated into two distinct phases. A volume of 100 mL from the liquid phase was neutralized with diluted HNO₃ and subjected to titration using a 0.1 N AgNO₃ solution until a K₂CrO₄ endpoint was reached. The titration was considered complete when the color transitioned from red to orange brick [8]. The percentage of NaCl content in CMC was determined using Equation (9).

$$\text{NaCl (\%)} = \frac{1.461 \times V}{m} \quad (9)$$

where

V is the amount of AgNO₃ (mL).

m is the weight of dried sample (g).

2.5.4. Ash Content

The ash content of CMC was assessed in accordance with the D7582-15 Standard Test Methods for Proximate Analysis of Coal and Coke by Macro Thermogravimetric Analysis (Mettler Toledo, TGA/DSC 3+, Greifensee, Switzerland) [38]. The ash yield represents the residue that remains after subjecting the samples to heating.

2.5.5. Inductively Coupled Plasma Mass Spectrometer (ICP-MS)

The ICP-MS instrument utilized in this study was the NexION 350X provided by PerkinElmer, Waltham, MA, USA. Data analysis and instrument control were performed using Syngistix™ for ICP-MS version 1.0. Sample preparation involved microwave digestion, which is a technique facilitated by the Titan MPS instrument from PerkinElmer, Rodgau, Germany. In this process, microwave energy was used to break down the sample matrix and convert the elements of interest into their atomic form. Subsequently, the ions generated were introduced into the ICP-MS system, separated by their mass-to-charge ratio, and quantified [39].

2.5.6. Fourier Transform Infrared Spectroscopy (FTIR)

The functional groups of both α -cellulose and carboxymethylcellulose were ascertained using an infrared spectrophotometer employing the KBr disc technique. This analysis was conducted utilizing a Nicolet iS50 model instrument from Thermo Scientific, Waltham, MA, USA, and the OMNIC software (version 9.12.1019). Pellets were prepared by grinding 0.2 mg of α -cellulose and carboxymethylcellulose with 2 mg of KBr. Transmission measurements were performed across the wavenumber range of 4000–400 cm^{-1} .

2.5.7. X-ray Diffractometry (XRD)

The X-ray diffraction patterns, used to assess the crystallinity of cellulose and CMC, were obtained using an X-ray diffractometer (Bruker AXS Model D8 Advance, Mannheim, Germany). The scattering angle (2θ) was scanned from 5° to 80° at a rate of 0.02° per step, with a duration of 0.2 s per step. The powder samples were exposed to X-ray beams with settings at 40 kV and 40 mA.

2.5.8. Field Emission Scanning Electron Microscopy (FE-SEM)

Morphological investigation of the cellulose and CMC samples was performed using field emission scanning electron microscopy or FESEM (JEOL, JSM 7800F, Tokyo, Japan). The samples were adhered to carbon tape containing a specimen stub and sputter-coated with a thin layer of gold using a sputter coater (QUORUM Q150R ES, Laughton, UK). The observation was run on a voltage of 2 kV with $500\times$ magnification.

3. Results and Discussion

The compositional data of the SCLs sample, serving as the raw material, are meticulously presented within Table 1. It is noteworthy that Table 1 illustrates the α -cellulose extracted, thereby quantifying 41.71 g per 100 g of dry biomass. These findings closely align with outcomes from similar studies, wherein corn husks were ascertained to harbor an α -cellulose content of 45.13 g per 100 g dry biomass [1]. Additionally, investigations conducted by Suriyatem et al. [23] unveiled the α -cellulose content of sugarcane bagasse to register at 42.5 g per 100 g of biomass. The diverse chemical composition evident in the original wood samples, even when derived from the same part and tree type, is attributed to an interplay of soil conditions, geographic locale, and prevailing climate factors [23].

Table 1. The α -cellulose content of the sample.

α -Cellulose	Weight of Powder (g)	Weight of α -Cellulose (g)	Yield of α -Cellulose (%)	Maximum Weight (g)	Minimum Weight (g)	Standard Deviation
SCLs	10.00	4.1706	41.71	4.6402	3.7468	0.2204

The α -cellulose content refers to the portion of cellulosic materials that remains insoluble in a solution of sodium hydroxide of mercerizing strength under specific conditions and for a defined duration [40]. Through this process, certain components and impurities, such as wax, hemicellulose, and lignin, are dissolved, and subsequent filtration and washing steps yield a refined form of cellulose known as α -cellulose [41]. This α -cellulose is a white, odorless, and tasteless powder with insolubility in water. Widely utilized as a raw material, α -cellulose finds applications in the manufacturing of propellants, paper, paperboards, fabrics, electrical cable insulators, cellulose derivatives, and various other products [42]. The visual characteristics of α -cellulose and CMC obtained from SCLs are documented in Table 2.

Table 2. Appearance of raw material, α -cellulose, and CMC from SCLs in different conditions.

Sample	Isolation of α -Cellulose	CMCSCL Synthesis with Various NaOH Concentrations (w/v)				
		20% NaOH	30% NaOH	40% NaOH	50% NaOH	60% NaOH
SCLs	α -cellulose					

3.1. Degree of Substitution (DS) and Percent Yield of CMC_{SCL}

The DS represents a critical property of CMC, thus significantly impacting not only its solubility but also its solution characteristics [43]. DS values falling within the range of 0.0 to 0.4 indicate that the CMC is insoluble but swellable, whereas values exceeding this range signify that the CMC is completely water soluble [23]. CMC derived from cellulose through alkalization and subsequent carboxymethylation employing monochloroacetic acid (MCA) generally falls within the range of 0.4–1.3 [15,18,20]. The DS values obtained in this study were well within the expected range, thus spanning from 0.46 to 0.86 (Figure 2).

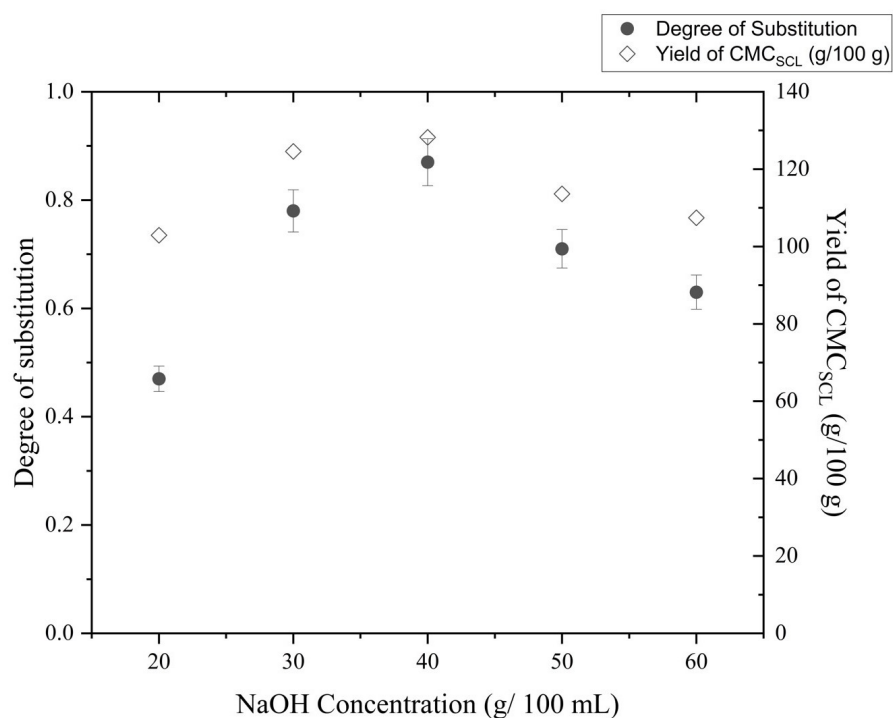


Figure 2. Effect of NaOH concentration on degree of substitution and percent yield of CMC_{SCL} synthesized with various NaOH concentrations.

The degree of substitution (DS) values exhibited an increase with rising NaOH concentrations (20–40 g/100 mL), thus reaching a peak DS of 0.86 at 40 g/100 mL NaOH, followed by a significant decline. This behavior can be elucidated through the carboxymethylation process, wherein two competitive reactions occur simultaneously. The initial reaction involves the interaction of cellulose hydroxyl groups with monochloroacetic acid (MCA), thereby yielding CMC_{SCL}, as demonstrated in Equation (2). The subsequent reaction, governed by NaOH and MCA, leads to the formation of sodium glycolate as a byproduct, as outlined in Equation (3).

The second reaction surpasses the first at higher alkaline concentrations. Excessive alkalinity leads to a predominant side reaction, thereby resulting in the formation of sodium glycolate as a byproduct and subsequently reducing the DS. Furthermore, the decrease in the DS observed at the elevated 40% NaOH concentration may also be attributed to the potential chain degradation of CMC polymers [3,4,44].

A similar trend has been noted in CMC derived from various cellulose sources, including corn cobs [4], sugar beet pulp [8], durian rind [18], cavendish banana pseudostems [20], lantana camara [21], and sago palm [22]. The DS values of CMC obtained from different cellulose sources are summarized in Table 3. Furthermore, the percentage yield of the CMC_{SCL} exhibited a trend related to NaOH concentration that was similar to the DS results (Figure 2). This aligns with findings from previous studies [3,18,20].

Table 3. The DS values of CMC obtained from different cellulose sources.

Sources of Cellulose	Degree of Substitution of CMC	Reference
Corn cobs	0.48–1.18	[4]
Sugar beet pulp	0.102–0.667	[8]
Durian rind	0.56–0.87	[18]
Cavendish banana pseudostem	0.26–0.75	[20]
Lantana camara	0.59–1.22	[21]
Sago palm	0.328–0.821	[22]
Sugarcane leaves (SCLs)	0.46–0.86	This work

The DS in CMC is a crucial factor determining its properties and applicability in specific industrial contexts. Generally, as the DS increases, the water solubility of CMC improves, thus influencing its viscosity and dissolution kinetics [4,45]. This characteristic is particularly significant in applications requiring rapid dissolution, such as in pharmaceutical formulations or food products, where CMC serves as a widely used thickener and stabilizer in the food industry [46]. For food applications, DS typically falls within the range of 0.6–0.95 [47]. CMC with a high DS can enhance the electrostatic repulsion between protein particles, thus preventing phase separation in acidified milk drinks [46]. Additionally, the film-forming ability of CMC is influenced by the DS, with higher values contributing to improved film formation, thus making it suitable for applications in coatings and packaging materials [48].

3.2. Purity, NaCl, and Ash Contents Analysis

The purity of the CMC was assessed following multiple washes with 80% methanol, thereby effectively eliminating reaction byproducts such as sodium chloride and sodium glycolate. For CMC intended for human consumption, purification to a minimum level of 99.5% is essential. In contrast, CMC designated for commercial use in industries such as paper coating, textile sizing and printing, ceramic glazing, and oil drilling muds must meet a slightly lower purity threshold of 98.0%.

The CMC content and sodium chloride percentages in the prepared CMC were meticulously analyzed and then compared to the established standards for commercial and pharmaceutical grades of CMC. The results have been succinctly summarized in Table 4. Upon examination of the data in Table 4, it is evident that the CMC content achieved in this study fell within the anticipated range of 98.81–99.34%. While it is slightly below the

99.5% purity benchmark set for CMC standards in the food and pharmaceutical industry, it comfortably surpasses the requisite 98.0% purity level for standard CMC in commercial applications. Moreover, the sodium chloride content in the prepared CMC was found to be remarkably low at just 0.04%. Notably, the sodium salt content in food additives should not exceed 0.5%, either individually or in combination, thereby underscoring the suitability of the prepared CMC for use in various applications.

Table 4. Determination of CMC, NaCl, and ash contents.

Parameters	Content of CMC or Purity, %	Content of NaCl, %	Content of Ash, %	Reference
CMC _{SCL} (20% <i>w/v</i> of NaOH)	98.81	0.04	11.46	This work
CMC _{SCL} (30% <i>w/v</i> of NaOH)	99.15	0.04	11.22	This work
CMC _{SCL} (40% <i>w/v</i> of NaOH)	99.34	0.04	14.40	This work
CMC _{SCL} (50% <i>w/v</i> of NaOH)	99.14	0.04	10.62	This work
CMC _{SCL} (60% <i>w/v</i> of NaOH)	>99.50	<0.5 (sodium salts)	-	This work
CMC food and pharmaceutical grade	99.18	0.04	9.79	[14,16,49–51]
CMC commercial grade	>98.00	<2.0 (sodium salts)	-	[43,49]

The results concerning the ash content of the prepared CMC are presented in Table 4. Notably, the ash content achieved in this study consistently fell within the expected range of 9.79–14.40%. The maximum ash content, 14.40%, was observed when utilizing a NaOH concentration of 40 g/100 mL. It is intriguing to observe that these results closely align with findings from analogous studies, where standard CMC samples were reported to contain an ash content of 14.23% [16]. This higher ash content suggests a greater degree of substitution (DS) in the CMC. It is conceivable that this phenomenon occurs because a higher number of hydroxyl groups are substituted by the sodium salts of carboxymethyl groups during the etherification reaction [11,16].

3.3. Heavy Metals of CMC_{SCL}

Heavy metals are potential environmental contaminants, with the capability to cause human health problems due to their toxic effects, even at very low concentrations [16]. The toxic metal content in the prepared CMC_{SCL} was determined and compared with the recommended values shown in Table 5. From the results in Table 5, it can be observed that the concentrations of As, Cd, and Hg were <0.04, <0.04, and <0.04 ppm, respectively. These values are within the World Health Organization's (WHO) permissible limits for food additives and emulsifiers.

Table 5. Concentration of heavy metals in the synthesized CMC_{SCL}.

Heavy Metal	As	Cd	Hg	Pb
Concentration in ppm (SCLs)	-	-	-	<50.00
Concentration in ppm (CMC _{SCL})	<0.04	<0.04	<0.04	3.64
Concentration in ppm (proposed permissible limit) [50–52]	3	1	1	1

Conversely, the concentration of Pb in CMC_{SCL} was 3.64 ppm, thus exceeding the permissible limits for food additives and emulsifiers set by the World Health Organization (WHO). When we consider the concentration of Pb in SCLs, which is <50 ppm, it is approximately 50 times the permissible limit. This underscores that during the processes of the isolation of α -cellulose and the synthesis of carboxymethyl cellulose, specifically through the alkalization and etherification reactions, the concentration of Pb in CMC_{SCL}

decreases. The presence of Pb is a result of its deposition through the application of herbicides during the cultivation process [53].

3.4. FTIR Analysis

The identification of the produced α -cellulose and CMC_{SCL} was conducted through Fourier Transform Infrared (FTIR) spectroscopy. The FTIR spectra of the synthesized CMC samples and α -cellulose are depicted in Figure 3. As observed in Figure 3, the spectra exhibited characteristic absorptions related to the cellulose backbone and the presence of the carboxymethyl ether group, notably at 1588–1591 cm^{-1} .

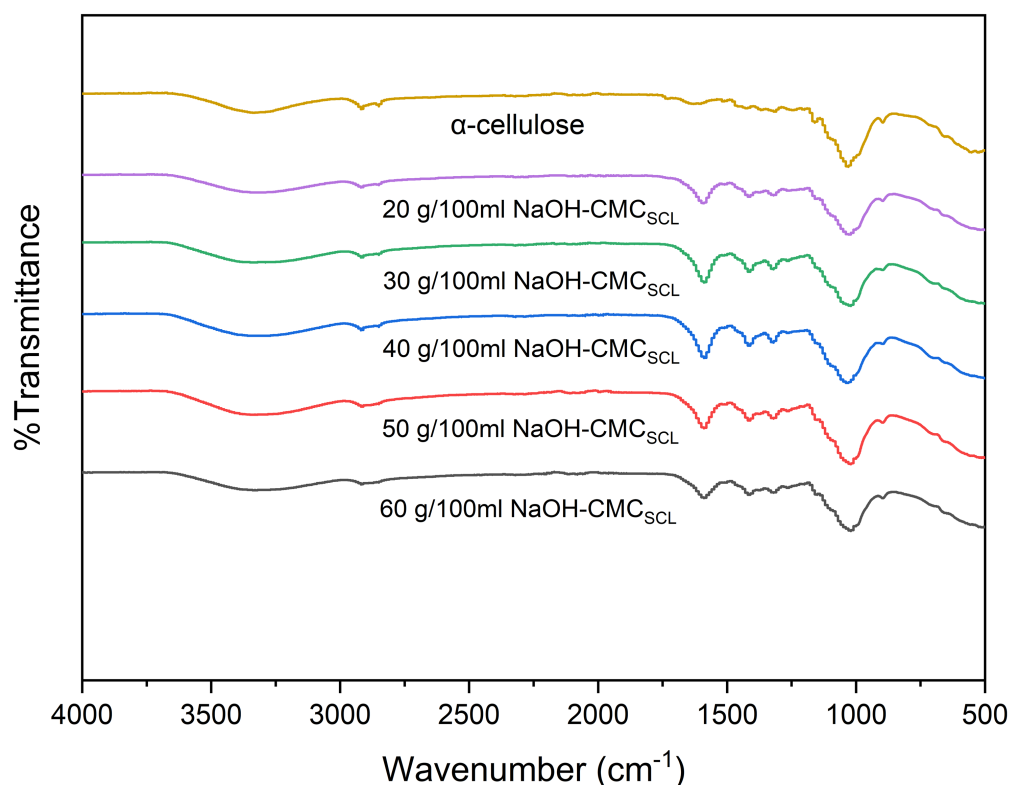


Figure 3. FTIR spectra of α -cellulose and CMC_{SCL} synthesized with various NaOH concentrations.

The broad band within the range of 3200–3600 cm^{-1} corresponds to O-H stretching, while the band between 2800 and 3100 cm^{-1} is attributed to C-H stretching vibrations. The band at approximately 1600 cm^{-1} represents C=O stretching, and the bands around 1420 and 1320 cm^{-1} are associated with $-\text{CH}_2$ scissoring and $-\text{OH}$ bending vibrations, respectively. The band spanning from 1000 to 1200 cm^{-1} is linked to $-\text{O}-$ stretching, while the band at around 1030 cm^{-1} corresponds to $-\text{CH}-\text{O}-\text{CH}_2$ stretching. Additionally, a wavelength near 890 cm^{-1} is indicative of the 1,4- β glycoside linkage of cellulose [1,5,18,22,23,54].

The absorbance bands in the CMC_{SCL} spectra exhibited consistency across different NaOH concentrations but displayed variations from those of α -cellulose. Notably, the emergence of a new and robust absorption band at 1588–1591 cm^{-1} provides clear evidence of stretching vibrations associated with the carboxyl group (COO^-), whereas the band at 1413–1415 cm^{-1} can be attributed to carboxyl groups in the sample salts [1,18,20]. A comprehensive dataset of FTIR spectra for the α -cellulose and CMC_{SCL}, synthesized with varying NaOH concentrations, are presented in Table 6. These results substantiate the occurrence of carboxymethylation within the cellulose molecules, thus aligning with prior findings [1,3,4,18,20,22,23].

Table 6. FTIR spectra data of α -cellulose and CMC_{SCL} synthesized with various NaOH concentrations.

Wave Number (cm ⁻¹)						Assignment
α -Cellulose	CMC _{SCL} (20% <i>w/v</i> of NaOH)	CMC _{SCL} (30% <i>w/v</i> of NaOH)	CMC _{SCL} (40% <i>w/v</i> of NaOH)	CMC _{SCL} (50% <i>w/v</i> of NaOH)	CMC _{SCL} (60% <i>w/v</i> of NaOH)	
3333.55	3332.01	3331.27	3331.72	3334.77	3334.22	OH stretching
2916.97	2917.63	2917.27	2917.38	2916.84	2916.86	CH stretching CH ₂ and CH ₃ groups
-	1591.93	1588.4	1588.12	1589.31	1589.04	C=O region (indicated CMC)
-	1415.46	1414.95	1413.48	1414.24	1413.23	CH ₂ bonding (indicated CMC)
1316.24	1320.27	1321.95	1322.41	1320.89	1320.38	OH in plane bending
1100.96	1096.9	1102.98	1099.1	1090.83	1098.93	C-O-C asymmetry bridge stretching
1031.26	1026.71	1020.39	1034.35	1020.05	1020.17	C-O stretching
896.51	896.34	896.34	896.51	902.42	896.34	β -Glycoside linkage

3.5. XRD Analysis

X-ray diffraction (XRD) analysis serves as a pivotal method for determining the crystallinity levels in polymers, with cellulose characterized by its inherent semicrystalline nature [16]. This study presents XRD patterns for α -cellulose and carboxymethyl cellulose sodium salt (CMC_{SCL}) synthesized at varying sodium hydroxide (NaOH) concentrations, as depicted in Figure 4. The crystallinity index (CI) was computed based on the ratio between the intensity of the 002 peak (I_{002}) and the minimum intensity (I_{AM}) observed between the 002 and 101 peaks [55]. Notably, the XRD patterns revealed prominent crystalline peaks at diffraction angles (2θ) of 15.89° (101), 18.00° (I_{AM}), and 22.05° (I_{002}). The calculated CI values for α -cellulose and CMC_{SCL}, across a range of NaOH concentrations from 20% to 60% *w/v*, were found to be 67.62%, 50.50%, 44.39%, 45.03%, 49.92%, and 43.37%, respectively. These findings indicate a decline in CI for CMC_{SCL} synthesized with increasing NaOH concentrations, thus suggesting a decrease in crystallinity. This observed reduction in crystallinity can be attributed to the disruption of hydrogen bonds during NaOH treatment [11,20], which is consistent with previous studies reporting similar effects in carboxymethyl cellulose (CMC) derived from various cellulose sources such as durian rind [18], corn husk [1], and brewer's spent grain [7], all of which demonstrated diminished crystallinity following alkaline processing.

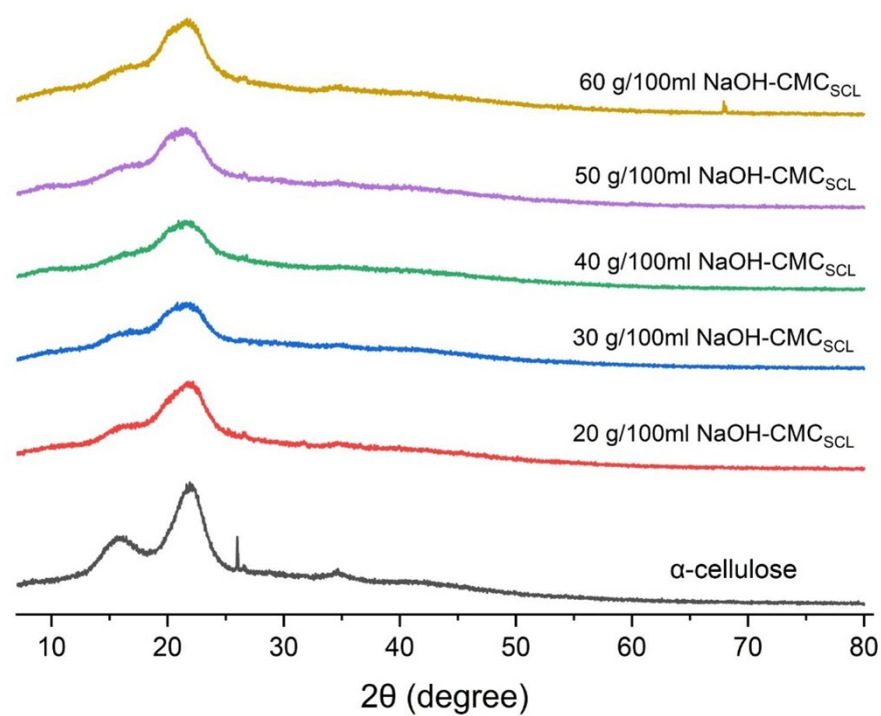


Figure 4. XRD patterns of α -cellulose and CMC_{SCL} synthesized with various NaOH concentrations.

3.6. Morphology of α -Cellulose and CMC_{SCL} Analysis

Field emission scanning electron microscopy (FE-SEM) is a sophisticated technique employed for imaging the microstructure of materials. In this study, FE-SEM analysis was utilized to assess the morphologies of α -cellulose and CMC_{SCL} synthesized with various NaOH concentrations. Table 7 presents the morphological characteristics of the α -cellulose and CMC_{SCL} synthesized using different NaOH concentrations. After alkali treatment, the surface of the SCL fibers became rougher, with diameters ranging from 80 to 150 μm . Prior to cellulose extraction, the fiber surfaces appeared to be covered by substances, possibly lignin and hemicellulose encrusting the cellulose within [23]. Following extraction with varying NaOH concentrations (20–60% *w/v* of NaOH), the FE-SEM images of the SCL cellulose demonstrated the removal of these encrusting substances from the fiber surfaces. In the case of the CMC_{SCL} synthesized with different NaOH concentrations, the resulting molecules exhibited a rod-like or ribbon-shaped structure, thereby suggesting the presence of some degree of crystallinity within the material. This structure aligns with images of typical CMC molecules as reported in previous studies [16,29].

Table 7. Morphological characteristics of α -cellulose and CMC_{SCL} synthesized using different NaOH concentrations.

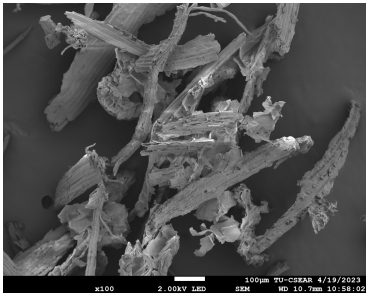
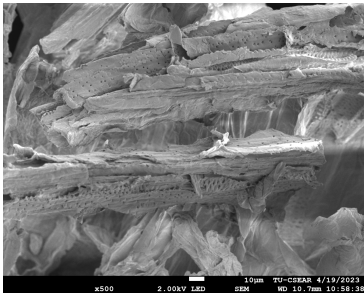
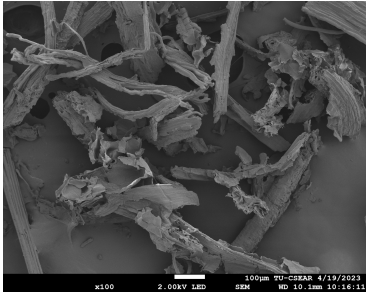
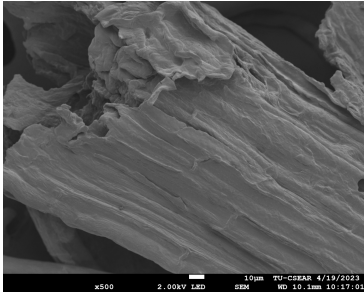
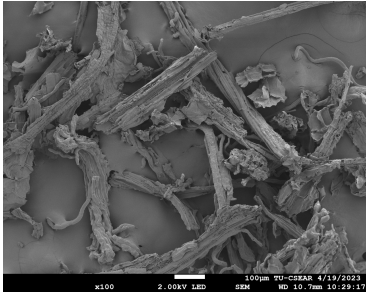
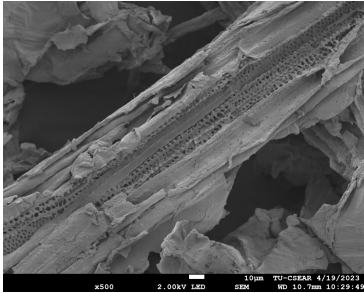
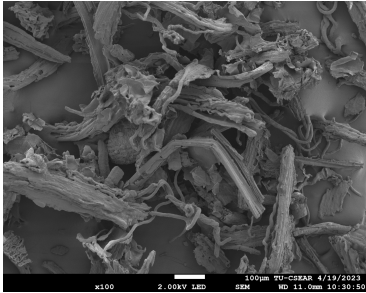
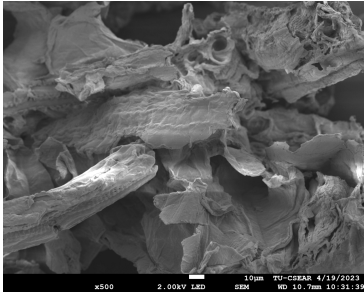
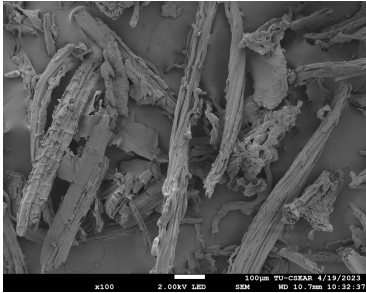
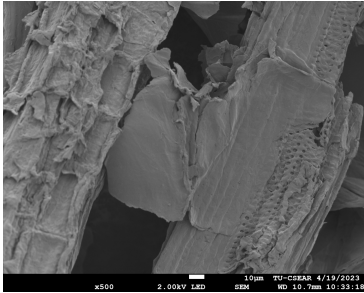
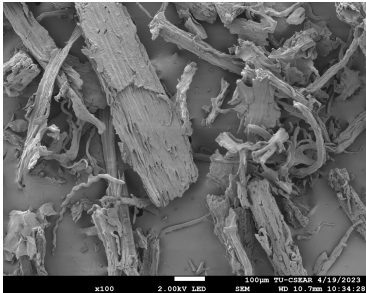
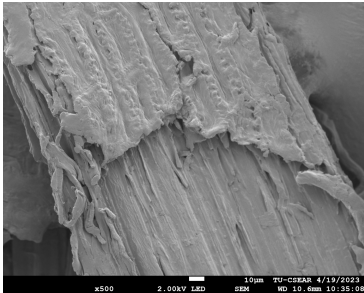
Materials	(Magnification: 100×)	(Magnification: 500×)
α -cellulose		

Table 7. Cont.

Materials	(Magnification: 100×)	(Magnification: 500×)
CMC _{SCL} (20% <i>w/v</i> of NaOH)		
CMC _{SCL} (30% <i>w/v</i> of NaOH)		
CMC _{SCL} (40% <i>w/v</i> of NaOH)		
CMC _{SCL} (50% <i>w/v</i> of NaOH)		
CMC _{SCL} (60% <i>w/v</i> of NaOH)		

4. Conclusions

In Thailand, surplus sugarcane leaves (SCLs) often end up as biomass waste left in the fields after the sugar cane harvesting process. Unfortunately, traditional methods of utilizing these SCL wastes are limited, and their disposal in landfills or through incineration leads to significant environmental issues, particularly air pollution. However, the innova-

tive approach of using them as a chemical feedstock for large-scale production offers a more contemporary solution. This concept holds promise for mitigating pollution problems. This study demonstrates the successful isolation of cellulose from SCLs. Chemical treatments involving alkali and sodium chlorite effectively removed noncellulosic constituents, thus resulting in fibers with a high α -cellulose content making them suitable for CMC production. The carboxymethylation of cellulose was achieved successfully, thereby generating samples with different average degrees of substitution (0.46–0.86) based on the reaction conditions. The highest DS achieved was 0.86 using 40 g/100 mL NaOH. Characterization techniques such as FTIR, XRD, FE-SEM, and ICP-MS provided a comprehensive understanding of the CMC's chemical structure, crystallinity, morphology, and purity level (99.34%), alongside an assessment of the heavy metal content within permissible limits. This process allowed for the production of high-purity CMC suitable for various industrial applications, including paper coating, textile sizing and printing, ceramic glazing, and oil-related uses. This research exemplifies a sustainable approach to utilizing agricultural waste, thereby potentially reducing environmental pollution and offering a pathway to diverse industrial applications of CMC.

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