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Synthesizing Cellulose and Its Derivatives from Pineapple Peel: A Systematic Literature Review

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Abstract: A systematic literature review (SLR) has been carried out to rove about the synthesis method of cellulose and its derivatives from pineapple peel. For now, based on our investigation, carboxymethyl cellulose (CMC) is the only cellulose derivative that can be obtained from pineapple peel. This study collected data from the Google Scholars database via Publish or Perish (PoP) software from 2012 to 2021. The results showed that pineapple peel cellulose (PPC) was synthesized by two methods i.e., chemical and fermentation methods, while CMC was obtained from PPC through additional steps such as alkalization and etherification. The PPC obtained from chemical and fermentation is cellulose I_α and cellulose I_β, respectively. Both types of PPC have a similar conformation of the heavy atom skeleton. Still, their hydrogen bonding patterns are different based on Fourier Transform Infrared (FTIR) and X-ray Diffraction (XRD) results. In this present SLR, PPC's special characteristics are discussed per functional groups and diffractograms data. The exploration of PPC and CMC-based pineapple peel contributes to specific results methodologically. Therefore, this study is necessary to specify a better result in obtaining cellulose and CMC-based pineapple peel as a source for the relevant application.

Keywords: pineapple peel; cellulose; carboxymethyl cellulose; systematic literature review.

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1. Introduction

Pineapple (*Ananas Mill.*) is a typical tropical fruit with distinctive flavor characteristics and several nutritional benefits. It is widely cultivated in the world's tropics for the relevant proceed products, such as canned pineapple and pineapple juice, which are favorite foods of most consumers [1]. Another deep processing method for pineapple fruit is bromelain extraction from pineapple peel. However, processing pineapple fruits and bromelain extraction produces a significant quantity of peel and residue (usually accounting for 30–50 % of the total fresh fruit weight). Hence, properly processing and handling these peels and residues are of practical and academic significance, especially in preventing biological resource waste and environmental pollution [2]. Pineapple peel and residue mainly consist of cellulose, hemicellulose, lignin, pectin, and other components. Despite having less color and rich cellulose (accounting for 20–25 % of the dry weight), pineapple peel and residues are seldom utilized as an industrial product, and little research on the use of cellulose has been carried out so far [2,3]. PPC has promising potential and prospects for materials production that can be applied in certain fields.

So far, PPC has been successfully obtained and modified in another form as hydrogels. Hu *et al*. (2013) reported their work on modifying PPC as hydrogels and adsorbents of heavy metals in waste [4]. In addition, the presence of PPC has opened up another potency, such as converting their structure to cellulose derivatives via its hydroxyl groups, namely methyl cellulose [5], ethyl cellulose [6], hydroxyethyl cellulose [7], hydroxypropyl cellulose [8], cellulose acetate [9], and carboxymethyl cellulose [10]. Carboxymethyl cellulose (CMC) is the most functional among several existing cellulose derivatives due to its low cost, biocompatibility, and biodegradable properties [11,12]. Hence, it has opened up many other potential applications, such as invisible eyes, wound dressing, and drug release systems [2,13]. Nevertheless, methodological exploration is highly desirable to determine how to synthesize PPC with suitable and recommended methods before modifying cellulose into various structures and forms, even applying it. One kind of study to get specific information about the methods of PPC and its derivatives synthesis, namely a systematic literature review (SLR).

SLR is a systematic and explicit method to identify, select, extract, and synthesize the available scientific information from the studies [14–16]. Thus, it is a good recommendation for a specific review of the methods of PPC and its derivatives synthesis. In this present SLR, we discussed the methods for obtaining PPC and its derivatives based on eligibility criteria. Based on the search and knowledge of authors, the study of the PPC and its derivatives using SLR has never been done. In addition, the Boolean algorithm used in database searches using Publish or Perish (PoP) gave excellent results when using Google Scholar as the data source because of the maximum limit of 1000 meta-databases. Therefore, this research can provide consideration for researchers among academics or practitioners in synthesizing cellulose, especially from pineapple peel.

2. Methods

2.1. Protocol.

The Preferred Reporting Items for Systematic Reviews and Meta-Analysis (PRISMA) method is used in this present SLR [17,18]. The review stages consist of developed and described eligibility criteria of information sources, literature search strategy, literature selection process, and data synthesis from selected literature [18]. All steps were carried out to obtain good cellulose and cellulose derivatives methods from pineapple peel based on various characterization. Hence, we can find the best method for synthesizing cellulose and its derivatives in detail.

2.2. Eligibility criteria.

The eligibility criteria are determined to create specific information from any chosen kind of literature. This part consists of inclusion and exclusion criteria. The inclusion criteria are (1) articles that contain some information about cellulose and its derivatives, which are synthesized from pineapple peel; (2) published articles from 2012 until 2021; (3) articles that provide related information about cellulose and its derivatives from pineapple peel. Nevertheless, the exclusion criteria are (1) unsystematic review articles; (2) articles that were published without DOI; (3) articles that were published in a language other than English.

2.3. Search strategy and selection process.

The database search was obtained from the Google Scholar database via PoP software from 2012-2021. In the PoP system, the search technique used the Boolean algorithm as keywords format to search the meta-data involving ("pineapple peel" OR "pineapple peels") AND ("cellulose" OR "carboxymethyl cellulose" OR "methyl cellulose" OR "hydroxyethyl cellulose" OR "hydroxyprophyl cellulose" OR "ethyl cellulose" OR "cellulose derivative") AND ("preparation" OR "synthesis" OR "extraction" OR "isolation"). This strategy generates n = 2397 meta-data ready to be selected via Mendeley Version 1.19.4 software, briefly described in Figure 1. Finally, the selection process resulted in 20 articles shown in Table 1 based on eligibility criteria.

2.4. Synthesis of the result.

The empirical trends focused on in this present SLR are synthesizing the result based on the exploration, obtaining, comparing, and discussing the results from available information. The obtained results are supported by available theories about cellulose and its derivatives.

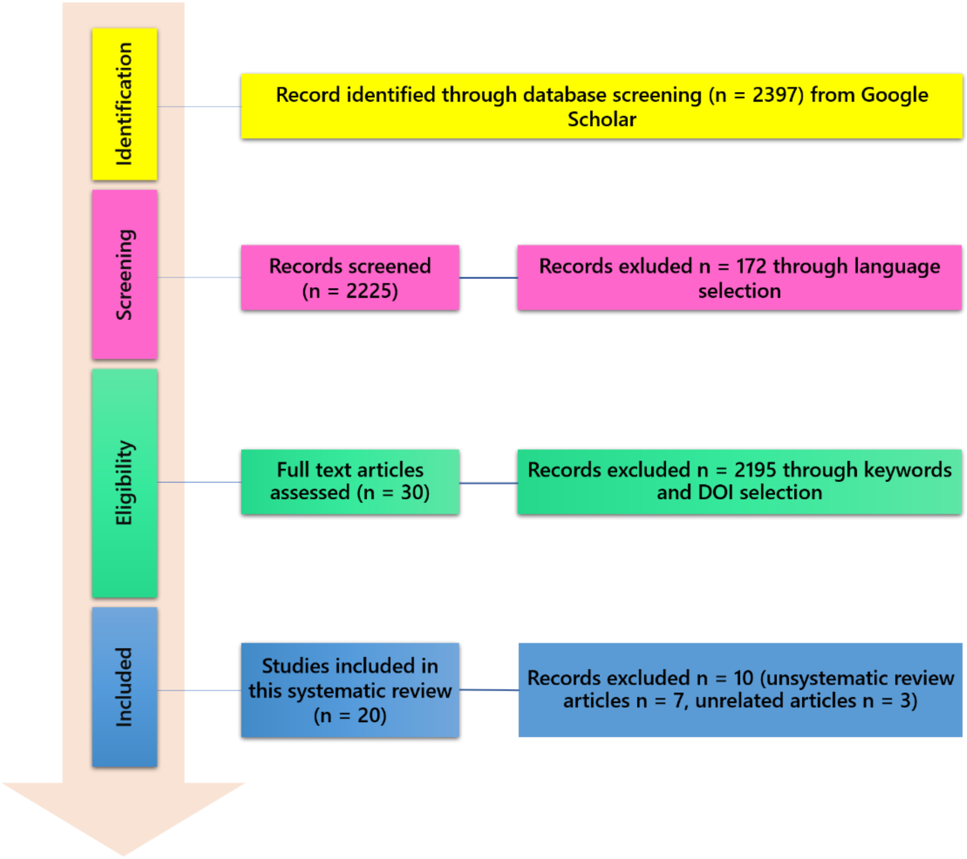

Figure 1. SLR flow chart of study selection.

Table 1. Selected articles for the synthesis of the result.

Year	Titles	Methods		Products	References
		Chemical	Fermentation		
2012	Physicochemical characteristics of pineapple (*Ananas Mill.*) peel cellulose prepared by different methods	☑	☑	Cellulose	[26]
2013	Impacts of some macromolecules on the characteristics of hydrogels	☑	–	Cellulose	[2]

Year	Titles	Methods		Products	References
		Chemical	Fermentation		
	prepared from pineapple peel cellulose using ionic liquid				
2014	Carboxymethyl cellulose from pineapple peel: Useful green bioplastic	☑	–	Cellulose & CMC	[36]
2015	Isolation of cellulose nanocrystals from bacterial cellulose produced from pineapple peel waste juice as culture medium	–	☑	Cellulose	[28]
2016	Modified pineapple peel cellulose hydrogels embedded with sepia ink for effective removal of methylene blue	☑	–	Cellulose	[35]
2016	Isolation of bacterial cellulose nanocrystalline from pineapple peel waste: Optimization of acid concentration in the hydrolysis method	–	☑	Cellulose	[29]
2017	Synthesis, characterization and properties of pineapple peel cellulose-g-acrylic acid hydrogel loaded with kaolin and sepia ink	☑	–	Cellulose	[37]
2017	Pineapple peel carboxymethyl cellulose/polyvinyl alcohol/mesoporous silica SBA-15 hydrogel composites for papain immobilization	☑	–	CMC	[66]
2017	Properties of natural rubber latex filled with bacterial cellulose produced from pineapple peels	–	☑	Cellulose	[25]
2018	Enhanced performances of polyvinyl alcohol films by introducing tannic acid and pineapple peel-derived cellulose nanocrystals	☑	–	Cellulose	[60]
2018	Enhanced swelling and multiple-responsive properties of gelatin/sodium alginate hydrogels by the addition of carboxymethyl cellulose isolated from pineapple peel	☑	–	Cellulose	[65]
2018	Utilization of pineapple peel for production of nanocellulose and film application	☑	–	Cellulose	[44]
2019	Green pH/magnetic sensitive hydrogels based on pineapple peel cellulose and polyvinyl alcohol: synthesis, characterization and naringin prolonged release	☑	–	Cellulose	[67]
2019	Green and facile fabrication of pineapple peel cellulose/magnetic diatomite	☑	–	Cellulose	[45]

Year	Titles	Methods		Products	References
		Chemical	Fermentation		
	hydrogels in ionic liquid for methylene blue absorption				
2019	Mechanical properties of bacterial nanocellulose membrane from pineapple peel waste after homogenization process	–	☑	Cellulose	[53]
2019	Crystallinity and morphology of the bacterial nanocellulose membrane extracted from pineapple peel waste using high-pressure homogenizer	–	☑	Cellulose	[54]
2020	Valorization of pineapple peel waste and sisal fiber: Study of cellulose nanocrystals on polypropylene nanocomposites	☑	–	Cellulose	[34]
2020	Effect of drying methods on the structure of bacterial cellulose from pineapple peel extract	–	☑	Cellulose	[51]
2021	FTIR analysis of alkali treatment on bacterial cellulose films obtained from pineapple peel juice	–	☑	Cellulose	[52]
2021	Properties of bacterial cellulose and it's nanocrystalline obtained from pineapple peel waste juice	–	☑	Cellulose	[27]

3. Results and Discussion

3.1. Pineapple peel cellulose extraction methods.

In this study, we found two methods in this SLR: chemical and fermentation. This diversity of methods certainly gives different cellulose polymorphs (cellulose I-IV), the structure of which depends on the cellulose source, the extraction method, or the treatment [19]. Cellulose I naturally produced two polymorphic structures, namely I_{α} and I_{β}. Cellulose I_{α} allomorph is obtained in algae and bacteria, while I_{β} allomorph is commonly found in plants [19,20]. In cellulose I_{α}, one chain in its structure is present in the triclinic unit cell, and there is no requirement for adjacent glucosyl residues in the same chain to be identical. However, among the two types of cellulose I, I_{β} is the most abundant form of cellulose with a high density of 1.63 $g/cm^{3,}$ which can be resulted from cellulose I_{α} via annealing in a weak alkaline solution. Cellulose I_{β} has a predominant packed parallel in a monoclinic unit, making it the most difficult form to hydrolyze. Therefore, the established pretreatment is required to disrupt its compact structure and the tight association within microfibrils [21].

Furthermore, other cellulose allomorphs can be converted from cellulose I by different chemical treatments, namely cellulose II, III, and IV. Cellulose I_{α} or I_{β} can be converted to cellulose II when both are treated with high concentration alkaline solution and then neutralized. This process is called mercerization. Meanwhile, cellulose I and II can be treated to convert both become III_{I} and III_{II}, as shown in Fig. 2. Cellulose III_{I} can result from Cellulose I_{β} via a stainless-steel vessel reaction with anhydrous liquid ammonia addition. Nevertheless, Cellulose III_{II} also can be obtained by similar treatment with ammonia, but it requires additional treatment, such as nitrogen atmosphere in the synthesis process [22,23].

Furthermore, cellulose IV is another form of polymorph that is reported to be formed through high-temperature treatments (about 260 °C) of cellulose III in glycerol [23]. However, the exact structure of cellulose IV (or its presence) remains controversial due to the poor reproducibility of cellulose IV synthesis and the poor analytical resolution for the artificially produced and allegedly-natural cellulose IV form [24]. In this study, we found only cellulose I polymorph. Therefore, we are discussing more the cellulose I structure from the methods to the characteristics obtained [22].

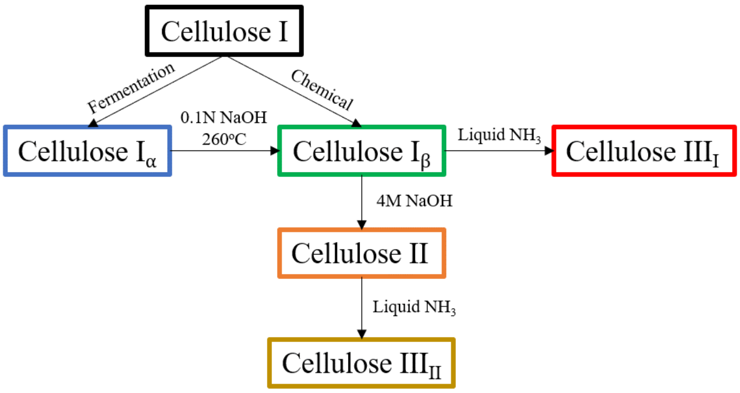

Figure 2. The conversion process of native Cellulose I into I_α, I_β, II, III_I, and III_{II} with some additional treatments.

On the other hand, we found another cellulose production technique, fermentation. Fermentation is a cellulose synthesis biosynthesis technique that utilizes carbon sources from polysaccharides and monosaccharides [25]. This process requires bacterial-assisted such as *Lactobacillus delbrueckiisubsp. Bulgaricus* [26], *Gluconacetobacter xylinus* [27,28], and *Acetobacter xylinum* [25,29]. These bacteria are the acetic acid bacteria group (AAB). AAB is strictly aerobic Gram-negative bacteria classified into *α-Proteobacteria* [30–32]. AAB group was known as strong cellulose producers with an important aspect due to their characteristic of being food-grade or generally recognized as safe bacteria [30,33]. The product from this method is called bacterial cellulose. It is a natural biopolymer composed of high-purity cellulose without impurities such as lignin, pectin, and hemicellulose [25]. Here we discussed how these two methods directly influenced the cellulose structure and its characteristics.

3.2. Chemical.

The chemical method is one of the promising methods in synthesizing PPC. The main steps that we found in this method are (1) preparation (pulped or slurried pineapple peel), (2) alkalization, (3) delignification or bleaching, (4) hydrolysis (5) purification [2,34,35]. However, we found several articles that did not perform the usual steps; for example, Zhang and Xia [26] successfully obtained PPC without the delignification step. Afterward, PPC is also obtained by Chumee and Khemmakama [36] with this method without hydrolysis. These phenomena triggered us to clarify this method. Therefore, further results can provide a clear chemical method for synthesizing PPC.

The method widely reported in this study for the preparation step is grinding the pineapple peel to become powder and pulped in the water. Then the pulped is separated for the next step [26]. Besides, several reports added filtration treatment with etamine in the pulped process; then, the residue was dried and ground to obtain a powder with a better mesh [2,35,37].

Furthermore, the next step is alkalization, or known as mercerization. This process involves the treatment of the fibers and the agro waste to ensure the removal of hemicellulose and lignin from pineapple peel to produce pure cellulose [38,39]. Normally, cellulose interacts with lignin and hemicellulose due to its microfibrils embedded in the lignin and hemicellulose sheath [8]. Hence the optimization of the mercerization process is necessary to carry out. Optimizing the treatment conditions such as time, temperature, liquor ratio, and alkali concentration is important to isolate cellulose with high quality and properties [39,40]. Mostly, sodium hydroxide (NaOH) and potassium hydroxide were utilized for mercerization at a certain pH, molarity, or concentration. Lignin and other compound-like proteins were removed using NaOH and temperature. The lignocellulose molecule and the sites react when in contact with an alkaline solution, then the lignin ester bond could be broken, and the COO^- groups were oxidized, as can be seen in Figure 4 [34,41].

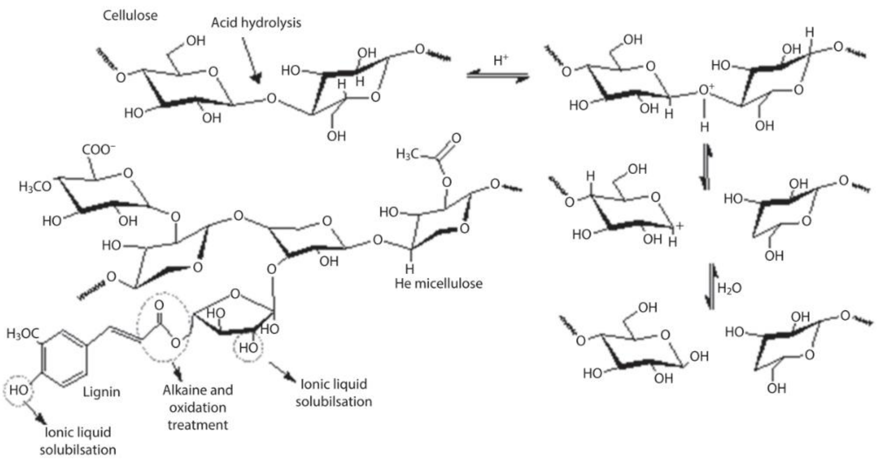

Figure 3. Selectivity of chemical treatments for isolation of lignocellulosic [41].

After that, the delignification process is important to carry out in obtaining high-quality cellulose from pineapple peel. The bleaching process completely removes lignin using chemicals, gases, and steam [42]. The process aims to break down the molecules consisting of chromatographic groups in lignin, remove the impurities, and whiten the pulp [34]. In our findings, bleaching is carried out by using various reagents such as sodium chlorite ($NaClO_2$) [2,35,37,43–45], sodium hypochlorite (NaClO) [34,41,46], calcium hypochlorite ($Ca(ClO)_2$) [36]. It also can be conducted through different reagents under different temperatures, concentrations, and pH conditions. Traditionally, chlorinated bleaching reagents were frequently used among other oxidative bleaching due to their great oxidation level, which is crucial in obtaining high cellulose quality [47].

$$\text{(Fibre)} - O^- Na^+ + HCl \rightarrow \text{(Cellulose)} - OH + NaCl \qquad (1)$$

In addition, one of the important processes in obtaining cellulose from pineapple peel is acid hydrolysis. This process remains the most widely used process to extract cellulose due to its reasonable price and shorter reaction duration [48–50]. HCl is the reagent that is extensively used for acid hydrolysis in synthesizing cellulose from pineapple peel. It works to bind Na^+ as a result of the mercerization process with the scheme, as shown in equation (1) [34]. Therefore, acid hydrolysis completes the previous steps in synthesizing cellulose from pineapple peel. Finally, the cellulose can proceed to purification, i.e., wash the residue with distilled water and ethanol 95% (v/v) several times until the filtrate turns neutral. Overall, the chemical method is still compatible due to its rapid and clear process during cellulose extraction from pineapple peel from mercerization until the purification step.

3.3. Fermentation.

Our findings in this method are divided into several steps, (1) preparation, (2) medium culture, (3) incubation, (4) harvesting pellicles, and (5) purification. In great measure, the fermentation steps we found in this SLR are similar. However, several types of bacteria are used for the synthesis of PPC, including *Acetobacter xylinum* and *Gluconacetobacter xylinus*.

In the preparation step, pineapple peel was cleaned, juiced, and filtered the pineapple peel [27,51,52]. Next, the juice was put into the aqueous solution containing sugar, ammonium sulfate, and acetic acid at a certain concentration, pH, or molarity to get a culture medium [29,51–54]. According to Castro *et al*. (2011), PPJ contains 2.14% (w/v) glucose, 2.4% (w/v) fructose, 2.10% (w/v) sucrose, and 0.31% (w/v) total nitrogen [55], where these ingredients can be used as other carbon sources than sugar. The prepared medium was cooled at room temperature for 24 hours before bacterial addition. After that, *Acetobacter xylinum,* at a certain concentration, was added into the medium and incubated for 10 days under room temperature [29,51,53,54]. This bacterium will form the cellulose between the outer and the cytoplasm membranes by means of polymerizing the glucose molecules to form β-1,4-glucosidic linkages forming cellulose changes. 10-15 parallel chains form a 1.5 nm-wide protofibril. In the second step, several protofibrils are assembled into 2-4 nm wide microfibrils, and in the third step a bundle of microfibrils is assembled into a 20-100 nm-wide ribbon. A matrix of interwoven ribbons constitutes the bacterial cellulose pellicle [55–57]. Next, the pellicles on the liquid medium's surface are cellulose. It was harvested and rinsed with sodium hydroxide and water until neutral pH [25,54].

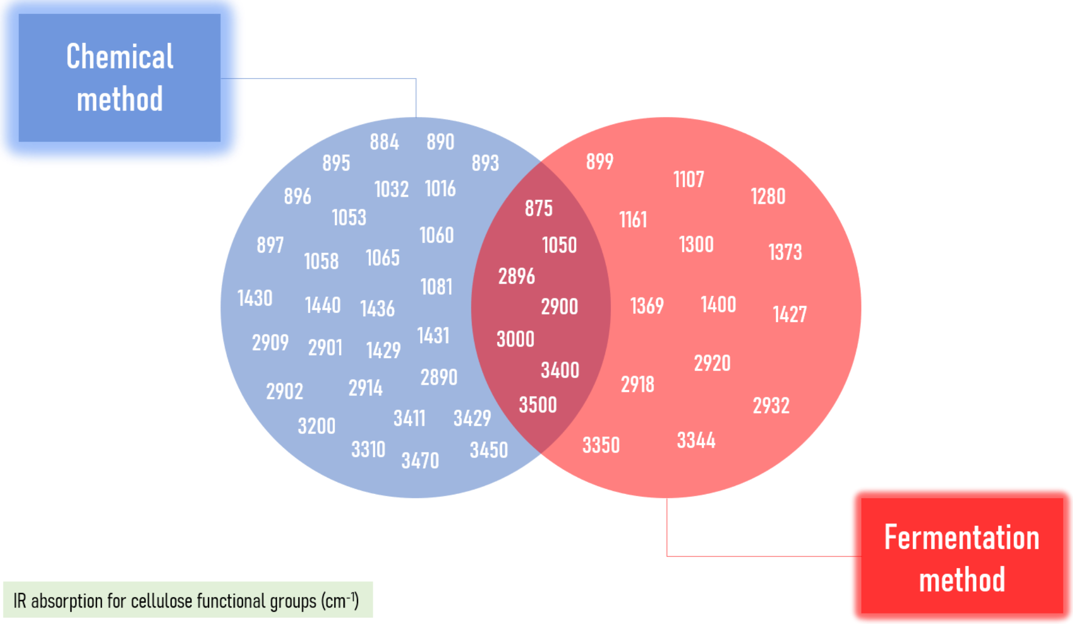

Figure 4. Venn diagram of cellulose functional groups from chemical and fermentation.

Meanwhile, the type of sugar and different incubation times were carried out by Saowapark *et al*. (2017), which used sucrose. Interestingly, the incubation time is faster than others, which is 7 days at room temperature [25]. Nevertheless, using different bacteria, such as *Gluconacetobacter xylinus* can also be utilized, but the required incubation time is longer, 14 days at room temperature [27,28]. Generally, the process is similar in that the substrate supplies energy to bacterial metabolism during the cellulose synthesis exhaustive energy-consuming pathway. Every carbon block the bacterial cell metabolizes into glucose can be used for cellulose production [32,58,59].

3.4. Cellulose characterization.

To confirm the success of a method in extracting cellulose, two characterization methods such as Fourier Transform Infrared (FTIR) and X-ray Diffraction (XRD), are needed. In this study, FTIR and XRD are standard characterizations to be used because they can identify the main characteristics of cellulose. FTIR was used to identify the functional groups that commonly occur in cellulose. In contrast, XRD was used to confirm the type of cellulose obtained from the diffraction angle, and the crystal structure obtained.

We compare the FTIR characterization from several reports obtained by different methods. For the chemical method, the most special functional groups that we found of cellulosic components were C–H rocking vibration of β-glycosidic linkages (875-897 cm^{-1}), C–O–C pyranose stretching vibration (1016-1080 cm^{-1}), C–H bending vibration from β-glycosidic (1429-1440 cm^{-1}), C–H stretching vibration (2896-2902 cm^{-1}), and O–H (3200-3500 cm^{-1}) stretching vibration. B-glycosidic linkages indicate the presence of cellulose, but some impurities based on Madureira *et al*. (2018) indicate there is detected acetyl and ester groups in hemicellulose or carboxylic acid from lignin at 1700 cm^{-1}. This is an imperfection of extraction, especially in the purification process. In the fermentation method, the specific functional groups' absorption is similar to the chemical method, where the main components of cellulose, such as C–H (β-glycosidic linkages) rocking absorption bands, C–O–C bond stretching, and C–H Bending of glycosidic linkages vibration have been identified at the 875-899 cm^{-1}, 1107-1161 cm^{-1}, and 1280-1427 cm^{-1}, respectively. The relation of cellulose functional groups from chemical and fermentation methods are described by the Venn diagram in Figure 5. The FTIR test on cellulose material still needs to be studied comprehensively and in-depth due to the results that appear in this study, namely the very varied in terms of the type of vibration, wavenumber, and absorption level obtained.

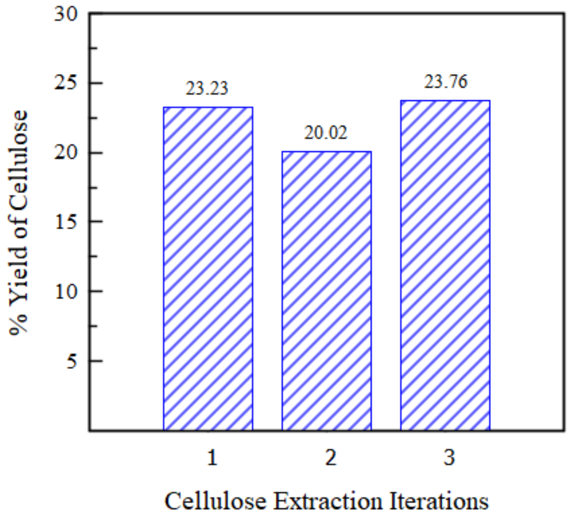

Figure 5. The yield percentage of PPC prepared by chemical method. Data is obtained from ref. [36].

Furthermore, the identified cellulose based on FTIR results can be continued with yield testing. This test was conducted to determine the percentage of cellulose obtained based on the ratio between sample weight and pineapple peel. The percent yield of cellulose was determined by equation (1) [36]. Our findings successfully obtained the PPC results with three iterations through equation (1). there it can be seen in Figure 6 that the PPC produced is 23.23%, 20.02%, and 23.76%. The material loss is attributed to removing impurities and non-cellulosic components, hence allowing acid access to cellulose [34]. Thus, we can determine the weight of cellulose obtained and the weight of impurities lost through equation (1). This parameter becomes important to be modified further to find a strategy for higher yield PPC.

$$\%\text{Yield} = \frac{\text{Weight of PPC}}{\text{Weight of pineapple peel}} \times 100 \qquad (2)$$

Meanwhile, the XRD results showed that cellulose characteristics from chemical and fermentation methods in the Table 2 have a similar type, namely cellulose I. Based on diffraction degree, it was found in three regions in ranges 14.9°-15.3°, 16.40°-16,6°, and 21.9°-22.5°, respectively [26,29]. Cellulose I also found the same type at 22° and 35° for the chemical method [43]. Specifically, cellulose I_β is the most finding in our study that is synthesized from pineapple peel via the chemical method. The former study confirmed the existence of more than one polymorph of cellulose. A crystalline cellulose form caused it to exist near the surface of a crystal which differed from the structure found at the crystal's center, where these two crystalline forms were called cellulose I_α and I_β. These were found with the same conformation of the heavy atom skeleton but different in their hydrogen bonding patterns. Cellulose I_α is produced by organisms, while cellulose I_β is produced by higher plants [19]. Thus, the celluloses which are resulted from pineapple peel in our study were dominated by cellulose I_β, which it confirmed due to the presence of characteristic crystalline peaks repeated at 2θ around 15.3°, 21.8°, 34.6° degrees of diffractogram. They claimed that the individual crystallite segments of cellulose are induced by a partial hydrolytic cleavage of the glycosidic bonds inside the cellulose chains due to the acid hydrolysis process [60,61].

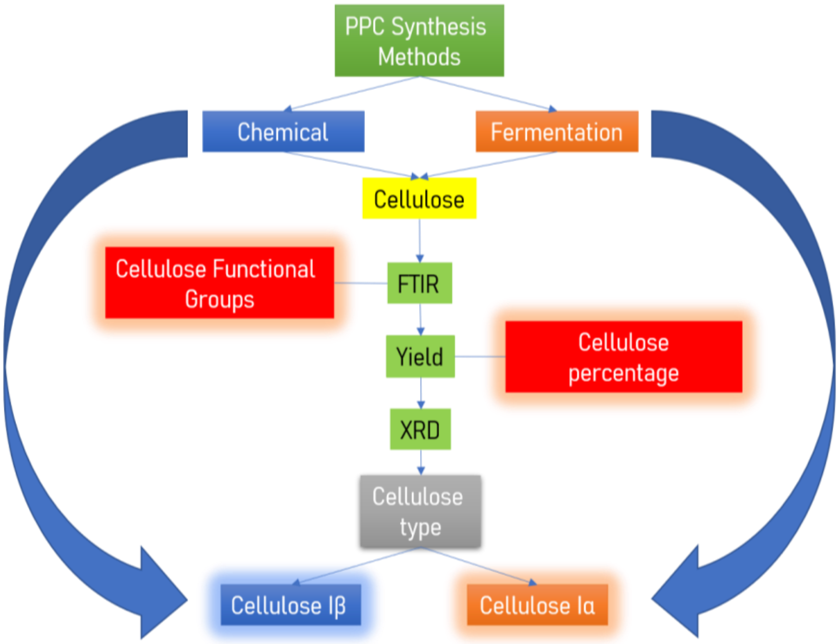

Figure 6. Standard scheme for synthesizing PPC via chemical and fermentation methods.

Furthermore, we obtained information on cellulose XRD patterns produced by fermentation. The type of cellulose resulting from fermentation is also cellulose I. Nonetheless, the characteristic crystalline peaks are different for each result due to their degree of diffractogram. Anwar *et al*. (2016) claimed that the XRD pattern of PPC obtained from the fermentation method presented at 14.9°, 16.40°, and 22.5°, where the crystallinity peak of cellulose I can be found at 22.50° and the amorphous region was identified at 16.40° [29]. Moreover, another finding has assumed that peaks identified at 18°, 22°, and 23° indicate a characteristic of cellulose I [51]. On the other hand, the specific cellulose from pineapple peel was also found via the fermentation method in our study, namely cellulose I_α. Sardjono *et al*. (2019) extended their findings from XRD characterization that cellulose I_α has a triclinic crystal shape with the dimensions a = 0.674 nm, b = 0.593 nm, c = 1.036 nm (chain axis), α= 117°, β= 113°, γ = 81°, and a single cellobiose residue per cell unit. Therefore, based on their crystal structure, we successfully identified the specific characteristics of cellulose from chemical and fermentation methods.

The discussion about the PPC synthesis method becomes more interesting due to its accompanied way of identifying the presence of PPC, yield percentage, and the types of cellulose obtained through XRD characterization. Thus, we found a standard scheme in Fig. 7 for the next study of cellulose synthesis experimentally. The scheme generated from this research is expected to be a benchmark in obtaining cellulose from pineapple peel. The first step to confirm the presence of cellulose in functional groups can be carried out by FTIR testing. If the presence of cellulose functional groups has been confirmed, it can be continued at the stage of obtaining yields from PPC. Finally, XRD can be used to study further the types of cellulose obtained through crystallographic phenomena.

3.5. Pineapple peel cellulose derivatives synthesis method.

Cellulose obtained from pineapple peel still has the potential to be modified in different forms or so-called derivatives. In this study, the obtained cellulose was modified into another form after several reports found the method that can be recommended, namely CMC. CMC has become recent progress in using pineapple peel as a solubility polymer. The degree of solubility of CMC depends on the substitution of carboxymethyl groups instead the hydroxyl groups in cellulose structure. CMC exhibits the greatest potential use in food, paper, cosmetics, textile, pharmaceuticals, and paint due to its good water solubility, biocompatibility, and biodegradability [36,62]. Therefore, CMC is significant in investigating how CMC can be produced from PPC.

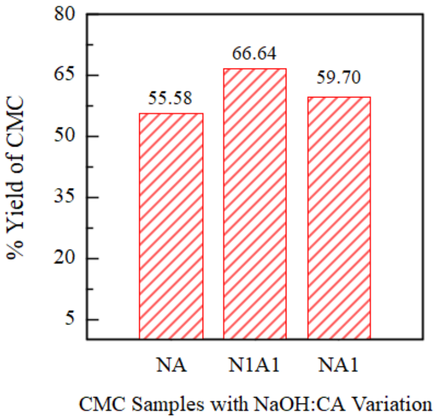

Figure 7. The yield percentage of CMC from PPC with NaOH:CA variation NA = 0.50:0.14; N1A1 = 0.25:0.07; NA1 = 0.50:0.07. Data is obtained from ref. [36].

3.6. Carboxymethyl cellulose.

CMC is converted from pure cellulose by the etherification process. There are two steps for the synthesis of CMC. The first step is activating cellulose with an aqueous NaOH in the slurry of an organic solvent. Then, the last step is the activation of cellulose reaction with chloroacetic acid or sodium chloroacetate. The reactions that were triggered by these processes and their residue are explained in equations (1) to (5) [63,64].

$$\text{Cellulose–OH} + \text{NaOH} \rightarrow \text{Cellulose–ONa} + H_2O \quad (3)$$

$$\text{Cellulose–ONa} + \text{Cl–}CH_2\text{–CO–OH} \rightarrow \text{Cellulose–O–}CH_2\text{–COOH} + \text{NaCl} \quad (4)$$

$$\text{Cellulose–ONa} + \text{Cl–}CH_2\text{–CO–ONa} \rightarrow \text{Cellulose–O–}CH_2\text{–COONa} + \text{NaCl} \quad (5)$$

$$\text{NaOH} + \text{Cl–}CH_2\text{–CO–OH} \rightarrow \text{HO–}CH_2\text{–CO–OH} + \text{NaCl} \quad (6)$$

$$\text{NaOH} + \text{Cl–}CH_2\text{–CO–ONa} \rightarrow \text{HO–}CH_2\text{–CO–ONa} + \text{NaCl} \quad (7)$$

Equation (2) is known as the alkalization process. It aims to activate the hydroxyl groups of cellulose, then carboxymethylation of hydroxyl groups can be conducted with chloroacetic acid or sodium chloroacetate to obtain CMC [65]. However, the residues that can be resulted in the carboxymethylation process are glycolic acid or sodium glycolate. Both are residual reactions between chloroacetic acid/sodium chloroacetate with NaOH. Thus, it needs a purification process, such as soaking in methanol. Based on our findings, other details mechanisms of CMC synthesis were described by Dai *et al*. (2017). They pre-alkalized the PPC mixing with the solvent composed of isopropanol, hydrogen peroxide, and sodium hydroxide at a certain concentration. Afterward, the etherification process was carried out by diazomethane. Then the mixture was neutralized with glacial acetic acid. The neutralized mixture was filtered and washed with methanol and ethanol [66].

$$\%\ \text{yield of CMC} = \frac{\text{Weight of CMC}}{\text{Weight of PPC}} \times 100 \quad (8)$$

The characteristics of CMC were identified by FTIR and determined its yield percentage. In the FTIR result, CMC performs the bands at 3464, 2909, and 1051 cm^{-1} due to the stretching vibrations of O–H, C–H, and C–O–C, respectively. The bands at 1423 and 1322 cm^{-1} resulted from the bending vibrations of C–H and O–H, respectively. The spectrum also showed a strong band of COO^{-} groups at 1653 cm^{-1}, corresponding to the typical adsorption of CMC [66]. In addition, the CMC yield can also be calculated by equation (7) [65]. Chumee and Khemmakama [36] obtained more than 55% yield of CMC at a certain mole ratio of NaOH:Chloroacetic acid (NaOH:CA), as shown in Figure 7. Firstly, they obtained >55% yield in the NA sample; then the yield was enhanced>66% in N1A1 sample when they reduced a-half of the ratio between NaOH:CA. However, the yield decreased <60% in the NA1 sample with controlling NaOH with different CA concentrations. Therefore, there are some interesting results that the variation ratio of NaOH and CA gives different results. This means that NaOH and CA have respective effects in producing the yield of CMC.

Table 2. Description.

Method	2theta	Crystal Plane	Cellulose Type	References
Chemical	15.3°, 16.6°, 22.1°	–	Cellulose I	[26]
Fermentation	15.3°, 16.6°, 21.9°	–	Cellulose I	
Fermentation	14.9°, 16.40°, 22.5°	–	Cellulose I_{α}	[29]
Chemical	15.3°, 21.8°, 34.6°	–	Cellulose I_{β}	[35]
Chemical	15.3°, 21.8°, 34.6°	–	Cellulose I_{β}	[37]

Chemical	15.3°, 21.8°, 34.6°	–	Cellulose I_{β}	[66]
Chemical	16°, 22°, 34°	–	Cellulose I_{β}	[60]
Chemical	16.1°, 22.5°, 34.6°	(110), (200), & (004)	Cellulose I_{β}	[44]
Chemical	22°, 35°	–	Cellulose I	[43]
Chemical	15.32°, 21.77°, 34.61°	(110), (200), & (004)	Cellulose I_{β}	[45]
Chemical	15.3°, 21.8°, 34.6°	–	Cellulose I_{β}	[67]
Fermentation	14.25°, 16.42, 22.45°	(100), (010), & (110)	Cellulose I_{α}	[54]
Chemical	17°, 22.5°, 28.9°, 34.55°, 37.17°	–	Cellulose	[34]
Fermentation	18°, 22°, 23°	–	Cellulose I	[51]

4. Conclusions

The present SLR study is successfully conducted to explore the utilization of pineapple peel for the synthesis of cellulose and its derivative CMC. PPC can be synthesized by chemical and fermentation methods. The main steps in synthesizing cellulose via chemical methods are preparation, alkalization, delignification, acid hydrolysis, and purification. In contrast, fermentation is divided into several steps: preparation, medium culture, incubation, harvesting pellicles, and purification. The standard characterization that is useful to confirm the cellulose presence is also carried out via FTIR and XRD studies. Each method gives a unique and informative result for the next study. Cellulose types from chemical and fermentation methods are cellulose I_{β} and cellulose I_{α}, respectively. Cellulose I_{β} is found in many plants, while cellulose I_{α} is often found in organisms. Structurally, both cellulose types we found have a similar conformation of the heavy atom skeleton but are different in their hydrogen bonding patterns. Meanwhile, CMC from PPC can also be obtained through conventional methods such as alkalization and etherification, then the presence of CMC can be confirmed by calculating the yield percentage.

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Conflicts of Interest

The authors declare no conflict of interest.

References

1. Charlène, L.-J.; Baptiste, Q.; Sophie, A.; Fabienne, R. Maintaining physicochemical, microbiological, and sensory quality of pineapple juice (Ananas comosus, var.' Queen Victoria') through mild heat treatment. *Processes* **2020**, *8*, 1–11, https://doi.org/10.3390/pr8091186.
2. Hu, X.; Wang, J.; Huang, H. Impacts of some macromolecules on the characteristics of hydrogels prepared from pineapple peel cellulose using ionic liquid. *Cellulose* **2013**, *20*, 2923–2933, https://doi.org/10.1007/s10570-013-0075-4.
3. Bardiya, N.; Somayaji, D.; Khanna, S. Biomethanation of banana peel and pineapple waste. *Bioresour. Technol.* **1996**, *58*, 73–76, https://doi.org/10.1016/S0960-8524(96)00107-1.
4. Hu, X.; Hu, K.; Zeng, L.; Zhao, M.; Huang, H. Hydrogels prepared from pineapple peel cellulose using ionic liquid and their characterization and primary sodium salicylate release study. *Carbohydr. Polym.* **2010**, *82*, 62–68, https://doi.org/10.1016/j.carbpol.2010.04.023.
5. Içten, E.; Purohit, H.S.; Wallace, C.; Giridhar, A.; Taylor, L.S.; Nagy, Z.K.; Reklaitis, G. V. Dropwise additive manufacturing of pharmaceutical products for amorphous and self emulsifying drug delivery

systems. *Int. J. Pharm.* **2017**, *524*, 424–432, https://doi.org/10.1016/j.ijpharm.2017.04.003.

6. Davidovich-Pinhas, M.; Barbut, S.; Marangoni, A.G. The gelation of oil using ethyl cellulose. *Carbohydr. Polym.* **2015**, *117*, 869–878, https://doi.org/10.1016/j.carbpol.2014.10.035.
7. Wang, W.; Li, F.; Yu, J.; Navard, P.; Budtova, T. Influence of substitution on the rheological properties and gelation of hydroxyethyl cellulose solution in NaOH-water solvent. *Carbohydr. Polym.* **2015**, *124*, 85–89, https://doi.org/10.1016/j.carbpol.2015.01.065.
8. Seddiqi, H.; Oliaei, E.; Honarkar, H.; Jin, J.; Geonzon, L.C.; Bacabac, R.G.; Klein-Nulend, J. Cellulose and its derivatives: towards biomedical applications. *Cellulose* **2021**, *28*, 1893–1931, https://doi.org/10.1007/s10570-020-03674-w.
9. Khoshnevisan, K.; Maleki, H.; Samadian, H.; Shahsavari, S.; Sarrafzadeh, M.H.; Larijani, B.; Dorkoosh, F.A.; Haghpanah, V.; Khorramizadeh, M.R. Cellulose acetate electrospun nanofibers for drug delivery systems: Applications and recent advances. *Carbohydr. Polym.* **2018**, *198*, 131–141, https://doi.org/10.1016/j.carbpol.2018.06.072.
10. Sahoo, P.K.; Jena, D.K. Synthesis and study of mechanical and fire retardant properties of (carboxymethyl cellulose -g-polyacrylonitrile)/montmorillonite biodegradable nanocomposite. *J. Polym. Res.* **2018**, *25*, 260, https://doi.org/10.1007/s10965-018-1659-3.
11. Kanafi, N.M.; Rahman, N.A.; Rosdi, N.H. Citric acid cross-linking of highly porous carboxymethyl cellulose/poly(ethylene oxide) composite hydrogel films for controlled release applications. *Mater. Today Proc.* **2019**, *7*, 721–731, https://doi.org/10.1016/j.matpr.2018.12.067.
12. Putro, P.A.; Sulaeman, A.S.; Erizal. Synthesis and characterization of swelling properties superabsorbent hydrogel carboxymethylcellulose-g-poly (acrylic acid)/natrium alginate cross-linked by gamma-ray irradiation technique. *J. Phys. Conf. Ser.* **2019**, *1171*, 012011, https://doi.org/10.1088/1742-6596/1171/1/012011.
13. Putro, P.A.; Sulaeman, A.S.; Erizal, E.; Ermawati, I.R. Absorban ion logam Cu(II) berbasis hidrogel superabsorban CMC-g-PKA/NaAlg dengan teknik iradiasi sinar gamma. *Pros. Semin. Nas. Fis. SNF2018* **2018**, *VII*, SNF2018-PA-1-SNF2018-PA-8, https://doi.org/10.21009/03.snf2018.02.pa.01.
14. Putro, P.A.; Sulaeman, A.S.; Maddu, A. Polyvinyl Alcohol-based Hydrogel: A Systematic Literature Review on Thermal Properties by Differential Scanning Calorimetry. *J. Phys. Conf. Ser.* **2021**, *2019*, 012101, https://doi.org/10.1088/1742-6596/2019/1/012101.
15. Sulaeman, A.S.; Putro, P.A.; Nikmatin, S. Thermal studies of hydrogels based on poly(acrylic acid) and its copolymers by differential scanning calorimetry: A systematic literature review. *Polym. Polym. Compos.* **2022**, *30*, 096739112210940, https://doi.org/10.1177/09673911221094022.
16. Entwistle, V.A.; Watt, I.S.; Davis, H.; Dickson, R.; Pickard, D.; Rosser, J. Developing information materials to present the findings of technology assessments to consumers: The experience of the NHS centre for reviews and dissemination. *Int. J. Technol. Assess. Health Care* **1998**, *14*, 47–70, https://doi.org/10.1017/S0266462300010527.
17. Moher, D.; Shamseer, L.; Clarke, M.; Ghersi, D.; Liberati, A.; Petticrew, M.; Shekelle, P.; Stewart, L.A. Preferred reporting items for systematic review and meta-analysis protocols (PRISMA-P) 2015 statement. *Syst. Rev.* **2015**, *4*, 1–9, https://systematicreviewsjournal.biomedcentral.com/articles/10.1186/2046-4053-4-1.
18. Sokouti, M.; Sokouti, B. A PRISMA-compliant systematic review and analysis on color image encryption using DNA properties. *Comput. Sci. Rev.* **2018**, *29*, 14–20, https://doi.org/10.1016/j.cosrev.2018.05.002.
19. Jin, E.; Guo, J.; Yang, F.; Zhu, Y.; Song, J.; Jin, Y.; Rojas, O.J. On the polymorphic and morphological changes of cellulose nanocrystals (CNC-I) upon mercerization and conversion to CNC-II. *Carbohydr. Polym.* **2016**, *143*, 327–335, https://doi.org/10.1016/j.carbpol.2016.01.048.
20. O'sullvian, A.C. Cellulose: the structure slowly unravels. *Cellulose* **1997**, *4*, 173–207, https://link.springer.com/article/10.1023/A:1018431705579.
21. Ling, Z.; Tang, W.; Su, Y.; Huang, C.; Lai, C.; Kirui, A.; Wang, T.; French, A.D.; Yong, Q. Stepwise allomorphic transformations by alkaline and ethylenediamine treatments on bamboo crystalline cellulose for enhanced enzymatic digestibility. *Ind. Crops Prod.* **2022**, *177*, 114450, https://doi.org/10.1016/j.indcrop.2021.114450.
22. Lee, C.M.; Mittal, A.; Barnette, A.L.; Kafle, K.; Park, Y.B.; Shin, H.; Johnson, D.K.; Park, S.; Kim, S.H. Cellulose polymorphism study with sum-frequency-generation (SFG) vibration spectroscopy: Identification of exocyclic CH2OH conformation and chain orientation. *Cellulose* **2013**, *20*, 991–1000, https://doi.org/10.1007/s10570-013-9917-3.
23. Kim, S.H.; Lee, C.M.; Kafle, K. Characterization of crystalline cellulose in biomass: Basic principles, applications, and limitations of XRD, NMR, IR, Raman, and SFG. *Korean J. Chem. Eng.* **2013**, *30*, 2127–2141, https://doi.org/10.1007/s11814-013-0162-0.
24. Wada, M.; Heux, L.; Sugiyama, J. Polymorphism of cellulose I family: Reinvestigation of cellulose IVl. *Biomacromolecules* **2004**, *5*, 1385–1391, https://doi.org/10.1021/bm0345357.
25. Saowapark, T.; Chaichana, E.; Jaturapiree, A. Properties of natural rubber latex filled with bacterial cellulose produced from pineapple peels. *J. Met. Mater. Miner.* **2017**, *27*, 12–16, https://jmmm.material.chula.ac.th/index.php/jmmm/article/view/335.

26. Zhang, Y.; Xia, X. Physicochemical characteristics of pineapple (ananas mill.) peel cellulose prepared by different methods. *Adv. Mater. Res.* **2012**, *554–556*, 1038–1041, https://doi.org/10.4028/www.scientific.net/AMR.554-556.1038.
27. Anwar, B.; Bundjali, B.; Sunarya, Y.; Arcana, I.M. Properties of Bacterial Cellulose and Its Nanocrystalline Obtained from Pineapple Peel Waste Juice. *Fibers Polym.* **2021**, *22*, 1228–1236, https://doi.org/10.1007/s12221-021-0765-8.
28. Anwar, B.; Bundjali, B.; Arcana, I.M. Isolation of Cellulose Nanocrystals from Bacterial Cellulose Produced from Pineapple Peel Waste Juice as Culture Medium. *Procedia Chem.* **2015**, *16*, 279–284, https://doi.org/10.1016/j.proche.2015.12.051.
29. Anwar, B.; Rosyid, N.H.; Effendi, D.B.; Nandiyanto, A.B.D.; Mudzakir, A.; Hidayat, T. Isolation of bacterial cellulose nanocrystalline from pineapple peel waste: Optimization of acid concentration in the hydrolysis method. *AIP Conf. Proc.* **2016**, *1708*, 040001, https://doi.org/10.1063/1.4941151.
30. Škraban, J.; Cleenwerck, I.; Vandamme, P.; Fanedl, L.; Trček, J. Genome sequences and description of novel exopolysaccharides producing species Komagataeibacter pomaceti sp. nov. and reclassification of Komagataeibacter kombuchae (Dutta and Gachhui 2007) Yamada *et al*., 2013 as a later heterotypic synonym of Komagataeib. *Syst. Appl. Microbiol.* **2018**, *41*, 581–592, https://doi.org/10.1016/j.syapm.2018.08.006.
31. Slapšak, N.; Cleenwerck, I.; De Vos, P.; Trček, J. Gluconacetobacter maltaceti sp. nov., a novel vinegar producing acetic acid bacterium. *Syst. Appl. Microbiol.* **2013**, *36*, 17–21, https://doi.org/10.1016/j.syapm.2012.11.001.
32. Gorgieva; Trček Bacterial Cellulose: Production, Modification and Perspectives in Biomedical Applications. *Nanomaterials* **2019**, *9*, 1352, https://doi.org/10.3390/nano9101352.
33. Castro, C.; Zuluaga, R.; Álvarez, C.; Putaux, J.L.; Caro, G.; Rojas, O.J.; Mondragon, I.; Gañán, P. Bacterial cellulose produced by a new acid-resistant strain of Gluconacetobacter genus. *Carbohydr. Polym.* **2012**, *89*, 1033–1037, https://doi.org/10.1016/j.carbpol.2012.03.045.
34. Agarwal, J.; Mohanty, S.; Nayak, S.K. Valorization of pineapple peel waste and sisal fiber: Study of cellulose nanocrystals on polypropylene nanocomposites. *J. Appl. Polym. Sci.* **2020**, *137*, 49291, https://doi.org/10.1002/app.49291.
35. Dai, H.; Huang, H. Modified pineapple peel cellulose hydrogels embedded with sepia ink for effective removal of methylene blue. *Carbohydr. Polym.* **2016**, *148*, 1–10, https://doi.org/10.1016/j.carbpol.2016.04.040.
36. Chumee, J.; Khemmakama, P. Carboxymethyl cellulose from pineapple peel: Useful green bioplastic. *Adv. Mater. Res.* **2014**, *979*, 366–369, https://doi.org/10.4028/www.scientific.net/AMR.979.366.
37. Dai, H.; Huang, H. Synthesis, characterization and properties of pineapple peel cellulose-g-acrylic acid hydrogel loaded with kaolin and sepia ink. *Cellulose* **2017**, *24*, 69–84, https://doi.org/10.1007/s10570-016-1101-0.
38. Agwuncha, S.C.; Owonubi, S.; Fapojuwo, D.P.; Abdulkarim, A.; Okonkwo, T.P.; Makhatha, E.M. Evaluation of mercerization treatment conditions on extracted cellulose from shea nut shell using FTIR and thermogravimetric analysis. *Mater. Today Proc.* **2021**, *38*, 958–963, https://doi.org/10.1016/j.matpr.2020.05.473.
39. Alila, S.; Besbes, I.; Vilar, M.R.; Mutjé, P.; Boufi, S. Non-woody plants as raw materials for production of microfibrillated cellulose (MFC): A comparative study. *Ind. Crops Prod.* **2013**, *41*, 250–259, https://doi.org/10.1016/j.indcrop.2012.04.028.
40. Li, Y.; Liu, Y.; Chen, W.; Wang, Q.; Liu, Y.; Li, J.; Yu, H. Facile extraction of cellulose nanocrystals from wood using ethanol and peroxide solvothermal pretreatment followed by ultrasonic nanofibrillation. *Green Chem.* **2016**, *18*, 1010–1018, https://doi.org/10.1039/c5gc02576a.
41. Camacho, M.; Ureña, Y.R.C.; Lopretti, M.; Carballo, L.B.; Moreno, G.; Alfaro, B.; Vega Baudrit, J.R. Synthesis and characterization of nanocrystalline cellulose derived from Pineapple peel residues. *J. Renew. Mater.* **2017**, *5*, 271–279, https://doi.org/10.7569/JRM.2017.634117.
42. Rizwan, M.; Gilani, S.R.; Durrani, A.I.; Naseem, S. Cellulose extraction of Alstonia scholaris: A comparative study on efficiency of different bleaching reagents for its isolation and characterization. *Int. J. Biol. Macromol.* **2021**, *191*, 964–972, https://doi.org/10.1016/j.ijbiomac.2021.09.155.
43. Madureira, A.R.; Atatoprak, T.; Çabuk, D.; Sousa, F.; Pullar, R.C.; Pintado, M. Extraction and characterisation of cellulose nanocrystals from pineapple peel. *Int. J. Food Stud.* **2018**, *7*, 24–33, https://doi.org/10.7455/ijfs/7.1.2018.a3.
44. Dai, H.; Ou, S.; Huang, Y.; Huang, H. Utilization of pineapple peel for production of nanocellulose and film application. *Cellulose* **2018**, *25*, 1743–1756, https://doi.org/10.1007/s10570-018-1671-0.
45. Dai, H.; Huang, Y.; Zhang, Y.; Zhang, H.; Huang, H. Green and facile fabrication of pineapple peel cellulose/magnetic diatomite hydrogels in ionic liquid for methylene blue adsorption. *Cellulose* **2019**, *26*, 3825–3844, https://doi.org/10.1007/s10570-019-02283-6.
46. Corrales-Ureña, Y.R.; Villalobos-Bermúdez, C.; Pereira, R.; Camacho, M.; Estrada, E.; Argüello-Miranda, O.; Vega-Baudrit, J.R. Biogenic silica-based microparticles obtained as a sub-product of the nanocellulose extraction process from pineapple peels. *Sci. Rep.* **2018**, *8*, 10417, https://doi.org/10.1038/s41598-018-28444-4.

47. Wang, T.; Zhao, Y. Optimization of bleaching process for cellulose extraction from apple and kale pomace and evaluation of their potentials as film forming materials. *Carbohydr. Polym.* **2021**, *253*, 117225, https://doi.org/10.1016/j.carbpol.2020.117225.
48. Tarchoun, A.F.; Trache, D.; Klapötke, T.M.; Derradji, M.; Bessa, W. Ecofriendly isolation and characterization of microcrystalline cellulose from giant reed using various acidic media. *Cellulose* **2019**, *26*, 7635–7651, https://doi.org/10.1007/s10570-019-02672-x.
49. Tarchoun, A.F.; Trache, D.; Klapötke, T.M. Microcrystalline cellulose from Posidonia oceanica brown algae: Extraction and characterization. *Int. J. Biol. Macromol.* **2019**, *138*, 837–845, https://doi.org/10.1016/j.ijbiomac.2019.07.176.
50. Trache, D.; Hussin, M.H.; Hui Chuin, C.T.; Sabar, S.; Fazita, M.R.N.; Taiwo, O.F.A.; Hassan, T.M.; Haafiz, M.K.M. Microcrystalline cellulose: Isolation, characterization and bio-composites application—A review. *Int. J. Biol. Macromol.* **2016**, *93*, 789–804, https://doi.org/10.1016/j.ijbiomac.2016.09.056.
51. Suryanto, H.; Muhajir, M.; Zakia, N.; Yanuhar, U.; Aminnudin, A.; Pradana, Y.R.A. Effect of Drying Methods on the Structure of Bacterial Cellulose from Pineapple Peel Extract. *Key Eng. Mater.* **2020**, *851*, 79–85, https://doi.org/10.4028/www.scientific.net/KEM.851.79.
52. Suryanto, H.; Aminnudin; Mahsuli; Wijaya, H.W.; Yanuhar, U. FTIR analysis of alkali treatment on bacterial cellulose films obtained from pineapple peel juice. *IOP Conf. Ser. Mater. Sci. Eng.* **2021**, *1034*, 012145, https://doi.org/10.1088/1757-899x/1034/1/012145.
53. Mahsuli, T.; Suryanto, H.; Larasati, A.; Muhajir, M. Mechanical properties of bacterial nanocellulose membrane from pineapple peel waste after homogenization process. *AIP Conf. Proc.* **2019**, *2120*, 050019, https://doi.org/10.1063/1.5115695.
54. Sardjono, S.A.; Suryanto, H.; Aminnudin; Muhajir, M. Crystallinity and morphology of the bacterial nanocellulose membrane extracted from pineapple peel waste using high-pressure homogenizer. *AIP Conf. Proc.* **2019**, *2120*, 080015, https://doi.org/10.1063/1.5115753.
55. Castro, C.; Zuluaga, R.; Putaux, J.L.; Caro, G.; Mondragon, I.; Gañán, P. Structural characterization of bacterial cellulose produced by Gluconacetobacter swingsii sp. from Colombian agroindustrial wastes. *Carbohydr. Polym.* **2011**, *84*, 96–102, https://doi.org/10.1016/j.carbpol.2010.10.072.
56. Laskar, J.; Robutel, P. High order symplectic integrators for perturbed Hamiltonian systems. *Celest. Mech. Dyn. Astron.* **2001**, *80*, 39–62, https://doi.org/10.1023/A:1012098603882.
57. Klemm, D.; Schumann, D.; Udhardt, U.; Marsch, S. Bacterial synthesized cellulose - Artificial blood vessels for microsurgery. *Prog. Polym. Sci.* **2001**, *26*, 1561–1603, https://doi.org/10.1016/S0079-6700(01)00021-1.
58. Krasteva, P.V.; Bernal-Bayard, J.; Travier, L.; Martin, F.A.; Kaminski, P.A.; Karimova, G.; Fronzes, R.; Ghigo, J.M. Insights into the structure and assembly of a bacterial cellulose secretion system. *Nat. Commun.* **2017**, *8*, 25–28, https://doi.org/10.1038/s41467-017-01523-2.
59. Wang, S.S.; Han, Y.H.; Chen, J.L.; Zhang, D.C.; Shi, X.X.; Ye, Y.X.; Chen, D.L.; Li, M. Insights into bacterial cellulose biosynthesis from different carbon sources and the associated biochemical transformation pathways in Komagataeibacter sp. W1. *Polymers (Basel).* **2018**, *10*, 963, https://doi.org/10.3390/polym10090963.
60. Dai, H.; Huang, Y.; Huang, H. Enhanced performances of polyvinyl alcohol films by introducing tannic acid and pineapple peel-derived cellulose nanocrystals. *Cellulose* **2018**, *25*, 4623–4637, https://doi.org/10.1007/s10570-018-1873-5.
61. Chen, Y.N.; Peng, L.; Liu, T.; Wang, Y.; Shi, S.; Wang, H. Poly(vinyl alcohol)-Tannic Acid Hydrogels with Excellent Mechanical Properties and Shape Memory Behaviors. *ACS Appl. Mater. Interfaces* **2016**, *8*, 27199–27206, https://doi.org/10.1021/acsami.6b08374.
62. Pushpamalar, V.; Langford, S.J.; Ahmad, M.; Lim, Y.Y. Optimization of reaction conditions for preparing carboxymethyl cellulose from sago waste. *Carbohydr. Polym.* **2006**, *64*, 312–318, https://doi.org/10.1016/j.carbpol.2005.12.003.
63. Toğrul, H.; Arslan, N. Production of carboxymethyl cellulose from sugar beet pulp cellulose and rheological behaviour of carboxymethyl cellulose. *Carbohydr. Polym.* **2003**, *54*, 73–82, https://doi.org/10.1016/S0144-8617(03)00147-4.
64. Rachtanapun, P.; Luangkamin, S.; Tanprasert, K.; Suriyatem, R. Carboxymethyl cellulose film from durian rind. *LWT - Food Sci. Technol.* **2012**, *48*, 52–58, https://doi.org/10.1016/j.lwt.2012.02.029.
65. Dai, H.; Ou, S.; Huang, Y.; Liu, Z.; Huang, H. Enhanced swelling and multiple-responsive properties of gelatin/sodium alginate hydrogels by the addition of carboxymethyl cellulose isolated from pineapple peel. *Cellulose* **2018**, *25*, 593–606, https://doi.org/10.1007/s10570-017-1557-6.
66. Dai, H.; Ou, S.; Liu, Z.; Huang, H. Pineapple peel carboxymethyl cellulose/polyvinyl alcohol/mesoporous silica SBA-15 hydrogel composites for papain immobilization. *Carbohydr. Polym.* **2017**, *169*, 504–514, https://doi.org/10.1016/j.carbpol.2017.04.057.