

ORIGINAL ARTICLE

Microcrystalline Cellulose from Coconut Pulp Residue: Hydrolysis Optimization for Bioplastic Reinforcement

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ABSTRACT - The volume of plastic waste in Indonesia grows every year, resulting in environmental pollution that affects both terrestrial and aquatic environments. Previous research has developed bioplastics as alternatives to conventional plastics.", however, PVA-based bioplastics lack sufficient mechanical strength and require reinforcement materials. Coconut pulp residue shows potential as a sustainable material for this role, but must be synthesized into microcrystalline cellulose (MCC) to function effectively. Converting cellulose into MCC begins with delignification, which produces alpha-cellulose through separation of the crystalline regions. Although previous studies have successfully produced MCC from coconut pulp residue, optimization of the hydrolysis process has not been thoroughly investigated. Therefore, this research investigates the optimum HCl concentration and heating duration for hydrolysis, along with their impact on MCC properties. A factorial design was applied using three HCl concentrations (1.5 M, 2.0 M, and 2.5 M) and three heating times (10, 20, and 30 minutes). The research began with cleaning and defatting of coconut pulp residue, followed by delignification, bleaching, and hydrolysis, and ended with MCC characterization through yield percentage and FTIR-based Hit Quality Index (HQI) scoring. Results showed that 2.0 M HCl for 10 minutes produced the optimum MCC, confirmed by FTIR functional groups, 37.25% crystallinity from XRD, and SEM-observed morphology.

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INTRODUCTION

Plastic serves as a common material which people use extensively throughout their daily activities. On a global scale, plastic waste constitutes over 12% of total municipal solid waste generated worldwide, making it one of the most significant single-material contributors to the global waste stream, alongside organic and food waste [1]. Plastics themselves are composed of polymers formed by chemical bonds between monomers and various additives. Plastics show high resistance to natural decomposition because of their polymeric structure which needs more than several years to break down [2]. Studies identify plastics as microplastics when they reach a state of degradation which results in their transformation into small fragments [3]. The small plastic particles known as microplastics spread throughout water bodies and land surfaces and air spaces before they could potentially harm human health through the food supply [4]. Recent studies now develop bioplastics as a sustainable alternative to conventional plastics [5]. Studies conducted by Supardi and Karmini demonstrates that bioplastics fulfil all requirements for food-grade materials which makes them suitable for food product packaging [6].

The development of bioplastics as an alternative to traditional plastics has become more popular because of increasing plastic waste problems, yet these materials show weaker mechanical properties and shorter lifespan than petroleum-based plastics. Therefore, it's required to develop reinforcement agents to improve their performance. Cellulose extracted from coconut pulp residue demonstrates potential to serve as this reinforcement agent which would improve the mechanical properties of bioplastic composites. Tropical countries provide favorable climatic conditions for extensive coconut cultivation, and Indonesia stands out as one prominent example of such a country [7]. According to 2023 data from the Indonesian Central Bureau of Statistics (BPS), Indonesia is among the world's leading coconut producers, with approximately 3.32 million hectares of coconut plantations yielding 2.83 million tons of coconuts annually. As in many other coconut-producing tropical nations, Indonesia's food industry generates substantial coconut milk output due to its widespread use in culinary practices. This extraction process, in turn, produces biomass waste, particularly coconut pulp residue, which is commonly discarded or repurposed as animal feed. Therefore, production of bioplastics using coconut pulp residue reinforcement agent presents a sustainable method to decrease food industry waste [8].

However, the extracted cellulose from coconut pulp residue needs additional processing to ensure its suitability as a reinforcement agent. The material needs to be processed into micron-scale particles while undergoing purification to eliminate lignin and hemicellulose components. The process exists to synthesize crystalline cellulose which will be purified. The purified crystalline form is commonly referred to as Microcrystalline Cellulose (MCC), which can be used

as an additive in bioplastic production. Previous research by Avbunudiogba *et al.* investigated the production of MCC from coconut pulp residue, however, the study did not optimize the hydrochloric acid (HCl) concentration and heating time during the hydrolysis process [9]. Although hydrolysis optimization has been explored in other studies, the raw materials were not derived from coconut pulp residue.

Based on this research gap, particularly the limited number of studies addressing hydrolysis optimization for MCC derived from coconut pulp residue, the present study aims to identify the optimum HCl concentration and heating duration for this process, using a factorial design with three HCl concentration levels and three heating time periods, following the experimental approach described by Liu *et al.* [10]. The resulting optimized MCC is then benchmarked against commercially available MCC to evaluate its properties. Overall, this research investigates how HCl concentration and heating duration affect the properties of MCC derived from coconut pulp residue, with the goal of producing an optimum reinforcement material for preliminary bioplastic applications.

EXPERIMENTAL METHOD

The production of MCC began with pre-treatment of the coconut pulp residue, followed by drying and chemical treatment to remove non-cellulosic substances, prior to acid hydrolysis under various processing conditions. Each treatment was carried out in triplicate (3 replications) to ensure reproducibility, while subsequent analyses were performed in duplicate (2x) to verify measurement consistency. The resulting MCC properties were analyzed to identify the optimum processing parameters, followed by further characterization to determine its functional groups, morphology, and crystallinity. The optimized MCC was then applied in a preliminary bioplastic formulation to assess its feasibility as a reinforcement agent.

Materials

This research uses coconut pulp residue obtained from local market (Langgar Market) in Semarang, Indonesia, along with technical-grade ethanol (96%), distilled water, citric acid (ACS Reagent $\geq 99\%$, Sigma Aldrich), sodium hydroxide (NaOH) pellets (EMSURE[®], Merck), sodium sulfite (Na₂SO₃) anhydrous (EMSURE[®], Merck), sodium hypochlorite (NaOCl) solution (4.00-4.99%, Merck), analytical-grade hydrochloric acid (37% HCl) from Merck, and commercially available MCC reference material, Avicel[®] PH 101.

Coconut Pulp Residue Pre-treatment

Mature coconut (*Cocos nucifera* L.) flesh obtained from Langgar Market, Semarang, was finely grated, pressed to extract its coconut milk, and the resulting pulp residue was rinsed with running distilled water. The residue was then boiled in distilled water for 20 minutes, filtered, and boiled again for another 20 minutes before being oven-dried at 50 °C for 24 hours. Ethanol 96% solution was used to treat the dried material at 60 °C for 30 minutes before the residue is being dried once again at 50 °C for 24 hours to obtain the final purified coconut pulp residue [11].

α -cellulose Delignification Process

The delignification process is based on Avbunudiogba *et al.* with some modifications. 1 g of treated coconut pulp residue was then soaked in 10 mL citric acid solution at 3.5% (w/v) concentration, then heated in a water bath at 90 °C for 2 hours [9]. Then, 2% (w/v) NaOH and 2% (w/v) Na₂SO₃ solution was added to the mixture, which then heated at 50 °C for another 1 hour before filtered. The residue is then immersed in 17.5% (w/v) NaOH solution at 80 °C for 30 minutes before rinsed with distilled water until the solution reached neutral pH (pH=7). The sample underwent bleaching with 3.2% (w/v) NaOCl solution before drying in oven at 50 °C for 12 hours. The dried sample was then grinded, followed by sieving through a 70-mesh sieve, to obtain fine α -cellulose powder.

MCC Hydrolysis

The obtained α -cellulose underwent hydrolysis through HCl solutions at three different concentrations (1.5, 2.0 and 2.5 M) while being heated at 90 °C for three time periods (10, 20 and 30 minutes) with a 1:10 solid-to-liquid ratio. The solution needed to be immersed in distilled water until it reached neutral pH through multiple cleaning procedures before oven drying at 60 °C for 48 hours to produce MCC powder [9].

MCC Characterization

The MCC characterization process required two distinct methods to determine yield percentage and functional group identification, before another physical property tests using XRD and SEM and FTIR spectroscopy. The experimental data underwent statistical analysis through mathematical modeling software to evaluate treatment variable differences according to Avbunudiogba *et al.* [9]. Yield calculation percentage is calculated by dividing the weight of MCC powder extracted through hydrolysis to the initial weight of α -cellulose before hydrolysis as shown in Equation (1).

$$\text{Yield \%} = \frac{\text{mass (after hydrolysis)}}{\text{mass (before hydrolysis)}} \times 100\% \quad (1)$$

XRD analysis was performed using a Bruker D8 Advance diffractometer with a scanning range of 2θ between 0° and 90°. Approximately 1 g of each sample was first ground using a ball mill to obtain a fine powder, then placed into a flat

sample holder and leveled evenly. The crystallinity index (CrI) was calculated using the Segal method according to Equation (2).

$$CrI = \frac{I_{200} - I_{amorf}}{I_{200}} \times 100\% \quad (2)$$

- CrI : Crystallinity Index
- I_{200} : maximum intensity at $2\theta = 19-22^\circ$
- I_{amorf} : maximum intensity at $2\theta = 16-18^\circ$

The solution for bioplastic preparation required 5 grams of polyvinyl alcohol (PVA) and two drops of dimethicone which was then dissolved in 100 mL of distilled water under stirring at 90 °C for 30 minutes to obtain a clear solution. Then, 2 mL of glycerol and 0.3 grams of citric acid were added to the mixture. Afterwards, 0.5 grams of MCC was added into the mixture while being stirred using a hand blender at 70 °C for 15 minutes. The bioplastic solution obtained under optimum conditions was poured into a 20 x 5 cm acrylic mold with a thickness of 1 mm to form the bioplastic films.

Data Analysis

The scoring matrix method enabled the analysis to find the best conditions through complete assessment of all evaluated parameters. Yield percentage and FTIR-HQI, despite being measured in different units and scales, were each converted onto a common 1-5 scoring scale using min-max normalization, following the general form shown in Equation (3).

$$F_{ij} = \frac{f_{ij} - \min(f_{ij})}{\max(f_{ij}) - \min(f_{ij})} \quad (3)$$

where f_{ij} is the raw value of parameter j for sample i , and F_{ij} is its normalized value. The lowest observed value for each parameter was assigned a score of 1 and the highest a score of 5, with intermediate values scaled proportionally between these bounds. This normalization approach follows established practice for combining criteria of differing units into a single comparable score in multi-criteria process and materials optimization and allows yield and HQI to be weighted equally without either parameter numerically dominating the total score due to its larger range or magnitude [12], [13], [14]. R-Studio software was used for statistical analysis to identify which independent variables affected the measured data obtained from the collected samples. Normality and homogeneity tests were first conducted using the Shapiro-Wilk and Levene's tests, respectively, to determine the appropriate statistical method for each measured parameter. Where the data met these assumptions, a parametric two-way ANOVA was applied, where the assumptions of normality or homogeneity of variance were not met, a non-parametric Aligned Rank Transform (ART) ANOVA was applied instead.

RESULT AND DISCUSSION

Yield Percentage Analysis

The yield percentage was determined by comparing the weight of MCC after the hydrolysis process to the initial weight before hydrolysis. The yield percentage data obtained are shown in Table 1.

Table 1. Yield percentage and scoring matrix for MCC hydrolysis

Sample	HCl concentration (M)	Heating time (min)	Yield percentage (%)	Scoring matrix
1	1.5	10	76.92 ± 0.375 ^a	4.87
2		20	73.71 ± 1.541 ^{ab}	4.02
3		30	69.77 ± 0.778 ^{ab}	2.99
4	2.0	10	77.43 ± 1.089 ^a	5.00
5		20	75.41 ± 2.524 ^{ab}	4.47
6		30	72.02 ± 3.946 ^{ab}	3.58
7	2.5	10	71.96 ± 2.305 ^{ab}	3.56
8		20	67.12 ± 0.516 ^{ab}	2.29
9		30	62.2 ± 2.107 ^b	1.00

Notes:

- The reported values represent the mean ± standard deviation (STDEV) of three replicates which were measured in two repetitions.
- Different superscript letters indicate significant differences based on the ART ANOVA test (p-value < 0.05).

The experiment produced its highest yield when the HCl solution reached 2.0 M concentration during 10 minutes of heating, which resulted in a 77.43% yield. The lowest yield occurred when the experiment used 2.5 M HCl solution during 30 minutes of heating, resulting in a 62% yield. The post-Hoc analysis of heating time on yield percentage showed that

only the comparison between 10 minutes and 30 minutes resulted in a statistically significant difference, as indicated by a *p*-value below 0.05. In contrast, the comparisons between 10 minutes and 20 minutes, as well as between 20 minutes and 30 minutes, did not demonstrate significant differences.

Prior to hypothesis testing, the yield percentage data were assessed for normality and homogeneity of variance to determine the appropriate statistical approach. The Shapiro-Wilk test indicated a significant departure from normality ($p = 0.0001915$), and Levene's test indicated a significant lack of homogeneity of variance ($p = 2.2 \times 10^{-16}$). As both assumptions of parametric testing were violated, a non-parametric Aligned Rank Transform (ART) ANOVA was applied instead. The ART ANOVA results showed that heating time had a statistically significant effect on yield percentage ($p = 0.04156$), while HCl concentration did not ($p = 0.2319$). The interaction between HCl concentration and heating time was not statistically significant ($p = 0.6690$), indicating that the effect of heating time on yield did not depend on the HCl concentration used.

Based on the statistical analysis, the yield percentage was significantly influenced by heating time, while HCl concentration did not show a statistically significant effect. According to Zhang *et al.*, increasing HCl concentration and prolonging heating time tend to reduce the resulting yield percentage [15], as strong HCl solutions generate additional H^+ ions that break down glycosidic bonds within the amorphous cellulose structure, with this reactivity accelerating at higher temperatures and longer heating times, enabling deeper depolymerization [16]. In the present study, this concentration-dependent effect was not statistically significant, possibly due to the relatively narrow HCl concentration range tested (1.5-2.5 M). By contrast, the significant effect of heating time observed here is consistent with the mechanism described above, since increased hydrolysis reaction time also promotes cellulose depolymerization. At sufficiently high acid levels or excessive heating durations, the crystalline structure of cellulose can also become damaged, resulting in additional degradation and reduced yield, an effect that may become more pronounced outside the concentration range tested in this study. The recovered mass from yield percentage calculations does not guarantee crystalline material, as it may contain other substances. Therefore, further characterization using Fourier Transform Infrared (FTIR) analysis is required to identify the produced MCC.

Functional Group Analysis

The optimization process of MCC was further evaluated using FTIR spectroscopy. Hit Quality Index (HQI) represents the similarity between spectra through its quantitative value [17], [18]. Average HQI values obtained from the FTIR measurements are presented in Table 2.

Table 2. Average HQI and scoring matrix for MCC hydrolysis

Sample	HCl concentration (M)	Heating time (min)	Average HQI	Scoring matrix
1	1.5	10	633.125 ± 5.33^b	1.00
2		20	638.875 ± 12.79^{ab}	1.95
3		30	643.25 ± 14.23^{ab}	2.67
4	2.0	10	656.125 ± 22.28^a	4.79
5		20	648.625 ± 22.44^{ab}	3.56
6		30	653.5 ± 12.65^{ab}	4.36
7	2.5	10	657.375 ± 13.88^a	5.00
8		20	652 ± 18.10^{ab}	4.11
9		30	637.5 ± 31.42^{ab}	1.72

Notes:

- The reported values represent the mean $\pm$ standard deviation (STDEV) of three replicates which were measured in two repetitions.
- Different superscript letters indicate significant differences based on the Two-way ANOVA test (p -value < 0.05).

Based on Table 2, the highest similarity to the MCC reference database was observed in the sample treated with 2.5 M HCl and a heating time of 10 minutes, yielding an HQI value of 657.375. Meanwhile, the lowest similarity to the MCC template was found in the sample treated with 1.5 M HCl and a heating time of 10 minutes, with an HQI value of 633.125. Interestingly, samples treated with 2.0 M HCl maintained a more consistent HQI pattern, thus outperforming those treated with 1.5 M and 2.5 M HCl.

The HQI serves as an indicator for determining which MCC sample most closely resembles the reference template, with higher HQI values reflecting greater similarity to commercial MCC [19]. Statistical testing confirmed that HCl concentration significantly affected the FTIR-HQI results. At the lowest HCl concentration (1.5 M), an insufficient heating time produced lower HQI values that gradually increased with longer heating times. Conversely, at the highest concentration (2.5 M), the opposite pattern emerged, where an excessively long heating time actually resulted in lower HQI values, which then increased as heating time was reduced. In other words, a higher HCl concentration must be balanced with a shorter heating time, and vice versa, in order to achieve optimal results. Notably, samples treated with 2.0 M HCl still exhibited a more stable HQI pattern than either the 1.5 M or 2.5 M treatments, suggesting that this concentration is less sensitive to variation in heating time.

Prior to hypothesis testing, the FTIR-HQI data were first evaluated for normality and homogeneity of variance. The Shapiro-Wilk test indicated no significant deviation from normality ($p = 0.25$), while Levene's test likewise showed no significant violation of homogeneity of variance ($p = 0.1246$). Therefore, the assumptions for parametric testing were

considered satisfied. A two-way ANOVA was subsequently performed, revealing that HCl concentration had a statistically significant effect on HQI ($p = 0.0255$), whereas heating time did not ($p = 0.7401$). Furthermore, the interaction between HCl concentration and heating time was also not statistically significant ($p = 0.1946$), thus indicating that no meaningful interaction occurred between the two factors, in other words, their combined effect on HQI was no greater than what would be expected from each factor acting independently.

Total Scoring Matrix Analysis

Table 3. Average HQI and scoring matrix for MCC hydrolysis

Sample	HCl concentration (M)	Heating time (min)	Yield percentage score	Average HQI score	Total scoring matrix
1	1.5	10	4.87	1.00	5.87
2		20	4.02	1.95	5.97
3		30	2.99	2.67	5.66
4	2.0	10	5.00	4.79	9.79
5		20	4.47	3.56	8.03
6		30	3.58	4.36	7.94
7	2.5	10	3.56	5.00	8.56
8		20	2.29	4.11	6.41
9		30	1.00	1.72	2.72

Total scoring matrix from MCC acid hydrolysis were achieved through the combination of percentage yield and average HQI analysis results. Scores for yield percentage, average HQI, and their total score to determine the optimum process condition are shown in Table 3. The sample treated using 2.0 M HCl and a heating time of 10 minutes obtained the highest scoring matrix value and was therefore determined as the optimum process parameters.

Validation of the Optimized MCC

While the optimization process revealed that the sample which received 2.0 M HCl treatment during 10 minutes heating time produced the optimum results, validation is needed through characterization using FTIR and XRD and SEM analysis. A reference material, commercially available MCC, Avicel® PH 101, is used to evaluate both characteristics and performance of the manufactured coconut pulp residue MCC samples. The FTIR analysis is conducted to evaluate whether optimized MCC retains its functional groups. XRD analysis was conducted to evaluate the crystallinity of the resulting sample structure, while SEM analysis was used to observe the surface morphology of the optimized MCC.

Functional Group Characterization

FTIR analysis records the characteristic absorption frequencies of functional groups present in the MCC sample. The compound MCC contains specific functional groups which make it unique from all other chemical substances. The FTIR spectrum shows particular peaks which indicate the presence of these functional groups.

Based on Figure 1, several characteristic peaks were observed at wavenumbers around 3350 cm^{-1} , 2900 cm^{-1} , 1750 cm^{-1} , 1650 cm^{-1} , 1430 cm^{-1} , 1370 cm^{-1} , 1100 cm^{-1} , and 895 cm^{-1} . These peaks correspond to the characteristic functional groups of MCC, as summarized in Table 4.

Table 4. FTIR peak range for synthesized MCC and Avicel® PH 101.

Peak range (cm^{-1}) MCC	Peak range (cm^{-1}) Avicel® PH 101	Characteristics
3500-3300	3500-3300	Hydroxyl group stretching (-OH)
2900	2900	C-H bond stretching
1750	-	Carbonyl group stretching (C=O)
1650	1650	O-H bond
1400	1430	CH ₂ bond
1370	1370	C-H bond
1200	1100	C-O-C bond
800	895	β -1,4 glycosidic bond

FTIR analysis of the optimized MCC (2.0 M HCl, 10 minutes) revealed characteristic absorption bands throughout the spectrum. The wide band between 3350–3300 cm^{-1} shows the stretching motion of cellulose hydroxyl (-OH) groups, confirming hydrogen bonding between cellulose chains. The peak at 2900 cm^{-1} results from C-H stretching vibrations in the cellulose backbone [20]. A carbonyl absorption band was also observed near 1750 cm^{-1} , indicating C=O stretching. The band at 1650 cm^{-1} shows O-H bending vibrations from water molecules chemically attached to the sample [21]. In the fingerprint region, the peak at 1430–1400 cm^{-1} shows symmetric CH₂ bending vibrations associated with cellulose crystallinity [22], the band near 1370 cm^{-1} reflects C-H bending vibrations [23], the region at 1200-1050 cm^{-1} shows

asymmetric C-O-C stretching of the pyranose ring, confirming the glucose ring structure [24], and the β -1,4-glycosidic bond appears as a peak between 895–800 cm^{-1} , confirming the cellulose structure [25].

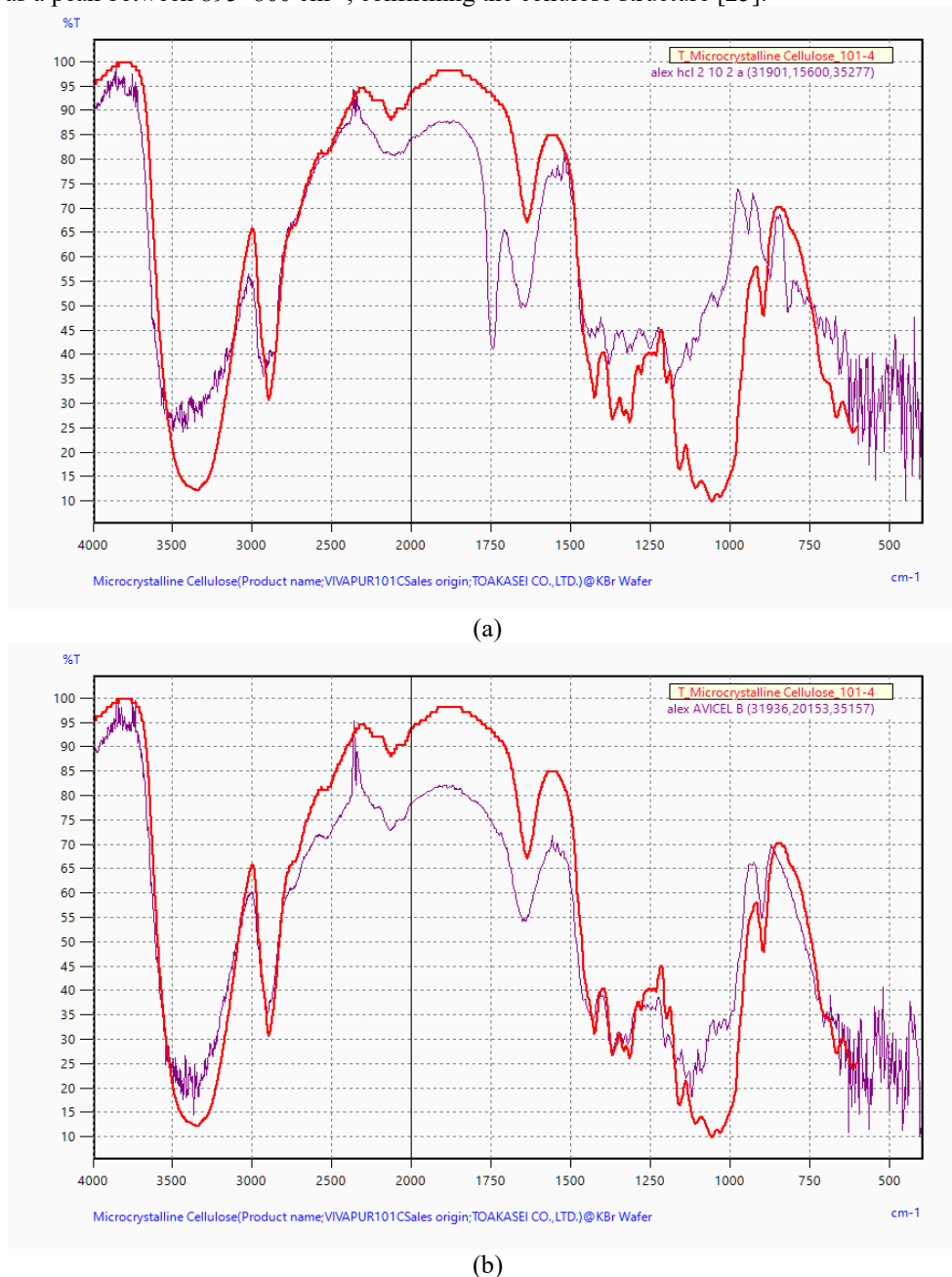


Figure 1. FTIR spectra of MCC produced under 2.0 M HCl and 10 minutes hydrolysis (a) and commercial MCC Avicel® PH 101 (b).

Most of these bands were also present in Avicel® PH 101, confirming that the synthesized MCC retains the expected cellulose functional-group profile. However, two differences were observed: the 1750 cm^{-1} carbonyl band was present in the synthesized MCC but notably absent in Avicel® PH 101, and the synthesized MCC showed lower relative peak intensity at both 1430 cm^{-1} and 895 cm^{-1} compared to Avicel® PH 101, which displayed more distinct, higher-intensity peaks at these positions.

The carbonyl band near 1750 cm^{-1} can most likely be attributed to acetyl and uronic ester groups associated with residual hemicellulose, as well as ester linkages originating from incompletely removed lignin fractions [22], [23]. In commercial MCC such as Avicel® PH 101, these non-cellulosic components are typically eliminated through more extensive delignification and bleaching, which explains the absence of this signal in the reference spectrum. Its presence in the synthesized MCC therefore suggests that small amounts of hemicellulose and/or lignin persisted despite the applied chemical treatment. Together with the lower peak intensities at 1430 cm^{-1} and 895 cm^{-1} , both literature-associated markers of crystallinity, this residual carbonyl signal points to the same underlying cause: incomplete delignification and bleaching during processing [26], [27]. This is consistent with the lower crystallinity index discussed in the following section.

Crystallinity Characterization

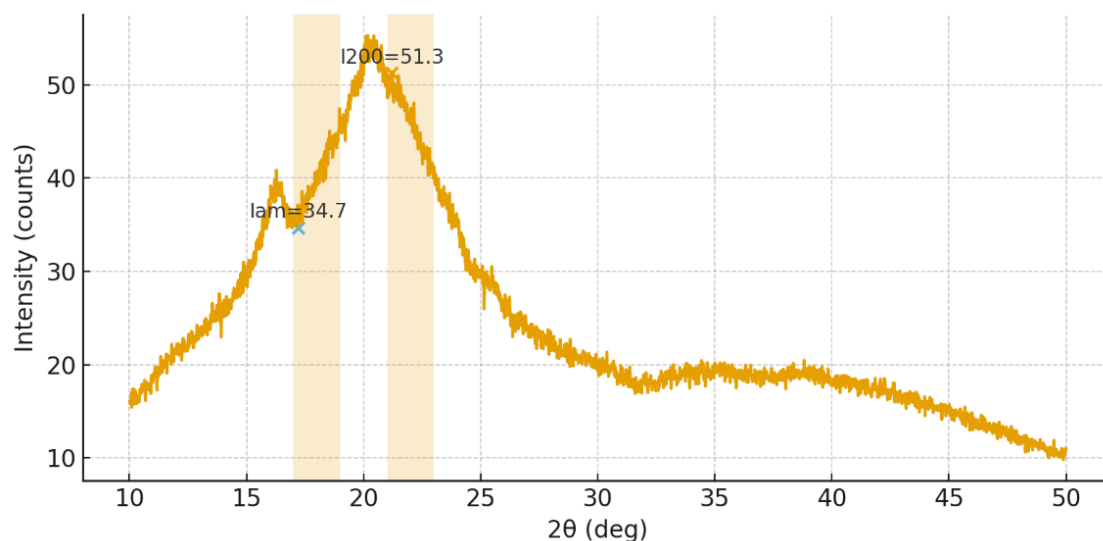


Figure 2. XRD spectra of MCC produced under 2.0 M HCl and 10 minutes hydrolysis.

The XRD pattern in Figure 2 displays broad and not very distinct diffraction peaks because the MCC sample contains a relatively limited proportion of crystalline material. The extent of this crystalline content is typically quantified using the crystallinity index (CrI), a parameter that reflects the degree of structural ordering within the sample. The crystallinity index results from analysing the main crystalline peak intensity (I_{200}) which exists between 2θ angles of $19\text{--}22^\circ$ through the Segal equation (Equation 2), and it was found that the calculated CrI value for the synthesized MCC was 37.25%.

This comparatively low CrI value indicates that the acid hydrolysis conditions applied in this study were insufficient to effectively remove the amorphous regions of cellulose, which are generally more susceptible to acid attack than the crystalline regions [28]. Consequently, a substantial proportion of amorphous cellulose likely remained intact alongside the crystalline domains, lowering the overall CrI. Several processing factors may have contributed to this outcome, including insufficient HCl concentration, inadequate hydrolysis duration or temperature, or incomplete penetration of the acid into the fiber structure due to residual lignin or hemicellulose from the pre-treatment stage [29].

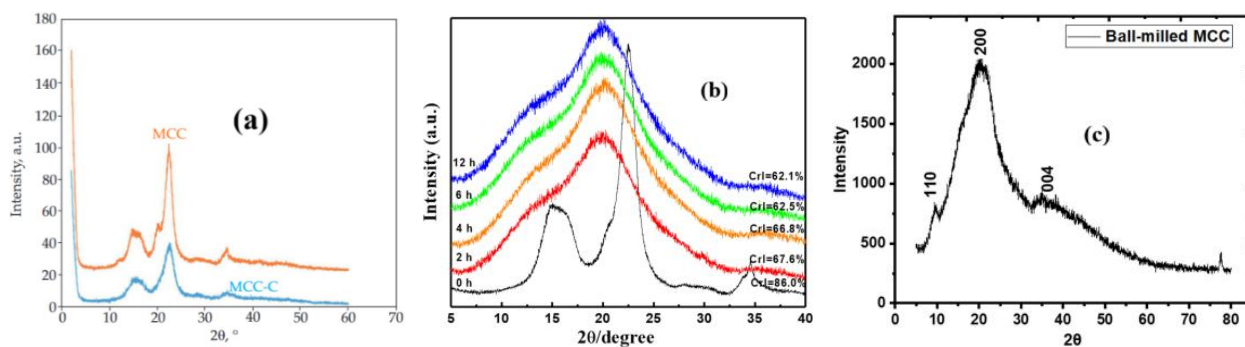


Figure 3. Comparison of XRD spectra of MCC from previous studies: Abdullah *et al.* (a), Wang *et al.* (b), Faye *et al.* (c).

The calculated CrI value for the synthesized MCC is considerably lower than the value reported by Abdullah *et al.*, who obtained a crystallinity index of 71.8% [30]. This difference is also reflected in the variation of peak intensities presented in Figure 3. For comparison, commercial MCC Avicel® PH 101 product shows a crystallinity index in the range of 80–90%. Beyond the hydrolysis conditions, the crystallinity index also appears to be sensitive to differences in pre-treatment method across studies. Also, according to previous research by Wang *et al.*, mechanical grinding using a ball mill as a pre-treatment step may reduce the crystallinity of cellulose [31]. The extended milling duration causes damage to the crystalline structure which results in reduced crystallinity. This phenomenon is illustrated in Figure 3(b) and is consistent with similar observations reported by Faye *et al.*, as shown in Figure 3(c) [32].

Nevertheless, this lower crystallinity does not undermine the selection of 2.0 M HCl and 10 minutes as the optimum hydrolysis condition, since this condition was determined based on yield and FTIR-HQI performance rather than crystallinity. Instead, it identifies crystallinity enhancement as a specific area for further optimization in future work.

Morphology Characterization

The SEM analysis enabled the morphological assessments of MCC for quality evaluation. The MCC sample images were taken at two different magnification levels which were 500x and 1000x to observe the particle shapes and surface details as shown in Figure 4.

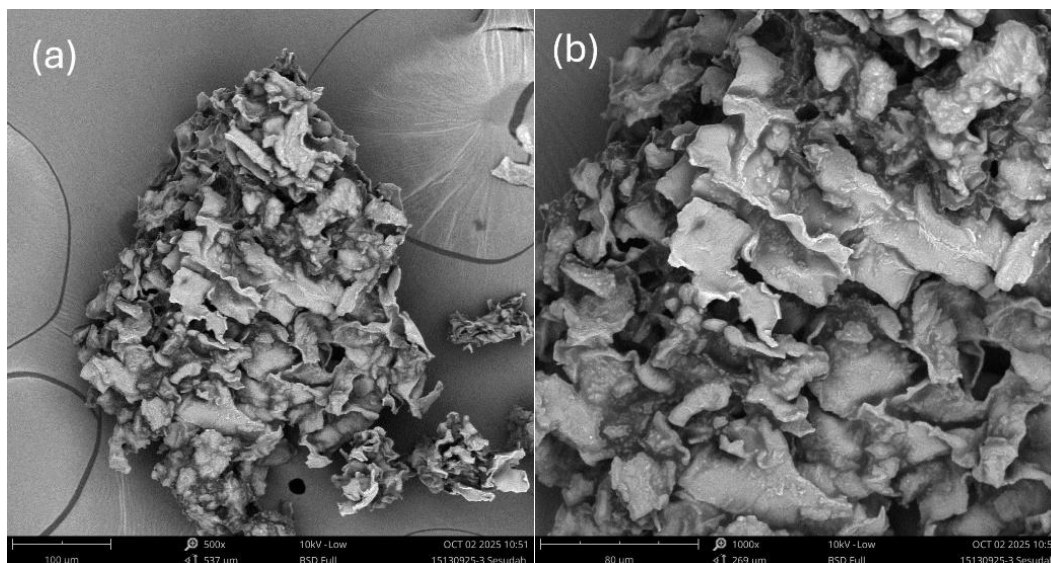


Figure 4. SEM image of MCC produced under 2.0 M HCl and 10 minutes hydrolysis under 500x Magnification (a) and 1000x Magnification (b).

SEM imaging of the optimized MCC (Figure 4) shows an irregular surface composed of sheet-like, folded structures. Particle length was estimated at 20-60 μm , with fragment pieces of 5-15 μm . The acid hydrolysis treatment caused the amorphous regions of cellulose to break down, forming these irregular microcrystals and removing most non-cellulosic components, including lignin and hemicellulose.

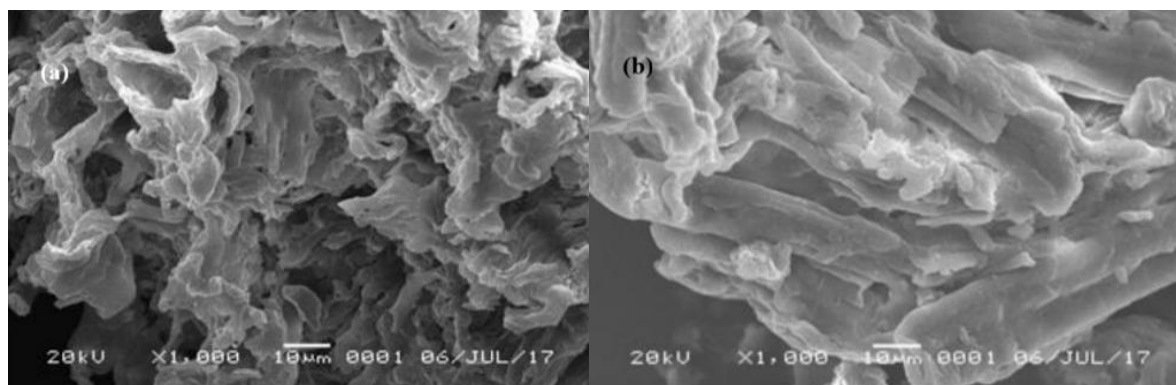


Figure 5. SEM image of MCC of previous studies: MCC from corn cobs found in previous study by Kunusa *et al.* (a) and Avicel® PH 101 MCC (b).

According to Abdullah *et al.*, native cellulose fibers typically retain a tubular or elongated cylindrical structure [30], the synthesized MCC no longer exhibits this shape, consistent with successful hydrolysis. In contrast, Avicel® PH 101 (Figure 5(b)) showed a more uniform morphology, with an estimated particle length of 8-20 μm and width of approximately 1-3 μm , smaller and more regular than the synthesized MCC. This folded morphology also resembles that reported for corn-cob-derived MCC by Kunusa *et al.* (Figure 5(a)) [33].

Based on these physical characteristics, the sample shown in Figure 4 can be identified as MCC. However, its surface morphology does not closely resemble that of Avicel® PH 101, suggesting that the synthesized MCC may still contain residual amorphous regions, consistent with its lower crystallinity index reported above. This larger, less uniform particle size relative to Avicel® PH 101 [8] also helps explain the surface texture differences observed in the preliminary bioplastic application discussed in the following section.

Nevertheless, the optimized MCC (2.0 M HCl, 10 minutes) shows a morphology broadly consistent with reference MCC structures, as its irregular, folded microcrystal formation resembles both Avicel® PH 101 and the corn-cob-derived MCC reported by Kunusa *et al.*, despite some differences in particle size, this supports the selection of 2.0 M HCl and 10

minutes as an appropriate condition for producing recognizable MCC, even though further refinement of particle uniformity could be pursued in future work.

Preliminary Application on Bioplastic

As a preliminary exploration of potential application, MCC was incorporated into a bioplastic mixture to produce two different bioplastic compositions, as shown in Figure 6. It should be noted that this assessment is qualitative and visual in nature. No quantitative mechanical measurements (e.g., tensile strength) were performed in this study, and the observations below are intended only as an initial indication of feasibility.

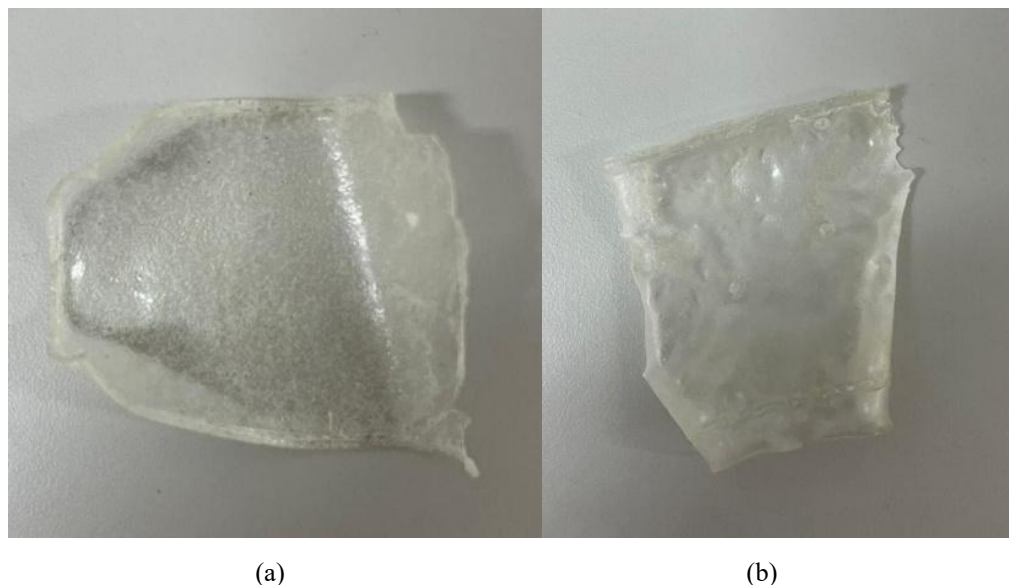


Figure 6. Bioplastic reinforced with coconut pulp residue MCC (a), with Avicel® PH 101 MCC (b).

Based on Figure 6(a), the bioplastic containing the synthesized MCC exhibits a relatively rough surface texture. This is likely due to the larger particle size and lower fineness of the MCC sample, which contributes to a coarse texture on the upper surface. However, the surface that was in contact with the acrylic casting plate appears smoother. The drying process causes water to evaporate mainly from the top surface which explains this difference. Consequently, larger and coarser MCC particles become more prominent on the top surface of the bioplastic.

The bioplastic in Figure 6(b) shows a uniform texture because it contains Avicel® PH 101 MCC as its reinforcement agent. This is likely due to the finer and more homogeneous particle size of Avicel® PH 101 compared to the synthesized MCC sample. The drying process of bioplastic material does not result in any surface appearance changes according to Arun *et al.* [8].

The observed texture of the bioplastics is consistent with the particle size estimations obtained from the SEM analysis (Figure 4(b), SEM of MCC at 1000x and Figure 5(b), SEM of Avicel-MCC template at 1000x), which showed that the synthesized MCC particles are up to three times larger than those of Avicel®. The surface of bioplastics becomes rougher when MCC particles increase in size compared to Avicel® PH 101.

CONCLUSION

This study demonstrated that HCl concentration had a significant effect on FTIR-HQI, however, yield percentage did not show a significant response to this parameter. Heating time significantly affected yield percentage, while HQI did not exhibit a significant response to changes in heating time. Yield percentage generally declined with longer heating durations, consistent with increased depolymerization of amorphous cellulose regions under prolonged acid exposure. The relationship between HCl concentration, heating time, and HQI proved more complex, at the lowest concentration (1.5 M), HQI increased with longer heating times, whereas the opposite trend occurred at the highest concentration (2.5 M), where HQI decreased as heating time increased. The 2.0 M HCl treatment produced the most stable HQI pattern across all heating durations, suggesting greater tolerance to variation in processing time. Taking both yield percentage and FTIR-HQI into account through the scoring matrix, 2.0 M HCl with a 10-minute heating time was identified as the optimum hydrolysis condition. Subsequent validation confirmed that the optimized MCC retains the characteristic functional groups of cellulose and exhibits a morphology broadly consistent with reference MCC, despite showing a relatively low crystallinity index (37.25%) and larger particle size compared to commercial Avicel® PH 101. These findings suggest that although the selected condition gives a good balance between yield percentage and FTIR-HQI, further improvements to the hydrolysis and pre-treatment steps could help increase the material's crystallinity and produce more uniform particles. A preliminary visual assessment also showed that the synthesized MCC could work as a reinforcement agent in bioplastic, and future studies should include mechanical testing to evaluate its performance more thoroughly.

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