



Green Extraction of Microcrystalline Cellulose from Cabbage Waste (*Brassica Oleracea L.*) Using Steam Explosion and Low-Concentration Chemical Treatment



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Abstract

The increasing generation of agricultural waste presents both environmental challenges and opportunities for the development of sustainable materials. This study explores the extraction of microcrystalline cellulose (MCC) from cabbage waste using a combination of steam explosion and low-concentration chemical treatments to optimize lignocellulose degradation. The process involved sequential delignification with NaOH, bleaching with NaOCl, and steam explosion applied to samples that had been soaked in oxalic acid at varying concentrations (0%–2%). After drying, the samples were analyzed using FTIR, XRD, and SEM to evaluate the extracted MCC product. The analytical results showed that the applied method significantly increased cellulose purity, from 31.05% in untreated fibers to 69.88% after steam explosion following soaking in 2% oxalic acid. FTIR analysis confirmed the removal of lignin and hemicellulose, while XRD analysis indicated an increase in crystallinity from 39% to 57%, suggesting improved structural integrity. SEM analysis revealed enhanced fiber separation and reduced particle size, indicating efficient defibrillation. These results highlight the potential of an environmentally friendly approach to producing high-quality MCC, supporting green chemistry principles and sustainable development goals (SDGs). The extracted MCC holds promising applications, particularly as a biopolymer for drug delivery systems, polymer composites, and food additives.

Keywords: Agricultural waste; Cabbage waste; Microcrystalline cellulose; Steam explosion; Sustainable materials

1. Introduction

The production of agricultural products in Indonesia continues to increase, and this growth is directly proportional to the amount of agricultural waste generated. Agricultural waste consists of residues and by-products from agricultural activities that are not utilized for commercial purposes, including crop residues, livestock waste, agrochemical waste, food processing waste, and other by-products. These wastes represent a renewable resource that can be utilized to produce various bio-based products, thereby reducing environmental impact and promoting a more sustainable agricultural sector. Unfortunately, the potential of agricultural waste has not been fully optimized. Therefore, the industrialization of agricultural products and their associated waste through the implementation of a bioeconomy approach is an inevitable necessity for Indonesia [1],[2].

Agricultural residues are abundant and serve as a valuable alternative for producing high-value products. One such product is microcrystalline cellulose (MCC), which is extracted from lignocellulosic materials. MCC is widely used as a natural additive in the development of bio-composite films, enhancing their mechanical properties and functionality. The utilization of MCC from agricultural waste not only adds economic value but also contributes to sustainable material development by reducing environmental impact and promoting the circular bioeconomy [3][4]. Agricultural waste, such as cabbage residue, presents a sustainable and cost-effective source for producing cellulose. Cabbage waste contains 31–67% cellulose, polysaccharides, and minerals, which can be utilized for nanocellulose extraction [5]. Microcrystalline cellulose (MCC) extracted from natural fibers is widely utilized in various industries, including pharmaceuticals, food, cosmetics, and polymer composites, due to its exceptional properties. These properties include non-toxicity, biocompatibility, hydrophilicity, biodegradability, crystallinity, and renewability, making MCC a sustainable and eco-friendly material. In the pharmaceutical industry, MCC is commonly used as an

excipient in tablet formulations. In the food industry, it serves as an emulsifier, stabilizer, or thickening agent. Additionally, MCC is incorporated into cosmetic formulations for skincare and personal care products. In polymer composites, MCC functions as a reinforcing filler to enhance mechanical properties. Due to its versatile applications and environmentally friendly nature, MCC holds great potential for sustainable material development [6][7]. This approach aligns with the Sustainable Development Goals (SDGs), promoting green chemistry, waste recycling, and bio-based industries [8].

Various methods have been explored for nanocellulose extraction, including mechanical, chemical, and hybrid techniques [9]. Among various isolation methods, acid hydrolysis is the most widely used due to its relatively low temperature and pressure requirements [10]. However, one drawback of this method is the use of high concentrations of strong acids, which can cause environmental pollution and pose corrosion risks. Furthermore, the yield of nanocellulose obtained using this method is relatively low, around 30–45% [11]. From the perspective of environmental and technical feasibility, the development of environmentally friendly and energy-efficient technologies is necessary. Steam explosion, when combined with chemical treatments, is a promising method for lignocellulose breakdown. This process induces defibrillation, fragmenting lignin and hemicellulose polymers into smaller units through hydrolysis and radical reactions [12]–[13]. Notably, steam explosion offers several advantages, such as low energy consumption, minimal chemical usage, reduced environmental impact, and cost efficiency. Although NaOH, NaOCl, and oxalic acid are used, their concentrations are kept low to minimize environmental impact.

In this study, we aim to characterize microcellulose extracted from cabbage waste using a combination of steam explosion and low-concentration organic acid treatment. This hybrid approach leverages the efficiency of steam explosion as a mechanical pretreatment while minimizing chemical consumption to develop an environmentally friendly and cost-effective nanocellulose production process.

2. Materials and Method

2.1 Material

All chemicals used in this study were of analytical grade and were utilized as received without further purification. The raw material consisted of the outer leaves of cabbage waste (*Brassica oleracea* L.), a cellulose-rich agricultural byproduct collected from Pasuruan, East Java, Indonesia. The bleaching and delignification processes were conducted using sodium hypochlorite (NaOCl) from Chemical Indonesia Multi Sentosa and sodium hydroxide (NaOH) (for analysis, CAS No. 1310-73-2) from Merck. Oxalic acid ($C_2H_2O_4$) (for analysis, CAS No. 6153-56-6) from Merck was employed as the organic acid reagent for low-concentration chemical treatment.

2.2 Extraction of Microcrystalline Cellulose

Cabbage waste is chopped into approximately 5 cm pieces before being placed into a blender. Then, water is added in a 1:2 (w/v) ratio of cabbage waste to the blending process. The mixture is blended at high speed. After blending, the mixture is washed and filtered. The resulting blend is then dried in an oven at 60°C for 8 hours. Once dried, the cabbage waste is ground into powder to ease the subsequent delignification process. 50 grams of cabbage powder is treated with 500 ml of 1% NaOH solution. The delignification process is carried out at 80°C for 120 minutes, followed by filtration. Bleaching is performed using a 2% NaOCl solution at 80°C for 60 minutes until the solid material turns white. The material is then washed with deionized water until the pH reaches neutral and dried in an oven at 60°C for 8 hours. 3 grams of the sample is treated with oxalic acid concentrations of 0%, 0.5%, 1%, 1.5%, and 2%. The sample is subjected to steam explosion at various temperatures of 130°C for 15 minutes. After the treatment, the solid residue is washed with deionized water until the pH is neutral (pH 7) and filtered. The resulting material is dried in an oven at 60°C.

2.3 Characterization

- Lignocellulose Composition Analysis (Chesson Method)

Determination of lignocellulosic components including cellulose, hemicellulose, and lignin was carried out using the Chesson method (1981):

$$\%Cellulose = \frac{c - d}{a} \times 100\% \quad (1)$$

$$\%Lignin = \frac{d - e}{a} \times 100\% \quad (2)$$

$$\%Hemicellulose = \frac{b - c}{a} \times 100\% \quad (3)$$

Where,

- a = initial dry weight of the sample
- b = weights of the sample with hot water
- c = weights of the sample after H₂SO₄ treatment 1 N
- d = weights of the sample after H₂SO₄ treatment 2%
- e = weights of the sample ash

- **Fourier Transform Infrared Spectroscopy (FTIR)**

Fourier Transform Infrared Spectroscopy (FTIR) analysis was performed using an Agilent Cary 630 spectrometer over a wavenumber range of 400–4000 cm⁻¹ to identify functional groups and confirm the removal of lignin and hemicellulose.

- **X-ray Diffraction (XRD)**

X-ray Diffraction (XRD) analysis was conducted using a Rigaku Miniflex 600 diffractometer equipped with Cu-K α radiation ($\lambda = 1.5406 \text{ \AA}$), operating at 40 kV and 15 mA. Scans were performed over a 2θ range of 5° to 45° at a rate of 2°/min. The Crystallinity Index (CrI) was calculated using the Segal method, based on the equation:

$$CrI (\%) = \frac{I_{002} - I_{am}}{I_{002}} \times 100 \quad (4)$$

where I_{002} is the peak intensity at $2\theta \approx 22\text{--}23^\circ$ (crystalline region), and I_{am} is the intensity at $2\theta \approx 15\text{--}18^\circ$ (amorphous region).

- **Scanning Electron Microscopy (SEM)**

Scanning Electron Microscopy (SEM) analysis was conducted using a Hitachi FlexSEM 1000 (version 01-10). Prior to imaging, samples were gold-coated. Micrographs were captured at 100 \times magnification to observe surface morphology and fibrillation behavior of the extracted cellulose.

3. Results and Discussion

3.1 Composition Analysis

The cabbage fiber used in this study primarily consists of cellulose, hemicellulose, and lignin. To optimize the cellulose extraction process, the removal of lignin, hemicellulose, ash, and other components was carried out through pre-treatment, delignification, and bleaching processes.

Table 1. Chemical composition of fibers after pre-treatment, delignification, and bleaching.

Process	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Other Component (%)
Native	29.53	23.83	13.08	33.56
Pre Treatment	31.05	16.80	1.02	51.13
Delignification	41.60	19.65	0.38	38.37
Bleaching	44.13	20.10	0.31	35.46

The analysis of chemical composition at each stage of the process indicates significant changes associated with the cellulose extraction from cabbage waste. At the pre-treatment stage, cellulose content remains relatively low (31.05%), while hemicellulose (16.80%) and lignin (1.02%) are not substantially degraded, indicating that this stage serves as an initial preparation step to soften the material's structure and facilitate the subsequent delignification process. After delignification, the fiber color changes to dark brown due to the degradation of lignin compounds in the fibers. Although delignification reduces lignin content, residual lignin remains and contributes to the dark color of the fibers. Lignin contains chromophores that absorb light, producing a dark brown or yellowish color [14]. At this stage,

cellulose content increases from 31.05% to 41.60%, while lignin content, which is highly soluble in NaOH, decreases significantly from 1.02% to 0.38%. This is due to NaOH's ability to break the bonds between lignin and cellulose. Lignin is linked through ester bonds that form a complex matrix reinforcing the plant cell wall. The alkaline conditions during delignification hydrolyze these bonds, allowing lignin fragments to be released into the solution [15]. The breakdown of ester bonds also destabilizes lignin, which subsequently degrades into small phenolic compounds. These fragments dissolve in the alkaline medium and can be rinsed away

Following delignification, which removes lignin, the bleaching process produces the color shown changes from the brownish color after delignification to white. This process further increases the cellulose content to 44.13% and reduces the lignin content to 0.31%. The bleaching process uses oxidizing agents to break the bonds in lignin into water-soluble compounds, such as aromatic aldehydes and carboxylic acids. These compounds are subsequently removed, resulting in whiter and purer fibers. By removing lignin, the fibers become purer, and the brightness or whiteness of the cellulose increases. Lignin is the primary cause of the brown color in fibers, so its removal results in whiter fibers [16]. However, the hemicellulose content increases during the delignification and bleaching processes. This is because the removal of lignin and other components makes the hemicellulose appear more dominant in percentage terms. Additionally, factors such as trapped hemicellulose exposure, detection of degradation products, and analytical methods also contribute to the observed increase in hemicellulose content. Jongaroontaprangsee et al., reported that alkaline treatments disrupt lignin–carbohydrate linkages, leading to partial solubilization of lignin and pectin, while hemicelluloses, though partially degraded, may become more detectable due to the exposure of previously trapped fractions. In addition, factors such as the release of bound hemicellulose, detection of hydrolysis products, and variations in analytical methods may also contribute to the apparent increase in hemicellulose content [17].

Table 2. Chemical composition of fibers after steam explosion.

Process	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Other Component (%)
Bleaching	44.13	20.10	0.31	35.46
OA 0% SE 130°C	45.65	19.85	0.28	34.22
OA 0.5% SE 130°C	48.10	19.10	0.26	32.54
OA 1% SE 130°C	56.95	18.38	0.18	24.49
OA 1.5% SE 130°C	67.90	17.95	0.15	14.00
OA 2% SE 130°C	69.88	16.58	0.11	13.43

The steam explosion process aims to remove hemicellulose, part of the lignin, and residual impurities in the fibers. In this process, the fibers are treated with high-pressure steam, followed by a rapid pressure drop to atmospheric levels, causing explosive decompression and hemicellulose degradation. To evaluate the effect of the steam explosion process, the initial composition of cabbage waste was analyzed prior to the treatment. After the pre-treatment, delignification, and bleaching steps, the cellulose content increased from 31.05% (pre-treatment) to 44.13%, while hemicellulose and lignin contents were reduced to 20.10% and 0.31%, respectively (Table 1.).

From Table 2. it is shown that the steam explosion process also drastically reduced the hemicellulose and lignin content to 19.85% and 0.28%, respectively, demonstrating the effectiveness of the process in removing lignin. When the OA concentration increased to 0.5%, the cellulose content slightly increased to 48.10%, accompanied by a slight decrease in hemicellulose (19.10%) and lignin (0.26%). Further increases in OA concentration to 1% resulted in a cellulose content of 56.95%, with a more significant decrease in hemicellulose and lignin, to 18.38% and 0.18%, respectively. At an OA concentration of 1.5%, the cellulose content rose sharply to 67.90%, while hemicellulose and lignin continued to decrease to 17.95% and 0.15%, respectively. The maximum increase occurred at an OA concentration of 2%, where the cellulose content reached 69.88%, while hemicellulose and lignin were reduced to 16.58% and 0.11%, respectively.

This process demonstrates that increasing the oxalic acid concentration in the steam explosion process enhances the isolated cellulose content, while hemicellulose and lignin undergo further degradation. This aligns with the findings of Chavez et al. (2023), where oxalic acid can catalyze the hydrolysis process and cellulose isolation, facilitating its separation from fibers. Oxalic acid depolymerizes and attacks long cellulose chains, resulting in molecules with a lower degree of polymerization indicating shorter cellulose chain lengths. This process not only breaks down the cellulose

structure but also modifies its surface by introducing carboxyl functional groups ($-\text{COOH}$), which can alter the chemical and physical properties of the cellulose. Therefore, oxalic acid plays a significant role in modifying cellulose in terms of both structure and surface properties [18].

The steam explosion process also contributes to the hydrolysis of glycosidic bonds in hemicellulose, meaning the breakdown of sugar molecule bonds within hemicellulose. Additionally, this process breaks the bonds between hemicellulose and lignin, leading to increased hemicellulose solubility in water (solubilization) [12].

3.2 Functional Group Analysis Using Fourier transform infrared spectroscopy (FTIR)

The FTIR spectrum in Figure 1. shows changes in the chemical characteristics of nanocellulose derived from cabbage waste with different treatment concentrations (0%, 0.5%, 1%, 1.5%, and 2%). FTIR spectra were analyzed qualitatively to identify the presence and relative changes of functional groups associated with lignocellulosic components. The peak intensity at 1603 cm^{-1} , corresponding to aromatic $\text{C}=\text{C}$ stretching indicates residual lignin structures. Nonetheless, the persistence of this band confirms that lignin was not entirely removed, even at higher acid concentrations. A noticeable shift in the 1423 cm^{-1} band, assigned to CH_2 vibrations from hemicellulose [19]. implies structural changes associated with hemicellulose degradation

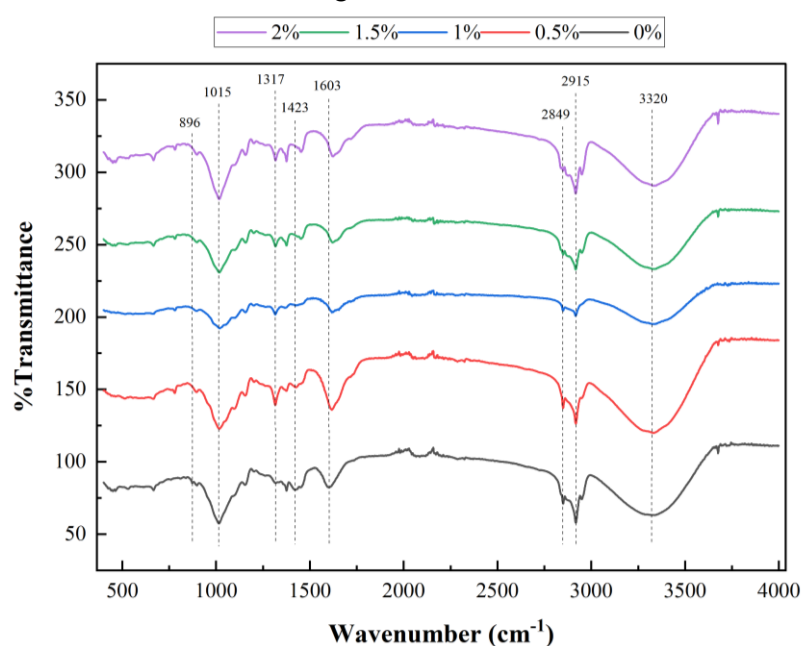


Figure 1. FTIR spectrum results for different oxalic acid concentrations.

Peak at 2915 cm^{-1} , represent aliphatic C-H stretching vibrations [18]. Conversely, peaks at 1015 cm^{-1} and 896 cm^{-1} , indicates the presence of characteristic cellulose groups, such as C-H and C-O-C vibrations in the anhydroglucose ring [20]. This indicates that the core cellulose structure is preserved. This result aligns with the analysis of lignocellulosic components, indicating an increased cellulose content; however, lignin compounds are still retained in the structure.

The intensity of the broad peak at 3320 cm^{-1} , indicating O-H stretching [13],[20], was observed in all samples. Variations in its width and intensity suggest differences in hydrogen bonding interactions and hydroxyl group availability, likely resulting from cellulose purification and internal structural rearrangement.

3.3 Crystallinity Analysis Using X-Ray Diffraction (XRD)

The diffractogram in Figure 2. shows that after the steam explosion treatment with increasing oxalic acid concentrations, each fiber exhibits characteristic diffraction peaks at 2θ values of $15\text{--}18^\circ$ for amorph region and $22\text{--}23^\circ$ for crystalline intensity, corresponding to the lattice planes (I_{am}) and (I_{002}), respectively. This aligns with the findings of Neenu et al. (2022), indicating that these peaks represent the characteristic structure of cellulose type I [21].

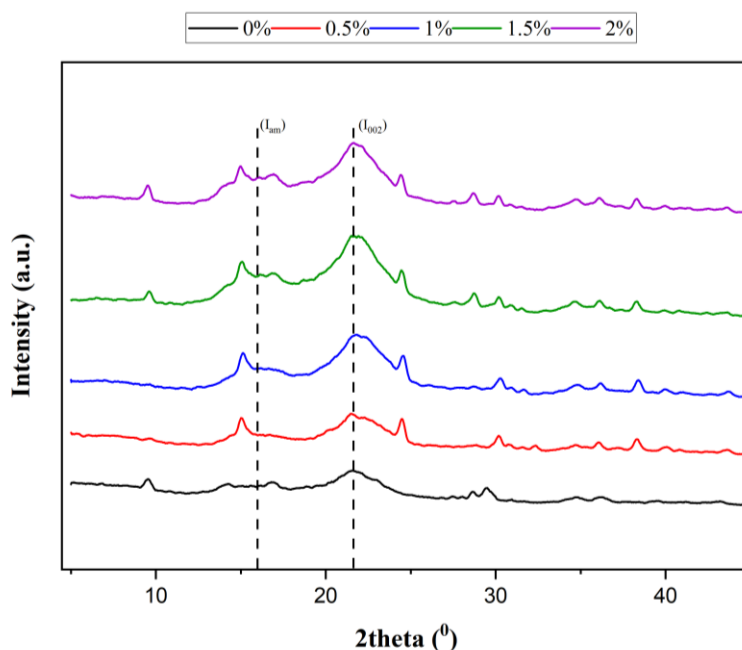


Figure 2. XRD analysis results for different oxalic acid concentrations.

In Figure 3, it is observed that the crystallinity index of cellulose increases significantly from 39% to 57% with rising oxalic acid concentrations from 0% to 2%. This result highlights that higher concentrations of oxalic acid promote more efficient removal of amorphous regions, particularly hemicellulose and lignin, thereby enriching the crystalline regions of cellulose. Oxalic acid acts as a mild organic acid that selectively hydrolyzes the disordered or less-ordered domains in lignocellulosic biomass. As noted by Chavez et al. (2023), hemicellulose and lignin predominantly exist in the amorphous domains of plant cell walls, while the crystalline regions are mainly comprised of tightly packed cellulose chains. These crystalline regions are embedded within a matrix of lignin, hemicellulose, and pectin, which collectively provide structural rigidity to the plant cell wall [18]. During oxalic acid treatment, this surrounding matrix begins to disintegrate as the acid reacts with ester and glycosidic bonds in hemicellulose and lignin. The removal of these matrix components facilitates the liberation of cellulose microfibrils from their entangled state. Furthermore, the action of oxalic acid on cellulose itself targets the amorphous regions, which are chemically less stable and more reactive to acid hydrolysis. The disruption of these regions leads to a reduction in polymer chain length and increases the relative proportion of crystalline domains within the overall cellulose structure [22].

In addition to the chemical role of oxalic acid, the steam explosion process also contributes significantly to enhancing cellulose crystallinity. This process involves subjecting the fiber to high-pressure steam followed by a sudden decompression. This sudden expansion disrupts the cell wall structure through mechanical shear and thermal stress, leading to the rupture of lignin-carbohydrate complexes, increased fiber porosity, and partial defibrillation of cellulose fibers [23]. The induced porosity significantly improves the accessibility of chemical reagents such as oxalic acid into the internal fiber matrix, enabling more efficient penetration, reaction kinetics, and selective hydrolysis of hemicellulose and amorphous cellulose fractions. According to Raju et al. (2023) the steam explosion process significantly enhances the reactivity of cellulose by promoting the hydrolysis of glycosidic linkages specifically within the amorphous regions of the cellulose matrix. These amorphous regions, characterized by their lower degree of polymerization and less-ordered molecular arrangement, exhibit a higher degree of accessibility to water, acid, and thermal degradation, making them more susceptible to cleavage under combined thermochemical treatment. The rapid depressurization during steam explosion creates mechanical shear forces and localized heat gradients that disrupt the hydrogen bonding and polymer chain integrity in these amorphous domains, thereby facilitating their depolymerization and solubilization. In contrast, crystalline cellulose domains, due to their dense packing and strong intermolecular hydrogen bonding. This tightly packed arrangement imparts significant resistance to thermal and chemical degradation, making these domains structurally robust and less prone to hydrolysis under typical pretreatment conditions [13].

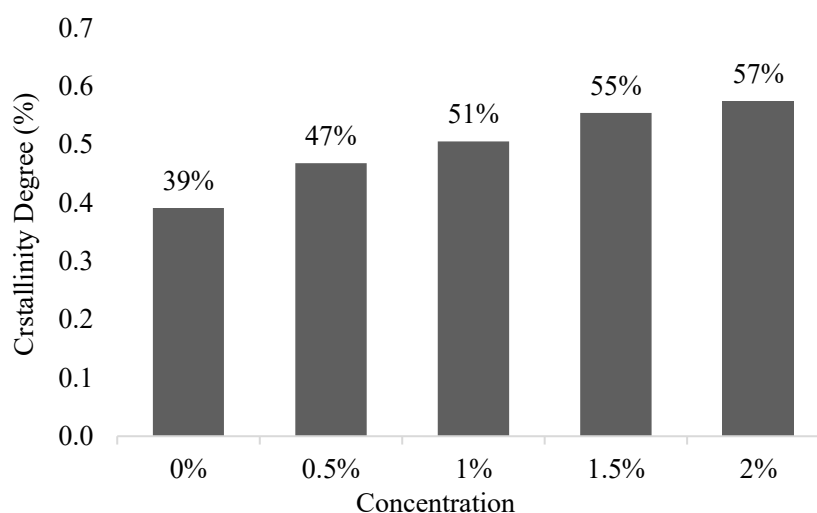


Figure 3. Crystallinity degree diagram for different oxalic acid concentrations.

This synergistic effect between acid hydrolysis and steam explosion results in an overall purification and restructuring of cellulose fibers. As more amorphous regions are degraded and solubilized, the remaining material becomes richer in crystalline domains. The high crystallinity observed at 2% oxalic acid treatment indicates that most of the amorphous matrix has been removed, leaving behind structurally ordered cellulose. As a result, higher oxalic acid concentrations not only accelerate the degradation of non-cellulosic materials but also facilitate the exposure and preservation of the more thermodynamically stable crystalline domains. These findings confirm the synergistic role of acid treatment and steam explosion in enhancing cellulose crystallinity through selective amorphous region removal.

3.4 Morphology Analysis Using Scanning Electron Microscope (SEM)

The surface morphology analysis using Scanning Electron Microscopy (SEM), as shown in Figure 4, reveals significant differences between samples treated with 0% and 2% oxalic acid. At 0% concentration, the particle surface morphology exhibits large and non-uniform structures, with a rough texture and incomplete fibrillation. This indicates that lignin and hemicellulose still dominate the structure, preventing complete cellulose isolation. In contrast, at 2% oxalic acid concentration, the particles appear smaller, more uniform, and exhibit a more distinct fiber structure. The enhanced delignification and bleaching processes with oxalic acid effectively remove lignin and hemicellulose, leading to better fiber isolation. This separation occurs due to the removal of hemicellulose and lignin during the initial treatment stage. Further acid treatment results in a more significant reduction in fiber diameter, attributed to the degradation of the amorphous regions, which are structurally disordered. These amorphous regions act as weak points prone to degradation, promoting fibril separation during acid hydrolysis [24].

At 2% oxalic acid concentration, the isolated cellulose particle size is recorded at 55 μm , indicating that oxalic acid facilitates the breakdown and dissolution of impurities such as hemicellulose and lignin. This enables the cellulose to be more purely isolated and organized into smaller sizes. Oxalic acid acts as an agent that accelerates the breakdown of complex biomass structures, resulting in finer and more well-separated cellulose particles. Conversely, at 0% oxalic acid concentration, the particle size remains larger, measuring 78 μm , due to the absence of oxalic acid's influence, which inhibits the effective breakdown of impurities and cellulose separation. Under these conditions, the cellulose particles remain larger as no chemical agent is present to accelerate the isolation process and reduce particle size.

During acid hydrolysis, the H^+ ions formed in the acidic solution penetrate the glycosidic bonds connecting glucose units in the cellulose polysaccharide chain. This leads to the cleavage of glycosidic bonds, subsequently breaking the long cellulose chains into smaller glucose units. Additionally, acid hydrolysis affects the amorphous zones of cellulose, which have looser bonding compared to crystalline zones. H^+ ions more easily penetrate and disrupt the amorphous zones, causing their separation. Therefore, acid hydrolysis not only cleaves glycosidic bonds but also enhances cellulose solubility by breaking down the more degradable amorphous regions [21].

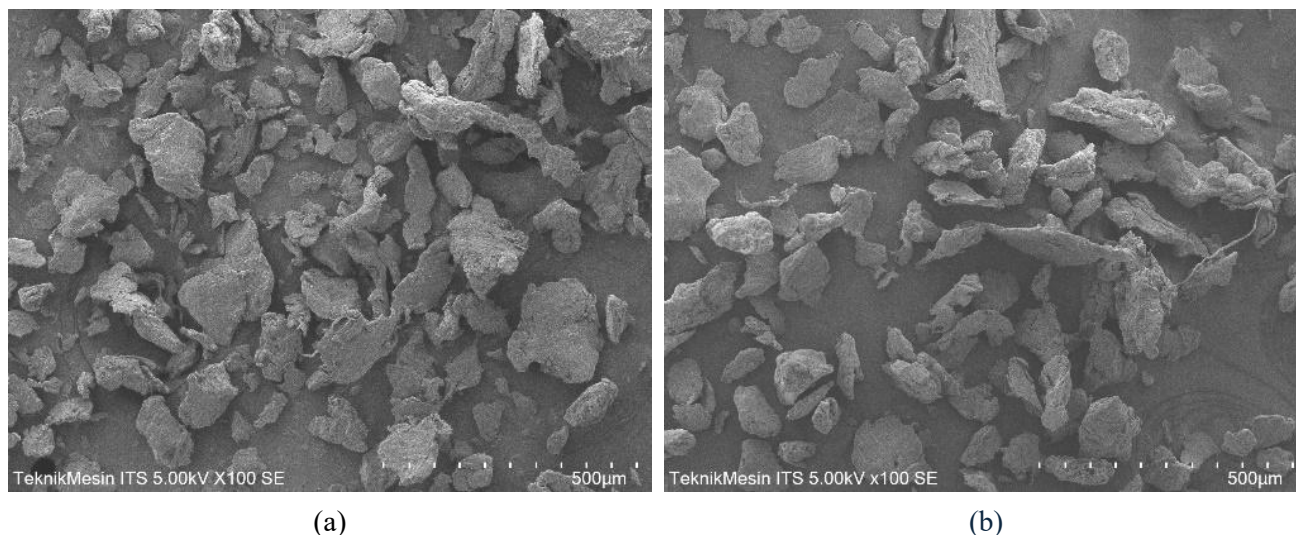


Figure 4. Results of (a) fibers treated with 0% oxalic acid at 130°C, (b) fibers treated with 2% oxalic acid at 130°C.

Under the condition of 2% oxalic acid, although cellulose structures undergo more effective breakdown, the hydrolyzed particles tend to re-agglomerate. This re-agglomeration may result from intermolecular interactions, including Van der Waals forces, hydrogen bonding, or residual impurities that remain within the system. Consequently, instead of dispersing as small individual particles, cellulose fragments tend to cluster into larger aggregates. This phenomenon occurs because the oxalic acid at this concentration promotes more efficient hydrolysis of the amorphous regions, producing smaller cellulose fragments. However, in the absence of sufficient external forces to maintain particle dispersion, these fragments may recombine and form aggregates. Such behavior is commonly observed in fibrous material systems, where newly generated particles tend to re-aggregate due to persistent inter-fiber interactions that have not been entirely disrupted [25].

Agglomeration can also be influenced by the drying process, particularly when excessively high temperatures are applied. Elevated drying temperatures may induce agglomeration of crystalline cellulose as a result of increased inter-particle attractive forces following rapid moisture loss. Moreover, excessively high temperatures pose a risk of degrading the cellulose structure, which can ultimately reduce its degree of crystallinity. According to previous studies, the optimal drying temperature range for crystalline cellulose, to minimize agglomeration while preserving high crystallinity, lies between 50°C and 80°C. Within this range, water evaporation occurs in a controlled manner, reducing the likelihood of particle adhesion and maintaining the integrity of the crystalline structure [26]. Nevertheless, the precise optimal temperature may vary depending on the drying technique employed, such as vacuum oven, spray drying, or freeze drying.

4. Conclusions

The integration of steam explosion with oxalic acid treatment successfully enhanced the extraction and purification of microcrystalline cellulose from cabbage waste. Subsequent application of the steam explosion process, especially when combined with oxalic acid at increasing concentrations, further improved the efficiency of cellulose isolation. The cellulose content peaked at 69.88% with 2% oxalic acid, while hemicellulose and lignin were reduced to 16.58% and 0.11%, respectively. XRD analysis showed a substantial increase in crystallinity index from 39% to 57% as oxalic acid concentration increased, attributable to the removal of amorphous regions and the preservation of crystalline cellulose I structure. FTIR spectroscopy qualitatively confirmed the presence of functional groups

characteristic of cellulose. Morphological observations via SEM revealed a progressive breakdown of fiber structure and improved fibrillation with increasing acid concentration. However, partial agglomeration of cellulose particles was noted at higher oxalic acid levels, likely due to residual inter-fibrillar interactions or drying conditions. Despite this, the overall approach proved effective and environmentally friendly, offering a promising method for converting agricultural waste into value-added cellulose.

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References

- [1] S. C. Z. Desobgo, "Utilization of Agricultural Waste for Water and Wastewater Treatment Processes," vol. 30, no. 1, pp. 295–320, 2024,
- [2] W. BUDHIJANTO, T. ARIYANTO, and R. B. CAHYONO, "Bioenergy Potential from Agricultural Residues and Industrial Wastes in Indonesia," *Journal of Smart Processing*, vol. 8, no. 6, pp. 253–259, Nov. 2019,
- [3] B. Debnath, D. Haldar, and M. K. Purkait, "A critical review on the techniques used for the synthesis and applications of crystalline cellulose derived from agricultural wastes and forest residues," *Carbohydrate Polymers*, vol. 273, p. 118537, Dec. 2021,
- [4] S. Ventura-Cruz and A. Tecante, "Nanocellulose and microcrystalline cellulose from agricultural waste: Review on isolation and application as reinforcement in polymeric matrices," *Food Hydrocolloids*, vol. 118, p. 106771, Sep. 2021,
- [5] D. Khukutapan, N. Chiewchan, and S. Devahastin, "Characterization of Nanofibrillated Cellulose Produced by Different Methods from Cabbage Outer Leaves," *Journal of Food Science*, vol. 83, no. 6, pp. 1660–1667, Jun. 2018,
- [6] M. H. Sainorudin, M. Mohammad, N. H. A. Kadir, N. A. Abdullah, and Z. Yaakob, "Characterization of Several Microcrystalline Cellulose (MCC)-Based Agricultural Wastes via X-Ray Diffraction Method," *Solid State Phenomena*, vol. 280, pp. 340–345, Aug. 2018,
- [7] J. Baruah, R. C. Deka, and E. Kalita, "Greener production of microcrystalline cellulose (MCC) from *Saccharum spontaneum* (Kans grass): Statistical optimization," *International Journal of Biological Macromolecules*, vol. 154, pp. 672–682, Jul. 2020,
- [8] T. Kobayashi and L. Nakajima, "Sustainable development goals for advanced materials provided by industrial wastes and biomass sources," *Current Opinion in Green and Sustainable Chemistry*, vol. 28, p. 100439, Apr. 2021,
- [9] S. Salimi, R. Sotudeh-Gharebagh, R. Zarghami, S. Y. Chan, and K. H. Yuen, "Production of Nanocellulose and Its Applications in Drug Delivery: A Critical Review," 2019.
- [10] K. O. Sodeinde *et al.*, "Cellulose isolated from *Delonix regia* pods: Characterisation and application in the encapsulation of vitamin A," *Industrial Crops and Products*, vol. 160, p. 113138, Feb. 2021,
- [11] Y. Li *et al.*, "Facile extraction of cellulose nanocrystals from wood using ethanol and peroxide solvothermal pretreatment followed by ultrasonic nanofibrillation," *Green Chemistry*, vol. 18, no. 4, pp. 1010–1018, 2016,

- [12] B. M. Cherian, A. L. Leão, S. F. de Souza, S. Thomas, L. A. Pothan, and M. Kottaisamy, "Isolation of nanocellulose from pineapple leaf fibres by steam explosion," *Carbohydrate Polymers*, vol. 81, no. 3, pp. 720–725, Jul. 2010,
- [13] V. Raju *et al.*, "Isolation and characterization of nanocellulose from selected hardwoods, viz., *Eucalyptus tereticornis* Sm. and *Casuarina equisetifolia* L., by steam explosion method," *Scientific Reports*, vol. 13, no. 1, p. 1199, Jan. 2023,
- [14] A. Rahayu, F. F. Hanum, N. A. Z. Amrillah, L. W. Lim, and S. Salamah, "Cellulose Extraction from Coconut Coir with Alkaline Delignification Process," *Journal of Fibers and Polymer Composites*, vol. 1, no. 2, pp. 106–116, Oct. 2022,
- [15] R. S. Abolore, S. Jaiswal, and A. K. Jaiswal, "Green and sustainable pretreatment methods for cellulose extraction from lignocellulosic biomass and its applications: A review," *Carbohydrate Polymer Technologies and Applications*, vol. 7, p. 100396, Jun. 2024,
- [16] T. Wang and Y. Zhao, "Optimization of bleaching process for cellulose extraction from apple and kale pomace and evaluation of their potentials as film forming materials," *Carbohydrate Polymers*, vol. 253, p. 117225, Feb. 2021,
- [17] S. Jongaroontaprangsee, N. Chiewchan, and S. Devahastin, "Production of nanofibrillated cellulose with superior water redispersibility from lime residues via a chemical-free process," *Carbohydrate Polymers*, vol. 193, 2018,
- [18] B. K. Chavez, K. Garces-Porras, D. C. Parada, and E. Flores, "Thermochemical isolation and characterization of nanofibrillated cellulose from *Stipa obtusa* fibers," *Carbohydrate Polymer Technologies and Applications*, vol. 6, p. 100344, Dec. 2023,
- [19] A. Pirozzi, F. Olivieri, R. Castaldo, G. Gentile, and F. Donsì, "Cellulose Isolation from Tomato Pomace: Part II—Integrating High-Pressure Homogenization in a Cascade Hydrolysis Process for the Recovery of Nanostructured Cellulose and Bioactive Molecules," *Foods*, vol. 12, no. 17, p. 3221, Aug. 2023,
- [20] M. G. Thomas, E. Abraham, P. Jyotishkumar, H. J. Maria, L. A. Pothan, and S. Thomas, "Nanocelluloses from jute fibers and their nanocomposites with natural rubber: Preparation and characterization," *International Journal of Biological Macromolecules*, vol. 81, pp. 768–777, Nov. 2015,
- [21] K. V. Neenu *et al.*, "Effect of oxalic acid and sulphuric acid hydrolysis on the preparation and properties of pineapple pomace derived cellulose nanofibers and nanopapers," *International Journal of Biological Macromolecules*, vol. 209, pp. 1745–1759, Jun. 2022,
- [22] T. M. Lacerda, M. D. Zambon, and E. Frollini, "Oxalic acid as a catalyst for the hydrolysis of sisal pulp," *Industrial Crops and Products*, vol. 71, 2015,
- [23] S. Bandyopadhyay-Ghosh, S. B. Ghosh, and M. Sain, "The use of biobased nanofibres in composites," in *Biofiber Reinforcements in Composite Materials*, 2015.
- [24] C. J. Chirayil, J. Joy, L. Mathew, M. Mozetic, J. Koetz, and S. Thomas, "Isolation and characterization of cellulose nanofibrils from *Helicteres isora* plant," *Industrial Crops and Products*, vol. 59, pp. 27–34, Aug. 2014,
- [25] L. Solhi *et al.*, "Understanding Nanocellulose-Water Interactions: Turning a Detriment into an Asset," 2023.
- [26] Y. Peng, D. J. Gardner, Y. Han, A. Kiziltas, Z. Cai, and M. A. Tshabalala, "Influence of drying method on the material properties of nanocellulose I: thermostability and crystallinity," *Cellulose*, vol. 20, no. 5, pp. 2379–2392, Oct. 2013,