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MICROCRYSTALLINE CELLULOSE PRODUCTION FROM SUGARCANE BAGASSE AS SUSTAINABLE PROCESS: A PILOT PLANT

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Abstract

This article overviews the microcrystalline cellulose (MCC) production process from sugarcane bagasse on a pilot scale and compares it with the commercial MCC. MCC is a versatile excipient used in the pharmaceutical, food, and cosmetic industries due to its excellent binding, disintegrating, and bulking properties. The production of MCC involves acid hydrolysis of alpha-cellulose, washing, and post-treatment. The article describes the steps involved in the production process, including preparing raw materials, acid hydrolysis, washing, drying, and particle size reduction. In addition, the article highlights the importance of careful control of process parameters to achieve the desired properties of the MCC. The degree of polymerization, Density (bulk), Crystallinity, Particle size distribution, and Specific surface area of the MCC produced on a pilot scale are 226, 0.27, 83%, 15-200 µm, and 1.2736 m^2/g, respectively. Accordingly, the produced MCC can be used in various pharmaceutical, food, and cosmetic applications, making it a valuable excipient for drug formulation and other applications.

Keyword: Sugarcane Bagasse; Acid Hydrolysis; α-Cellulose; Microcrystalline Cellulose

1. Introduction

In Egypt, agricultural wastes are known as one of the richest resources. In recent years, there has been a rise in studies exploring the potential of agricultural by-products as resources, which can be used to make various products at a low cost. This not only offers an affordable and sustainable option, but can also address the issue of waste disposal, which is a leading cause of environmental contamination. Cellulosic waste has become a particular focus of research in this area, and interest has grown significantly [1-4]. Many researchers have investigated ways to produce crystalline cellulose (MCC) from agricultural wastes such as rice straw, cotton stalk, and sugarcane bagasse. Microcrystalline cellulose (MCC) is a fine, white, odorless, crystalline powder of purified, partially depolymerized cellulose prepared by treating alpha-cellulose with mineral acids. MCC is insoluble in water, dilute acids, alkali, and most common organic solvents [5]. It has a smaller degree of polymerization and a higher specific surface area than other cellulose fibers [6]. Preparing MCC conventionally involves treating alpha cellulose with strong mineral acids, which breaks down the microfibrils that comprise it. The crystalline sections of these microfibrils are made up of microcrystals arranged in a rigid linear formation, while the paracrystalline regions are composed of

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disorganized cellulose chains. As a result of van der Waals interactions and hydrogen bonds, cellulose chains form crystalline regions called cellulose crystallites. The diameter of these crystallites was found to be the same as that of cellulose microfibrils. During acid hydrolysis, the amorphous phase easily hydrolyzes. Consequently, the cellulose chain fragments are smaller and more crystalline, known as MCC. This leads to a decrease in the degree of polymerization of the cellulose chain with minimal weight reduction. Microcrystalline cellulose biocomposites are emerging as an alternative to other polymeric composites. There is potential for eco-friendly bio-composites to be a new product of the 21st century and partially rectify several environmental problems. MCC has been widely used, especially in pharmaceuticals, cosmetics, food, and other industries [7,8]. Researchers used MCC as starting material for cellulose-reinforced nanocomposites [9]. It likewise reported that the type of cellulose, its origin, and the preparation method affect the overall properties of MCC [10]. MCC can be used as filler, which produces tablets of high mechanical and physical properties such as high rigidity, fast dissociation time, and a high rate of drug release. Furthermore, it is used in the production of direct-pressure tablets. MCC tablet production is growing due to many advantages; for example, uniform particle size does not require granulation, produces more stable tablets, and is profitable economically. Also, MCC can decrease sedimentation in suspension and dry syrup [11]. The use of this excipient in direct compression is widespread due to its impressive ability to bond and form tablets with excellent mechanical properties when dry. However, the flow properties are relatively weak due to the small size of the particles; it appears as low bulk density due to its molecular shape [12]. The production of MCC from agricultural waste presents an attractive opportunity for sustainable and cost-effective resource utilization. Agricultural residues such as sugarcane bagasse, wheat straw, corn cobs, and rice husks are potential sources of MCC [13]. The extraction process typically involves several steps, including pretreatment, hydrolysis, and purification. Various pretreatment methods, including physical, chemical, and biological approaches, can be used to improve the accessibility of cellulose and remove impurities [14]. Acid hydrolysis is the most common method used to extract MCC, but other methods, such as alkali hydrolysis and enzymatic hydrolysis, are also being explored. Purification of MCC is essential to remove impurities and unwanted by-products and can involve washing, filtration, or centrifugation [15,16]. Using agricultural waste as a source of MCC, we can reduce waste, lower costs, and create value-added products while improving the sustainability and efficiency of MCC production [17-19]. Further research is necessary to optimize extraction and develop high-quality MCC with consistent properties. This article aims to provide a detailed overview of the production process of MCC from sugarcane bagasse on a pilot scale. Also, it describes the different steps involved in the production process, including the preparation of raw materials, acid hydrolysis, washing, drying, and particle size reduction. The article also emphasizes the importance of carefully controlling process parameters to achieve the desired properties of the MCC. The MCC produced on a pilot scale has various applications in the pharmaceutical, food, and cosmetic industries, making it a useful excipient for drug formulation and other applications.

2. Materials and Experimental

2.1. Material

Sugarcane bagasse was collected from the local juice stores in Cairo, Egypt. Sodium hydroxide (NaOH), sodium hypochlorite (12%), and Sulfuric acid (H_2SO_4) was purchased from Elnacer Company, Egypt. The bagasse was washed in running water for half an hour, dried in the sun, cut, and ground using a mill we designed and manufactured.

2.2. Experimental

2.2.1. Compositional Analysis of Bagasse

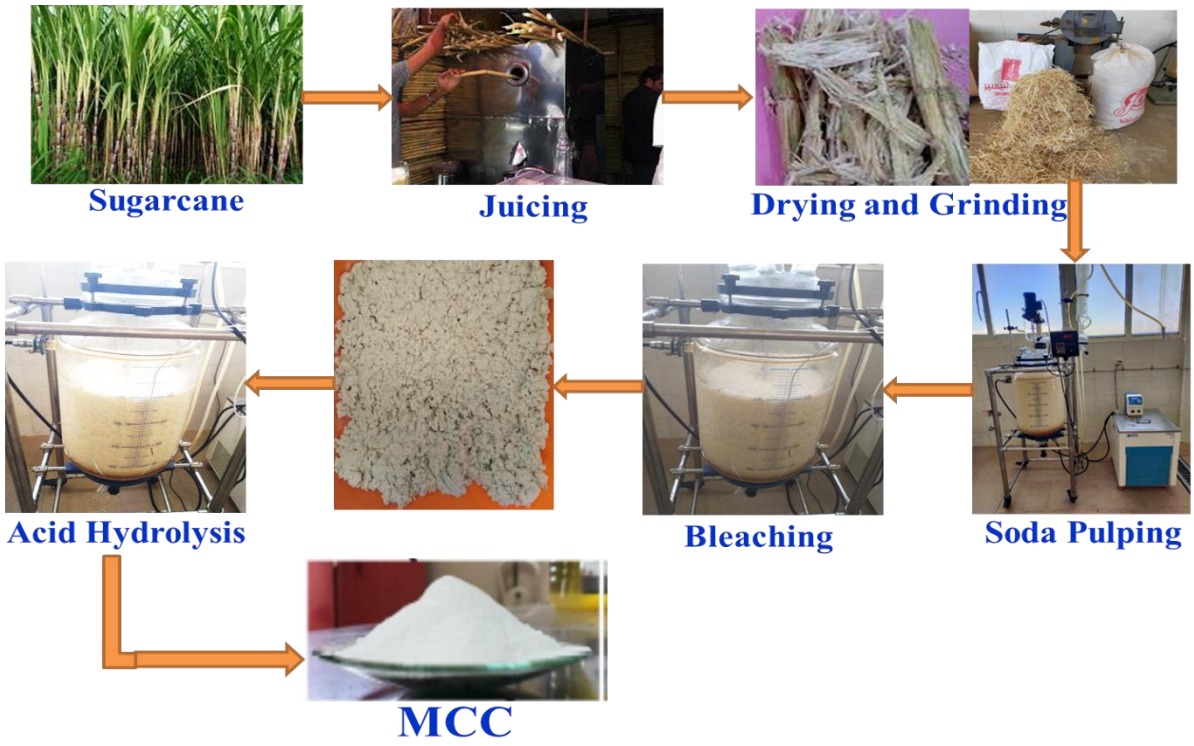
The compositional analysis of bagasse raw material was performed according to the known standard methods for determining chemical constituents, including lignin (T 222 om-15), hemicelluloses (T9 wd-75), holocellulose (ASTM D-1104 standard), α-cellulose (T 203 cm-09,), and ash content (T 244 cm-11), which was shown in **Table 1**. Three replicates were used for each sample.

Table 1: Chemical composition of SCB raw materials (% based on oven-dry weight)

Lignin (Klason lignin)	Hollocellulose	α-cellulose	Hemicellulose	Ash
21	78	60	18	1.30

2.2.2. Preparation of Microcrystalline Cellulose (MCC)

Microcrystalline cellulose was extracted from sugarcane bagasse through pulping followed by bleaching and then acid hydrolysis as follows (**Scheme 1**):



Scheme 1: Flow chart of MCC production.

2.2.2.1. Pulping Process

Sodium hydroxide is the primary chemical pulping agent used in soda pulping. The pulping procedure was as follows: 4 kg of oven-dried bagasse was placed in a 50-liter stainless steel reactor at 170 °C for 2 hours after reaching the temperature. NaOH was used in cooking at a concentration of 12% alkali, related to the oven-dry weight of bagasse. The bagasse-liquor ratio for the pulping process (Kg/L) was 1:6. Once the pulping process was completed, the pressure was relieved and the resulting pulp was disintegrated, rinsed with tap water until it was devoid of alkali and lastly, left to air dry. We calculated the yield of this pulping process at 49%.

2.2.2.2. Bleaching Process

Sodium hypochlorite was used to bleach the produced pulp. First, we treated 3 kg pulp with 2000 ml of sodium hypochlorite (12%) at 45 °C for 1 hour. Then, the pulp produced was filtered and washed with water. The calculated the yield of this bleaching process at 42%.

2.2.2.3. Acid Hydrolysis

The bleached pulp was hydrolyzed using sulfuric acid (2 M). The hydrolysis process was achieved by refluxing bleached pulp in 2 M sulfuric acid solution for 45 minutes in a liquid ratio of 1:10. The hydrolyzed pulps was then washed with distilled water and acetone, followed by drying in the air till constant weight. The yield was about 25% based on the bleached pulp weight.

2.3. Characterizations

2.3.1. Fourier transforms infrared spectroscopy

Fourier transform infrared spectrometer (JASCO, Japan) was used to measure the FTIR spectra of the samples, which were prepared by pressing a KBr disc containing 2 mg of sample and 98 mg of KBr. The measurements were taken in the range of 400–4000 cm-1.

2.3.2. X-Ray Diffraction

The XRD patterns were investigated on a Diano X-ray diffractometer using a CoKα radiation source energized at 45 kV and a Philips X-ray diffractometer (PW 1930 generator, PW 1820 goniometer) with CuK radiation source (λ=0.15418 nm), at a diffraction angle range of 2θ from 10 to 70° in reflection mode.

The crystallinity of MCC samples was determined by application of the Segal method as follows:

$$C_s = \frac{I_{200} - I_{Am}}{I_{200}} x100(\%)$$

Where C_s is the crystallinity (%), I_{200} is the reflection intensity of (200) plane diffraction and I_{Am} is the minimum intensity near 18.58 of 2θ angle.

2.3.3. Surface morphology

An environmental scanning electron microscope (FEI IN SPECTS Company, Philips, Holland) was used to analyze the surface morphology of MCC without coating. The images were obtained with an accelerating voltage of 10–15 kV.

2.3.4. Bulk, Tapped Density, and Flow Properties

Bulk and tapped densities were determined following established procedures outlined in prior research [8]. In summary, approximately 10 grams of the sample were introduced into a 50 ml graduated cylinder. Bulk density was computed as the ratio of mass to volume. To obtain the tapped density, mechanical tapping was applied until there was no further alteration in the volume of the sample. The resulting volume was recorded and used in subsequent calculations. Subsequently, the bulk and tapped densities were employed to assess the flow characteristics and compressibility of the MCC powder by determining the Hausner ratio and Carr's compressibility index.

$$\text{Hausner ratio} = \frac{\text{Tapped density}}{\text{Bulk density}}$$

$$\text{Carr index \%} = \frac{\text{Tapped density} - \text{Bulk density}}{\text{Tapped density}} X\ 100$$

Each sample was replicated five times.

2.3.5. Particle size distribution determination

Particle size distribution of MCC was measured using a laser-diffraction analyser (MicroTrac MT3000EX, Nikkiso, Japan). All MCC samples were sonicated in an ultrasonic processor for 180 s before measuring.

3. Results and discussion

3.1. FT-IR Analysis

Figure 1 displays the Fourier Transform Infrared (FTIR) spectrum of MCC. As can be seen, the spectrum contains the signature peaks of pure cellulose, such as the O-H stretching vibration at 3427 cm-1, the stretching vibration of CH and CH2 groups at 2921 cm-1, the adsorbed water at 1633 cm-1, the bending vibration of CH2 and OH groups at 1421, 1377, and 1311 cm-1, the stretching vibration of C-O for the glycosidic bonds and C-O of primary and secondary hydroxyl groups at 1166 and 1055 cm-1, as well as the out-of-plane deformational vibration of O-H groups at 866 cm-1.

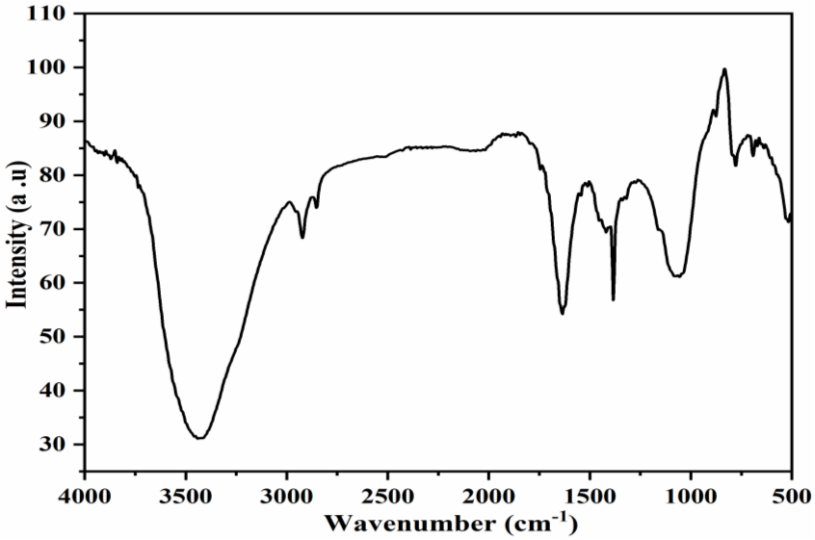


Figure 1: FTIR of MCC prepared from bagasse pulp.

3.2. X-ray diffraction of MCC

Figure 2 displays the X-ray diffraction patterns of the prepared MCC, which was used to determine its crystallinity. The obtained XRD pattern for MCC exhibited diffraction peaks at 2θ angles of 34.5, 22.5, and 15.8, which correspond to diffraction from (0 4 0), (0 0 2), and (1 ī 0) planes, respectively, in accordance with the known diffraction peaks of cellulose I. Based on these patterns, the calculated crystallinity of the MCC was found to be 83%.

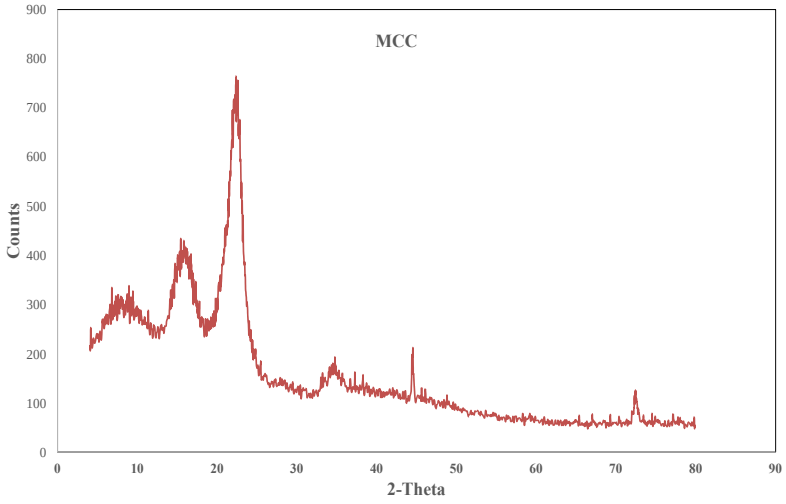


Figure 2: X-ray pattern of the prepared MCC.

3.3. Morphological characteristics of MCC

Figure 3 presents scanning electron microscopy (SEM) images of the MCC particles at different magnifications. The MCC was isolated from bagasse and dried; the SEM images revealed some interesting features of the MCC particles. At lower magnifications, the MCC particles appeared to be heavily aggregated, indicating the presence of strong inter-particle forces. This could be due to the hydrogen bonding between the cellulose microcrystals, which promotes aggregation. In addition, the MCC particles were long and rod-shaped, with a relatively smooth surface. The rod-like shape of the MCC particles is typical of cellulose microcrystals, which tend to form elongated shapes due to the anisotropic nature of cellulose. This morphology is important because it affects the behavior of MCC in different applications, such as reinforcement in composites or as a filler in coatings. The relatively smooth surface of the MCC particles

also suggests that the MCC was well-processed, with a low degree of impurities or irregularities.

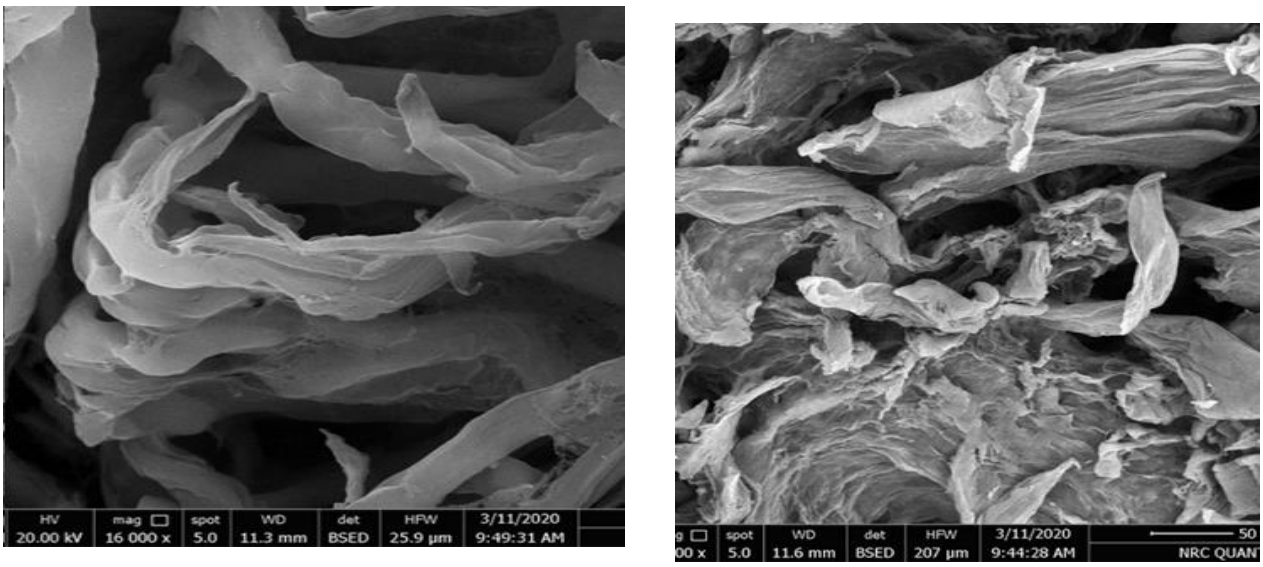


Figure 3: SEM images of the prepared MCC.

3.4. Particle size distribution and Surface area

The particle size distribution can affect the final product's physical and chemical properties, as shown in **Figure 4**. For example, smaller particle sizes can increase the surface area of the powder, which can affect its flowability and compressibility. Uniform particle size distribution can also ensure consistent mixing and blending of the powder with other ingredients. The average particle size of the prepared MCC from bagasse pulp, measured with light scattering, is 15-200 µm with an average of 49.95 µm. Avicel PH 101 has a particle size range of 20-200 µm, which means that its particle size distribution is closer to that of MCC prepared from bagasse pulp. The performance of MCC can be influenced by its surface area (**Figure 4**), which is an important property to consider for various applications. MCC derived from bagasse has a higher surface area of 1.2736 m^2/g compared to the average surface area of Avicel PH 101, which is 1.09 m^2/g.

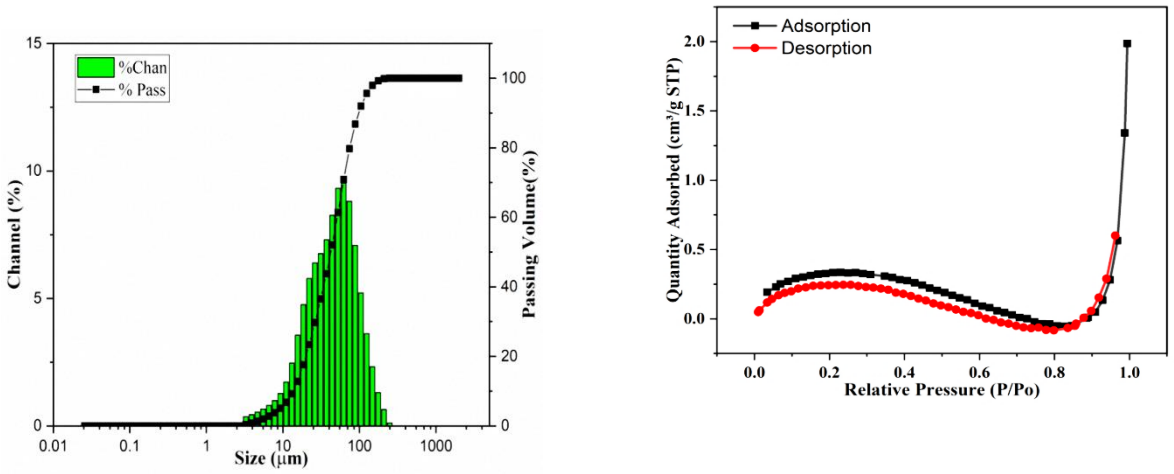


Figure 4. Particle size distribution (left)Adsorption/ desorption curve (*right*) of the prepared MCC.

3.5. Powder flow properties

Table 2 presents the bulk and tapped densities of the MCC, which are essential parameters to consider

for the material's handling and processing. Bulk density is the measure of the weight of a powder per unit volume, and it is usually determined by pouring the powder into a measuring cylinder without any compaction. Tapped density, on the other hand, is the measure of the weight of a powder per unit volume after tapping or vibration. Tapping the powder helps remove trapped air pockets and reduces the powder's volume. The Hausner ratio (HR) and Carr's compressibility index (CI, %) were calculated from the bulk density and tapped density values. The Hausner ratio measures inter-particulate friction or powder flowability. A higher Hausner ratio indicates a higher degree of inter-particulate friction, leading to poor powder flowability. On the other hand, the compressibility index (CI) measures the powder's ability to withstand compression and regain its original volume after the compression force is released. A higher CI value indicates poor flowability and powder bridge strength and stability. The flowing character of the MCC can be rated based on the CI and HR values. If the CI value is below 15%, the powder is considered free-flowing; if it is between 15 and 25%, it is moderately compressible. If the CI value is above 25%, the powder is considered cohesive; if it is above 35%, it is very cohesive. A high HR value also indicates poor flowability, which can lead to issues in processing, such as bridging or rat-holing. Therefore, the values of the Hausner ratio and Carr's compressibility index provide insights into the flow character of MCC, which is an important factor in the manufacturing and processing of pharmaceuticals and other industries that use powder formulations. Understanding these properties can help select the right type of MCC and optimize the manufacturing processes.

Table 2: Some physicochemical properties of the prepared MCC**.**

Crystallinity (%)	83
Bulk density (g/cm3)	0.27
Tapped density (g/cm3)	0.35
Carr index (%)	23.07
Hausner ratio	1.3

3.6. Characterization of MCC and comparison to the Commercial MCC

Characterization of MCC is an essential aspect of its utilization and applications. It involves determining MCC's physical and chemical properties, such as particle size distribution, crystallinity, degree of polymerization, and surface area. Commercial MCC is the most used type of MCC, and it is obtained from wood pulp. However, MCC can also be produced from agricultural wastes, such as rice straw, sugarcane bagasse, and cotton stalk. The characterization of MCC produced from these sources can be compared to commercial MCC to determine their quality and suitability for different applications (**Table 3**). One of the critical properties of MCC is particle size distribution, which affects its followability, compressibility, and mechanical strength. The particle size of commercial MCC typically ranges from 20 to 200 microns. MCC produced from bagasse varies from 15 to 200 microns. Crystallinity is another critical property of MCC, and it affects its water sorption, enzymatic degradation, and mechanical properties. Commercial MCC has a high degree of crystallinity, about 75%, while MCC produced from bagasse is almost the same (78%). The surface area of MCC is also essential property that affects its performance in different applications. MCC produced from bagasse may have a higher surface area than commercial MCC, which can influence its water sorption and binding properties.

Table 3. Comparing prepared MCC with Avicel PH 101

Properties	Avicel PH 101	Prepared MCC
Form	Solid, powder	Solid, powder
Appearance	White crystalline powder	White crystalline powder
Density (bulk)	0.37	0.27
Tapped density (g/cm3)	0.51	0.35
Crystallinity %	78.0	83
Particle size distribution	20—200 µm	15-200 µm
Specific surface area	0.78 – 1.4 m2/g	1.2736 m²/g
Degree of polymerization	317	226

3. Conclusion

Characterizing MCC produced from bagasse is essential in evaluating their quality and suitability for various applications. The characterization involves the analysis of their physical, chemical, and structural properties, including particle size, surface area, morphology, crystallinity, moisture content, and purity. By characterizing these properties, it becomes possible to understand how the MCC will perform in different applications and to compare their properties to those of commercial MCC. Comparing the properties of MCC produced from bagasse to those of commercial MCC can provide valuable insights into their performance and identify their advantages and limitations. For example, as we discussed earlier, bagasse-derived MCC has surface area higher than commercial MCC, which can be advantageous in specific applications. However, bagasse-derived MCC may also have different properties due to differences in the source of bagasse, processing conditions, and preparation methods. These variations may affect the performance of the MCC in specific applications and should be considered when selecting the appropriate type of MCC. Using MCC produced from bagasse as an alternative to commercial MCC is a promising approach that can provide a sustainable and cost-effective solution while reducing waste disposal issues. Bagasse is a renewable and abundant source of cellulose, and using it to produce MCC can reduce the environmental impact of waste disposal while providing a valuable product. Also, bagasse-derived MCC is often less expensive than commercial MCC, making it a more cost-effective option for some applications.

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5. Conflicts of Interest:

The authors declare no conflict of interest.

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