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Effect of Leaf Powdering Technique on the Characteristics of Date Palm-Derived Cellulose

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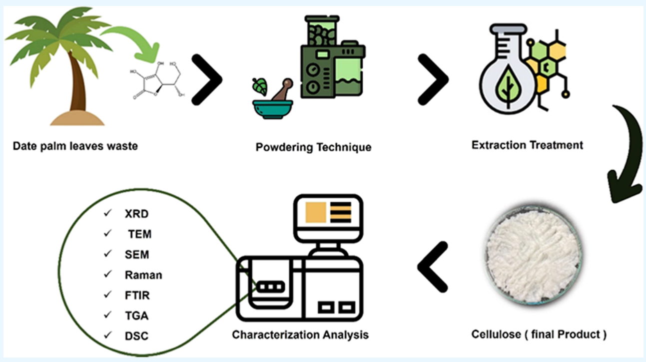

ABSTRACT: The date palm tree (*Phoenix dactylifera* L.) is the oldest cultivated tree and is very commonly seen in the Arab countries. In recent times, researchers are working on the conversion of the plant-based biowaste into value-added products. Cellulose is identified as one of the best options to be synthesized from plant-based materials due to its immense application possibilities. It is a natural hydrophilic polymer consisting of linear chains of 1,4-β-D-anhydroglucose units, and the most used method for cellulose extraction is acidic hydrolysis. However, in this study, a very sustainable, ecofriendly, and simple process of isolating cellulose from date palm leaves is discussed. In this study, the best mechanical approach (ball milling, grinding, or its combination) for changing the leaves into powder form, as well as the sustainable and simple chemical extraction of cellulose from those date palm leaves, is analyzed. SEM analyses confirmed that the mechanical treatment process affected the appearance of the cellulose formed. Raman spectrum confirmed the difference in stretching vibrations among the cellulose obtained. From the results obtained, it was noted that cellulose derived utilizing the grinding technique and subsequent chemical treatment was considered as the finest cellulose prepared with respect to its properties and structure, and the greatest yield obtained for Cellulose 2 was 42%. As a future scope, this cellulose developed can be used to produce advanced materials like nanocellulose.

1. INTRODUCTION

Date palm (*Phoenix dactylifera*) is the most commonly seen agricultural crop across many Gulf Cooperation Council (GCC) nations.[1,2] Given that it provides the inhabitants of the Arabian Peninsula with food, medicine, and even building materials, it is known as the "elixir of life".[3−5] For centuries, this tree has played a crucial role in the establishment and growth of civilizations in the arid regions of the Arab world.[6,7] In recent years, date palm-related knowledge, practices, and traditions have been recognized by United Nations Educational, Scientific, and Cultural Organization (UNESCO) as part of the world's intangible cultural heritage.[8] Additionally, many efforts are made, aiming to preserve the long-threatened culture as it serves as an important emblem of the Arab region's prosperity.[9] Due to their remarkable ability to endure extremely harsh conditions and requiring little maintenance, date palm trees are suitable crops for the Arab world.[10,11] According to Shahbandeh,[12] the date production has been increased from 7.1 to 9.45 million metric tons from years 2010 to 2020. There are over 62 million date trees in the Middle East and North Africa. The 16th largest date-producing nation in the world is Qatar,[13] and as of 2010, there were 581,336 date palm trees covering 2469 ha, producing 21,491 mt of dates. It is the main fruit tree produced in the nation, and date production accounts for 7.2% of all agricultural output.[14] It is estimated that 2 million tons of dry palm fronds is generated annually during the date harvest,[15−17] As a result, when the palms are pruned and refined seasonally, large amounts of waste biomass are produced. According to the estimates in Saudi Arabia, each palm tree produces between 20 and 35 kg of biomass waste each year, amounting to about 1 million metric tons (MMT) of biowaste in total.[18]

In general, a significant quantity of waste is produced by the agricultural sector in countries that adopt an agricultural-driven economy.[19] Given the severity of the global warming issue and the tremendous volume of waste produced, this would ultimately be harmful to both the economy and environment.

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The date palm plants produce massive amounts of trash, much of which is currently composted and burned, causing environmental degradation.[4] Effective waste management is still lacking in the palm industry, particularly the date palm and oil palm industries.[20] Recent studies on the uses and products of palm trash have been widely published, but proper industrial use of such applications remains in its early stages.[21,22] Hence, a proper and easy-to-handle method of waste extraction as well as beneficial product synthesis is necessary to execute a management plan of recycling such waste into useful products. This could be the future sustainable method that intends for the global reduction of ecological footprints as well as impacts. It must be noted that the date palm is abundant and rich in cellulosic fibers.[23] Using the cellulose-rich date palm biomass waste has the potential to support the long-term growth of biorefineries.[24] Hence, the diverse date palm waste components can be utilized to create new, sustainable bioproducts like cellulose.[25] It would additionally provide significant environmental and financial advantages, yet also result in the production of high-value biomaterials with superior chemical and physical characteristics. Table 1 presents some of the previous studies that utilized date palm cellulose into broad applications in different sectors.

Table 1. Investigation into Various Applications of Date Palm-Derived Cellulose

source	application	reference
palm fiber	reinforcement material	Braiek et al.[26]
date palm fruit stack	CNF membrane−−removal of *E. coli*	Hassan et al.[27]
palm RACHISES	hardboard production	El-Morsy et al.[28]
date pits and corn starch	food starch film	Alqahtani et al.[29]
date palm leaves	peroxidase extraction	Al-Senaidy and Ismael[30]
date palm fronds	oriented strand board production	Hegazy et al.[31]

Moreover, the attraction toward natural fibers has grown as a result of the increasing desire for more renewable and sustainable materials. Cellulose makes up the majority of the plant cell wall, which gives the structures strength and stiffness.[32] Plant-based cellulose fibers have several benefits over synthetic fibers, such as low cost, light weight, local availability, and exceptional mechanical qualities.[33] Date palm fiber and other natural fibers including jute, ramie, sisal, roselle, wood, cotton, coconut, and oil palm can all be used to extract cellulose.[34] A natural fiber consists essentially of rigid, crystalline cellulose microfibrils that are encased in an amorphous matrix made of lignin and hemicellulose.[35] Microfibrillar angle (MFA) refers to the angle that the cellulose microfibrils in the majority of plant fibers make with the cell axis.[36] MFA is considered to be an essential element in determining the mechanical characteristics of fibers, particularly their elasticity and rigidness.[37] Due to their lightweight, natural fibers have excellent specific characteristics in addition to being environmentally benign.[38] One of the sources of natural fibers is the date palm tree that could be a potential source of cellulose that can be extracted from palm leaves' waste.[39] Cellulose is composed of crystalline and amorphous regions, and the proportion of the two differs, depending on the cellulose source. The amorphous structure of cellulose is intrinsically less ordered relative to the crystalline structure, which helps interaction with a reactant.[40]

Date palm waste is generated from a variety of parts, which include date pits, leaves, fronds, and trunk. Due to its exceptional properties, researchers are currently interested in the fabrication of cellulose nanofibers and nanocrystals. Scientists have demonstrated that different biomasses have yielded cellulose nanofibers. Despite the great effort of extracting cellulose, many of them include complex processes and use a great number of chemicals. It is reported that the chemical and physical properties of cellulose prepared can vary significantly based on the preparation process. It has been noted that cellulose nanofibers (CNFs) derived from various date palm tree components exhibit variances in their structure, shape, level of polymerization, and surface qualities. Table 2 summarizes the average chemical weight percentage of different constituents present in various parts of a date palm tree.[41] Due to the weight percentage content and less ash content, in addition to higher extraction ability, the date palm tree leaf was selected to extract cellulose in this study.

Table 2. Different Constituents Present in Various Parts of Date Palm Tree[a]

part	cellulose %	hemicellulose %	lignin %	ash %	extractive %
leaf	57.75	20.00	15.30	1.75	8.20
leaflet	40.21	12.80	32.20	10.54	4.25
trunk	38.26	28.17	22.53	5.96	5.08

[a]Adopted from Mirmehdi et al.[41]

In spite of the fact that date palm waste can be considered as a source for the production of cellulose, due to numerous factors including the condition of the tree and its constituents, the properties of the cellulose obtained can vary significantly. Current research works have mainly focused on developing cellulose isolation techniques that are inexpensive, effective, and ecologically favorable. However, converting the current laboratory size technologies to pilot/industrial scale and being able to generate cellulose with consistent qualities remain the major challenges. Different studies have been carried out for isolating cellulose from date palm leaves, for instance, AlHamzani and Habib[42] obtained cellulose with a high degree of crystallinity and consistent appearance; however, extensive chemical treatment was performed. Elseify et al.[23] obtained a crystallinity degree of 76.01% of nanocellulose, but the only drawback is the significant weight mass loss due to high temperatures during the synthesis process and application of ball milling technique for leaf powdering. There were a few studies reported on the extraction of cellulose from date tree, especially from date palm tree spathe sheath and date palm seeds. There were also a few works reported on the preparation of cellulose from the leaflets of cellulose, however, with repeated chemical treatment.[43] Alhamza et al.[44] also reported on the preparation of cellulose nanofibers from date palm tree leaflets by the ball milling technique. Despite the fact that the ball milling technique is a cost-effective technology with vast potential and is applicable for preparing cellulose nanofibers or nanocrystals, it has got some drawbacks. The disadvantages of the ball milling technique include its high specific energy consumption (energy is mainly consumed on the wear of balls and wall armor, friction, heating of material, etc.), large weight, loud noise during working, contamination of the product (as a result of wear and tear which occur principally from the balls

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and partially from the casing), low working efficiency, and large-sized ball mill that weighs several hundreds of tons so that the one-time investment is high. Furthermore, hypochlorite is one of the common oxidizing agents that can be employed to treat cellulosic pomace in order to remove colored lignin and chromophoric compounds and produce high-quality cellulose. According to Dai et al.[45] bleaching process ensures the breakage of some water-insoluble bonds in lignin, resulting in the production of soluble compounds. For this reason, this study included necessary investigations to study the effect of increasing amount of oxidizing agent hypochlorite (NaClO) on the properties of the resultant cellulose. In our study, we investigated the effect of mechanical grinding pretreatment technique to isolate cellulose from date palm tree leaves. No studies have reported the synthesis of cellulose from date palm tree leaves by using the simplest grinding method as the leaf-powdering technique to the best of our knowledge. Comparatively, a very few studies have been conducted on the extraction of cellulose from date palm leaves. For that particular reason, we focused on using waste leaves to synthesize good-quality cellulose using the simplest powdering technique, i.e., grinding. In this study, natural cellulose was extracted from palm leaves under different mechanical treatments (ball milling, grinding, and both), followed by chemical treatment techniques to develop cellulose with a high yield, enhanced chemical composition, morphological stability, crystallinity index, and thermal stability (Figure 1). The characteristics were investigated through elemental analytical techniques such as SEM, Raman analysis, powder XRD, FTIR spectroscopy, DSC, and TGA.

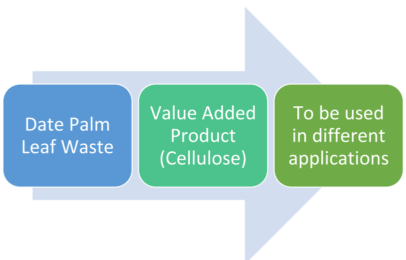

Figure 1. Schematic representation of the current study.

2. EXPERIMENTAL SECTION

2.1. Materials and Equipment. Sodium hydroxide (NaOH) extra-pure pellets and sodium hypochlorite (NaClO) were obtained from Sigma Aldrich. Extraction solutions were made using deionized water from Milli-Q system having 0.20 μm resistivity (Millipore, France). Date palm leaflet wastes were obtained from the campus of Qatar University. The ball milling machine used was a laboratory horizontal planetary ball mill, model no. WXQM-0.4 at 1000 rpm, and all the samples were ball-milled for almost 30 min time. The grinding machine used was a Spice Dry Grinding Machine SGM00, and the sample was ground for 30 s.

2.2. Extraction of Cellulose from Date Palm. For isolating cellulose, date palm tree leaves were initially cut into 5 cm sized parts and washed thoroughly with water for removing any particles of dust or dirt. The samples of leaves were subsequently dried in an oven for 5 h at a temperature of 70 °C till it is completely dried. Date palm leaf powder was obtained through three powdering mechanical techniques such as ball milling, grinding, and a combination of both grinding and ball milling consecutively. The preparation of cellulose using all three different types of mechanical techniques followed the same procedure and conditions. Consequently, 20 g of date palm powder was stirred with 15 g NaOH diluted in 300 mL of DI water on a hot plate stirrer at a fixed temperature of 70 °C for 4 h. The next step consisted of washing the solution several times with distilled water to remove NaOH from the slurry paste. The slurry paste obtained was then added to 60 mL of NaClO and 125 mL of distilled water and stirred for almost 4 h at a temperature of 70 °C. Similarly, the solution was washed several times to remove any residing chemicals. The resultant paste was positioned in an oven at 70 °C for 8 h till the specimens were completely dried. The cellulose obtained using the ball milling method was named as "Cellulose 1"; the cellulose obtained using the grinding method was named as "Cellulose 2"; and the cellulose obtained using the ball milling method and grinding together is known as "Cellulose 3" in this study. Furthermore, Cellulose 2 is the cellulose extracted from ground leaves treated with 48.0% NaClO, and Cellulose 4 is the cellulose extracted from ground leaves treated with 64.0% NaClO.

2.3. Calculation of the Content of α-Cellulose. In order to prepare the sample, 20 mL of 17.5% sodium hydroxide solution is macerated with 0.5 g of the fibrous material in a 100 mL beaker. After letting the mixture sit for 10 min, 30.0 mL of water is added and thoroughly blended. 1 h is given for the mixture to stand, with periodic stirring. 3 mL of the liquid is transferred to a Gooch crucible for suction-free draining after being stirred once more. In a 100 mL volumetric flask, the filtrate and wash water are collected. The cellulose is moved to a crucible, put in a 400 mL beaker, and then 25 mL of 12 M sulfuric acid is added to the beaker at room temperature. The mixture is washed with an additional 50 mL of acid after a few minutes. Alpha-cellulose is dissolved, 25.00 mL of potassium dichromate solution is added, and the mixture is heated to 140−150 °C before being maintained at that temperature for roughly 10 min while being bubbled with a fine stream of air to prevent bumping. 50 mL of water is added when the solution has cooled to 130−140 °C, and the mixture is then cooled to 60 ° C or below. After that, 0.5 M ferrous ammonium sulfate solution is used to electrometrically titrate the residual dichromate. The second step involves adding 50 mL of strong sulfuric acid gently down the side of a 400 mL beaker, while continuously swirling a mixture of 50.00 mL of the filtrate and 5.00 mL of potassium dichromate solution. After that, the resultant solution is heated and titrated.

The percentage of alpha-cellulose is calculated by dividing the volume of dichromate corresponding to it by the total volume of dichromate for all alpha-, beta-, and gamma-fractions.

$$\begin{aligned}\text{Alpha} - \text{cellulose (\%)} \\ = V\ (\text{mL})\ \text{dichromate used for alpha cellulose}/V\ (\text{mL}) \\ \text{dichromate used for gamma cellulose} + V\ (\text{mL}) \\ \text{dichromate used for gamma cellulose}\end{aligned}$$

2.4. Characterization of Extracted Cellulose. The extracted cellulose was characterized for its properties using several analytical techniques. The FTIR (760 Nicolet) technique was employed to confirm the main functional groups of the isolated cellulose. Approximately 5−10 mg of

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finely ground or ball-milled cellulose samples was placed onto the face of a KBr plate, and scanning was done within the range of 400−4000 cm^{-1}. Approximately 5−10 mg of finely ground or ball-milled cellulose samples was placed onto the face of a KBr plate, and the scanning was within the range of 400−4000 cm^{-1}. XRD analysis was carried out using PANalytical, Empyrean, the Netherlands. To characterize the prepared cellulose crystalline phase pattern, the finely ground or ball-milled cellulose powder was poured onto a holder, and the excess was wiped off. The XRD analysis was performed with CuK$\dot{\alpha}$ radiation at a wavelength of λ = 1.540 Å for determining the crystalline phase of the synthesized samples. SEM/EDX (Nova Nano SEM 50 Series, FEI, USA) was performed to analyze the cellulose sample microstructure and morphology. In the current work, the SEM instrument employed was NovaNano SEM 450, and its voltage capacity is in the range of 200 V−30 kV. The chemical structure of the isolated cellulose was obtained using the Raman analysis. The instrument used for Raman spectroscopic analysis was a Thermo Fisher Scientific DXR Raman microscope. TGA determined the sample's thermal stability and the fraction of the volatile components. DSC analysis (NETXSCH DSC 214 Polyma) provided information about the sample's thermal transitions and physical properties.

3. RESULTS AND DISCUSSION

3.1. XRD Characterization. XRD analysis is a key parameter in studying the effect of different mechanical techniques on the characteristics of cellulose yielded. Figure 2 presents the XRD analysis patterns of microfibrous cellulose extracted from date palm leaves using three different powdering techniques. It can be noted that approximately all samples exhibited a similar pattern of peaks, with varied intensities of the peaks. The diffraction patterns for the cellulose showed intense peaks that clearly coincide with the ones reported by previous literatures by Elseify et al.[23] For example, the presence of three distinct angles in the cellulose, 15.6°, 22.6°, and 34.2°, is confirmed through the intensity of the diffraction peaks obtained by Pattnaik et al.[46] Non-cross-linked composites containing nanofiber cellulose exhibit a clear peak at 22°. Additionally, a study[47] pointed out that the peaks of the composites corresponding to this A-type amylose allomorph become stronger as the percentage of the filler increased.[47] In this case, cellulose obtained via the different mechanical techniques exhibited clear variation in the intensity peaks. The isolated cellulose from ground leaves depicted diffraction peaks at 22° with a higher intensity compared to other samples, which corresponds to the 002 reflections. Nagalakshmaiah et al.[49] confirmed that such behavior in the XRD patterns occurs only for highly purified cellulose.

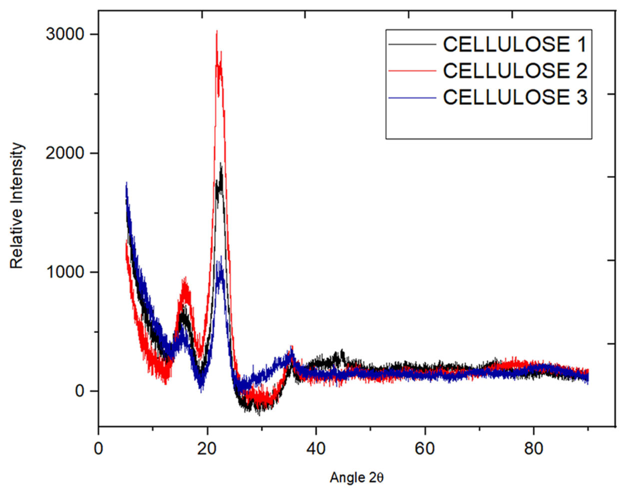

Figure 2. XRD pattern of pure cellulose extracted from date palm leaves. Cellulose 1: cellulose extracted from ball-milled leaves; Cellulose 2: cellulose extracted from ground leaves; Cellulose 3: cellulose extracted from ground and ball-milled leaves together.

The XRD pattern displayed in Figure 3 corresponds to cellulose isolated with a similar mechanical technique, i.e., grinding, however, with a changed amount of oxidizing agent during the cellulose synthesis. Cellulose 2 is the cellulose extracted from ground leaves treated with 48% NaClO, and Cellulose 4 is the cellulose extracted from ground leaves treated with 64.0% NaClO. Findings obtained clearly show that cellulose treated with 64.0% NaClO exhibits a higher intensity diffraction peak located at 22° compared to the cellulose treated with 48% NaClO. It can be noted that increasing the amount of hypochlorite resulted in even more pure cellulose. Waste material from date palm contained structural lignin and hemicellulose. In the bleaching process, raw samples were deprived of hemicellulose and some lignin, resulting in a higher crystallinity index. Following further lignification, the crystallinity index of the bleached sample increased, suggesting that lignin has been removed.[48] According to Shi and Liu,[50] the primary peak, designated as the diffraction intensity of crystalline zones, is represented by the I_{002} peak intensity (the greatest intensity of the I_{002} lattice diffraction), whereas the secondary peak denotes the diffraction intensity of the amorphous zone. The principal peaks of Cellulose 2 and Cellulose 4 samples were somewhat separated from one another, which suggested that the structure's crystallinity had increased.[32] The variation in the width of the crystallization peak reflects the intensity changes in the hydrogen bonding between the molecules of cellulose.[47] Date palm fibers have a crystallinity index within the upper range of values.[49] Isolated cellulose from ground leaves (Cellulose 2) seems to be the most similar to the ones in

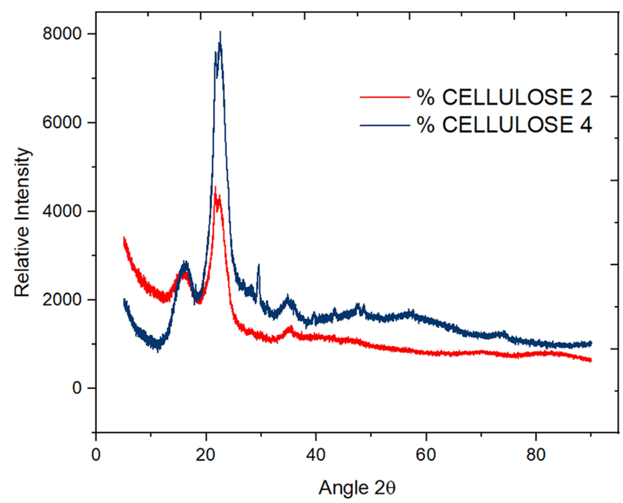

Figure 3. XRD pattern of pure cellulose extracted from date palm leaves. Cellulose 2: cellulose extracted from ground leaves treated with 48% NaClO; Cellulose 4: cellulose extracted from ground leaves treated with 64.0% NaClO.

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the literature of Nagalakshmaiah et al.[49] The crystallinity of Cellulose 2 was noted to be 60.2% Also, it is confirmed that increasing the concentration of hypochlorite resulted in a higher quality cellulose.

3.2. FTIR Spectroscopy. Figure 4 displays the FTIR spectra of extracted cellulose samples, including Cellulose 1, Cellulose 2, and Cellulose 3. It can be observed from Figure 4 that the spectra of all samples exhibit a similar trend of absorption peaks. With reference to the functional groups of cellulose, as noted from this figure, it can be noted that lignin and hemicellulose were successfully eliminated during the cellulose isolation process. Accordingly, the resulting spectra obtained exclude the absorption peaks at 1738 cm^{-1}, attributable to the C═O stretching vibration of carboxylic groups found in hemicellulose and lignin. The peaks noted at 2899 and 3451 cm^{-1} are attributable to the stretching vibrations of C−H and O−H groups, respectively. Wulandari et al.[51] also reported comparable stretching vibrations of O−H within the same range. Moreover, the peaks at 1382 cm^{-1} are caused by the vibrations of C−H and C−O groups in polysaccharide rings. Additionally, the absorption peak at 1060 cm^{-1} indicates C−O−C in the pyranose ring. Highlighting the distinctive features of the resulting three spectra, it can be observed that Cellulose 2 exhibits a higher intensity vibrational peak at almost 1000 cm^{-1} compared to Cellulose 1 because of crystallinity. This finding supported the lower crystallinity confirmed by the XRD analysis of Nagalakshmaiah.[49] The isolated cellulose from ground palm leaf powder shows the most similar FTIR pattern with the referenced literature reported by Fethiza Tedjani et al.[52] Peaks showed relative amounts of the I crystal form, as represented by the band at 708 cm^{-1}. The O−H and C−H stretching vibrations are noted at 3329.98 and 2890.29 cm^{-1}, respectively. The broad band at 1644 cm^{-1} is due to the OH bending mode of water. The coupling modes of C−O and C−C vibrations are assigned to the band at 1025.94 cm^{-1}.[52]

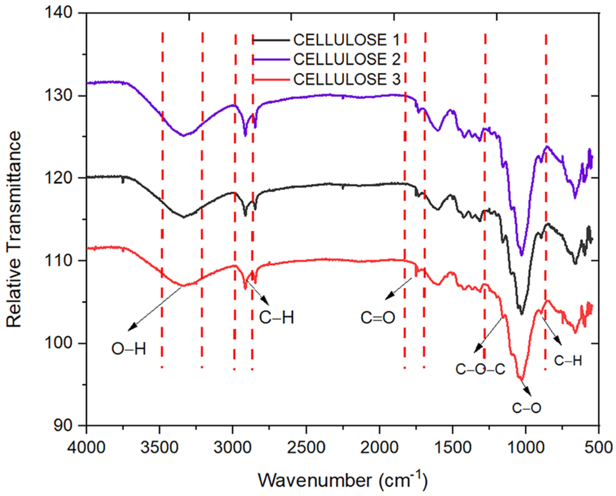

Figure 4. FTIR spectra of pure cellulose extracted from date palm leaves. Cellulose 1: cellulose extracted from ball-milled leaves; Cellulose 2: cellulose extracted from ground leaves; Cellulose 3: cellulose extracted from ground and ball-milled leaves together.

The FTIR spectra presented in Figure 5 show cellulose extracted from palm leaves powdered with a similar mechanical technique while varying the bleaching amount with NaClO. Cellulose 2 is the cellulose extracted from ground leaves treated with 48% NaClO, and Cellulose 4 is the cellulose extracted from ground leaves treated with 64.0% NaClO. As per the study carried out by Dai,[45] the bleaching process ensures the breakage of some water-insoluble bonds in lignin, resulting in the production of soluble compounds. From Figure 5, it can be noted that the spectra of cellulose treated with 64% NaClO exhibit no absorption band at 1725 cm^{-1} with respect to the C═O group vibration, which implies the presence of hemicellulose. The results obtained are in agreement with the results presented by Luo and Wang[53] Luo and Wang[53] claimed that increasing the amount of hypochlorite from 48 to 64.0% secured the production of higher quality cellulose.

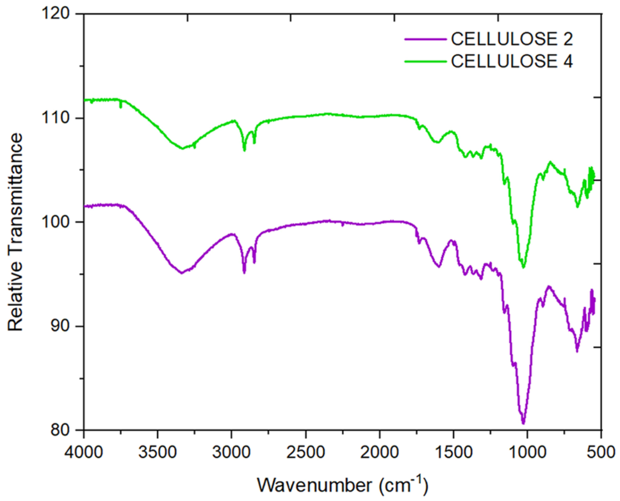

Figure 5. FTIR spectra of pure cellulose extracted from date palm leaves. Cellulose 2 represents cellulose extracted from ground leaves treated with 48% NaClO, and Cellulose 4 represents cellulose extracted from ground leaves treated with 64.0% NaClO.

3.3. SEM Analysis. In this study, the surface morphologies of different cellulose specimens extracted from the leaves of date palm biomass were examined under a scanning electron microscope. The specimens were coated with Au/C employing a vacuum sputter coater before SEM analysis to increase their conductivity as well as the quality of the pictures obtained. It was noticed clearly that the mechanical treatment process affected the cellulose appearance. From Figure 6a,c, it is noted that some nodular agglomerates were present on the surface of the cellulose separated from the date palm waste. The surface was dominated with the components of lignin and hemicellulose that are cemented together to form bundles of continuous individual cells. Similar findings were reported by Alhamzani and Habib[42] for raw and untreated cellulose fibers. From Figure 6a,c, it can be confirmed that the extraction method was not efficient to remove the extractives and other impurities, unlike Figure 6b,d that used the grinding method. Figure 6b,d clearly confirm that the structure is smooth rod, and this could be due to the breaking of the links between hemicellulose and lignin. The elimination of the binding component also caused the fiber bundles to disperse into individual fibers, resulting in fibers with reduced diameters. All of the treatments (alkali and bleaching treatments) that cleaned the surface of the fibers produced a smoother surface of the extracted cellulose. Moreover, it is possible to describe the clearly distinct cylindrical rodlike frameworks seen in the cellulose fiber as being highly porous and having an appreciable diameter, which can offer a substantial surface area for physical

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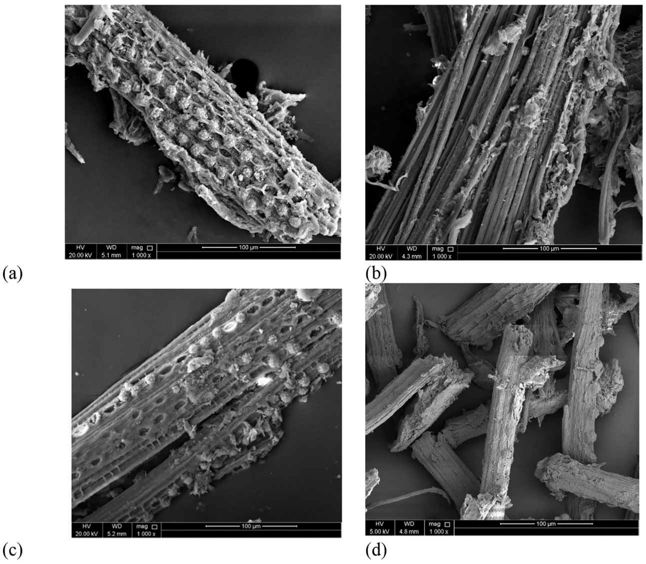

Figure 6. SEM images of cellulose isolated from date palm leaves (a–d) from different methods. (a) Cellulose 1––ball milling, (b) Cellulose 2––grinding, (c) Cellulose 3––both ball milling and grinding, and (d) Cellulose 4––increased bleaching treatment.

and chemical interactions as well as electrical conduction as a morphological benefit.

3.4. DSC Analysis. DSC analysis was performed to determine the stability, compatibility, and phase transitions of excipients. In DSC analysis (Figure 7), two types of samples

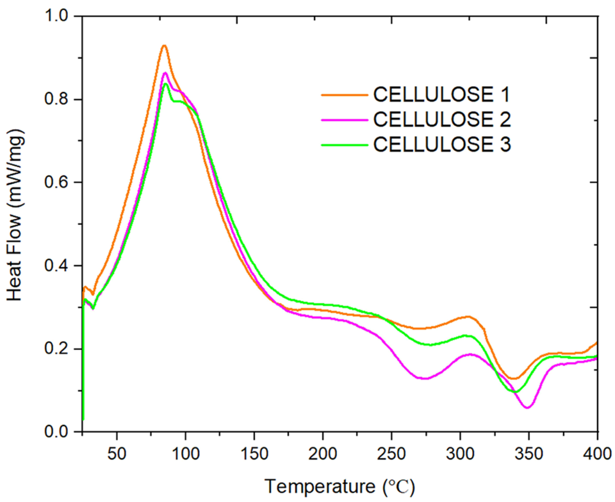

Figure 7. DSC analysis of the three types of cellulose isolated from date palm leaves.

were found to exhibit an early endothermic band in the range of 60–100 °C. As mentioned by Owi et al.[54] these peaks were due to the heat energy absorbed and water molecules volatilized. There was an additional small peak at 150 and 185 °C for each type of samples at different variations in the 160.5–170.5 °C regions. The same results were obtained in a research work performed by Chan and others.[55] Meanwhile, a larger peak was evident at 240–287 °C. As an illustration, the endothermic peak developed by melting is observed at around 250 °C. As a result of quenching after heating, there was little crystallization after glass transition and subsequent melting, indicating that the sample was in an amorphous state.[56] Furthermore, Cellulose 2 showed an exothermic peak at almost 200.3 °C, but Cellulose 1 and Cellulose 3 (195.9 °C) are more expected to show the exotherms as shoulder bands instead of sharp peaks. It is possible that the degradation of cellulose occurs more uniformly in Cellulose 2 than in other powdering methods.[66] Moreover, all types of samples show an exothermic peak between 290 and 325 °C, which was associated with decarboxylation and depolymerization of cellulose.[57] Thus, through the DSC analysis, it was confirmed that Cellulose 2 exhibited enhanced characteristics than Cellulose 1 and Cellulose 3.

3.5. TG Analysis. TGA was carried out to investigate the thermal stability of the obtained cellulose samples. TGA measurements of the thermal degradation of samples from 30 to 600 °C at 10 °C/min were carried out. Figure 8 shows the

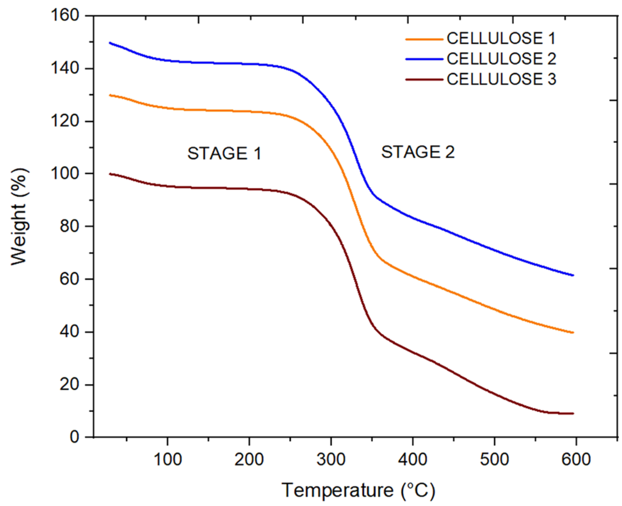

Figure 8. TGA of three types of cellulose isolated from date palm leaves.

three similar degradation ranges that took place in the three samples Cellulose 1, Cellulose 2, and Cellulose 3, with moisture and volatile chemicals being eliminated at temperatures 329.31, 328.45, and 329.78 °C, respectively. The degree of crystallinity, type, and/or source of cellulose impact the thermal stability of the cellulose formed.[58] The three different cellulose samples, developed using three different mechanical techniques, were thermally decomposed into two weight loss stages, which corresponded to the slow pyrolysis and quick pyrolysis stages. Water volatilization and evaporation were linked to weight loss at the early and slow pyrolysis stages of the curve. In the meantime, rapid pyrolysis took place between 250 and 350 °C owing to the cellulose decomposition process. Dehydration and cellulose degradation caused further weight loss to occur rather quickly.[59] Alhamzani and Habib[42] found TGA results of cellulose similar to this study. In the context of the current investigation, it can be said that TGA shows the effectiveness of isolated Cellulose 2 compared to both Cellulose 1 and Cellulose 3.

3.6. Raman Analysis. A significant focus during the early stages of Raman spectroscopy was on identifying bands with the characteristics of various cellulose polymorphs, studying the orientation/organization of cellulose in materials, and carrying out the band assignment work.[60] Several studies have been conducted using flexible substrates such as cellulose along with portable Raman spectroscopy, such as Agarwal[61] and

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others' studies, providing the detailed insights on molecular dynamics, crystallinity, polymorphism, and chemical structure information. As shown in Figure 9, there is a very high degree of similarity between the vibrational modes of all three types of cellulose, for mechanical powdering below 1000 cm^{-1}, which indicates that each method is characterized by the same vibration. A similar Raman spectroscopic result was reported in a study carried out by Wiley and Atalla.[62] A significant difference in frequency is observed in the range of 1000 cm-r and above.[62] The band noted at 2330 cm^{-1} in the cellulose Raman spectrum can be attributed to one or more of the following processes: first, certain oxidative mechanism occurs at the time of chemical modification conditions, or, next could be the high tendency for the powdering technique to show the effect on the surface area and small dimension of the fibers. This band is assigned to the C−C triple bond stretching mode. A region between 2000 and 2500 cnrl contains several tightly spaced, medium-intensity bands on the Raman spectra of Cellulose 1 and Cellulose 2. As explained in this article, cellulose exhibits the same frequency bands in its infrared spectrum as its Raman spectrum, but with a much higher intensity. Furthermore, the cellulose obtained matches the result mentioned in the study by Alves.[63] There is a variation in intensity heights of the peaks in the region of 3000 cm^{-1}, which indicates a difference in stretching vibrations among the three types. For more explanation, the OH stretching vibration occurs in this region, which reflects the main change occurring in the intensity of the results obtained. The O−H movements are separated from the other internal vibrations of the cellulose molecule, as noted in a study.[63] The hydroxyl stretching movements are involved in intermolecular hydrogen-bond

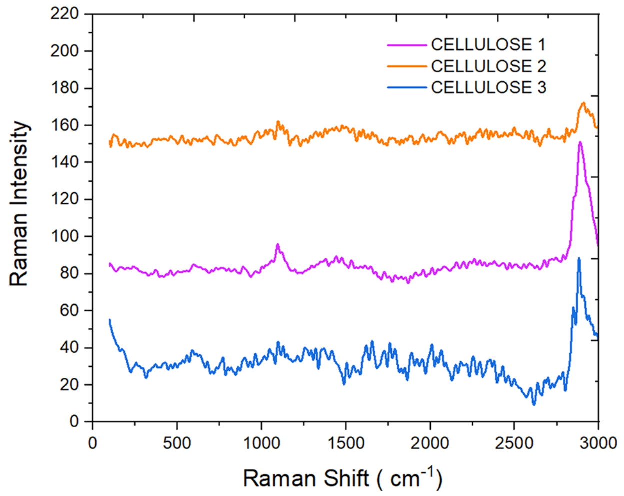

Figure 9. Raman analysis of cellulose isolated from date palm leaves.

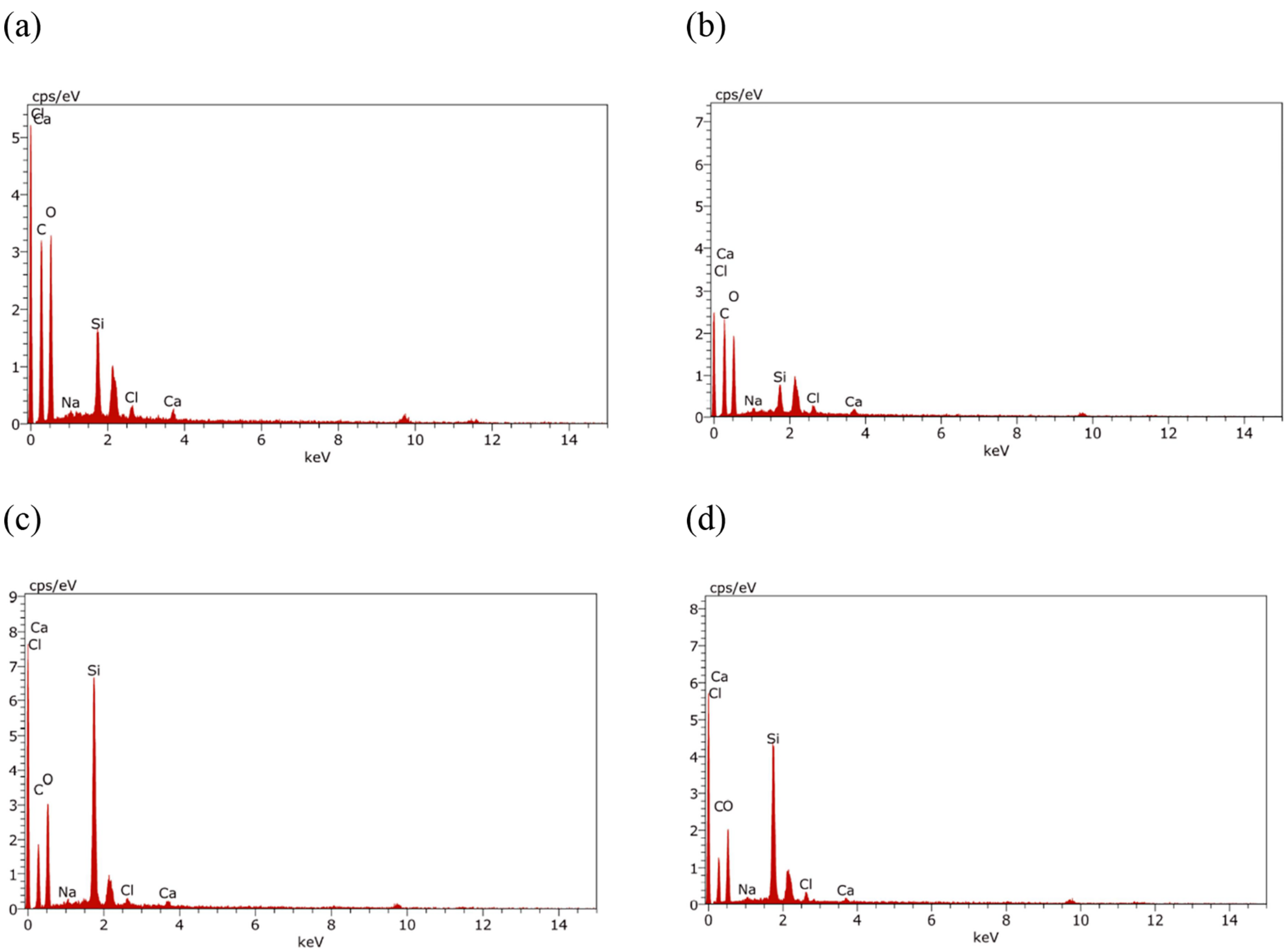

Figure 10. EDX analysis of cellulose isolated from date palm leaves: (a) Cellulose 1, (b) Cellulose 2, (c) Cellulose 3, and (d) Cellulose 4.

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interactions, which help to link with the lattice modes.[63] It has been shown by Raman analysis that isolated Cellulose 2 is more efficient than isolated Cellulose 1 and Cellulose 3.

3.7. EDX Analysis. The purity of the different types of cellulose can be confirmed using the EDX analysis. Figure 10a−d shows the EDX results of Cellulose 1, Cellulose 2, Cellulose 3, and Cellulose 4. It can be noted that although NaClO is used for the preparation, the amount of Cl present in the cellulose samples obtained is very less. Cellulose 1, Cellulose 2, Cellulose 3, and Cellulose 4 demonstrated 0.9, 0.8, 0.32, and 0.45% of Cl, respectively. The cellulose extraction process involves multiple steps, including delignification, alkaline treatment, and bleaching. NaClO is employed in the bleaching step, which primarily targets residual lignin and other impurities to enhance cellulose purity.[64] Maintaining optimal conditions including temperature, pH, and concentration of NaClO during the NaClO bleaching process is essential for minimizing the generation of unwanted byproducts such as dioxins. By carefully controlling these parameters, the bleaching reaction can be carried out efficiently, ensuring high-quality cellulose extraction, while reducing the environmental impact associated with the process. Hence, considering the significantly lesser amount of chlorine in the final cellulose sample, we can consider this as a sustainable process.

3.8. Plant Extract Analysis. FTIR analysis was performed for determining the leaf extract composition, and the results are presented in Figure 11. The FTIR results of the date palm

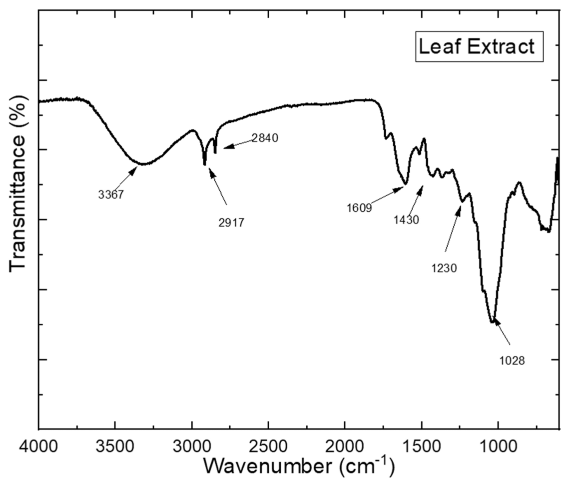

Figure 11. FTIR analysis of the date palm leaf extract.

leaf extract confirmed a broad peak at almost 3367 cm^{-1} that is related to the stretching vibration of −OH groups of phenolic as well as polyphenolic components present in the extract. Also, peaks were observed in the region between 2800 and 3000 cm^{-1}, and these are due to the stretching vibrations of C−H of aromatic skeletons like aromatic acids as well as flavonoids. A unique C═C stretching was observed at 1609 cm^{-1}, which is because of the aromatics together with the carbonyl C═O stretching of flavonoids and polyphenols. Several distinctive peaks related to the stretching of C−C and bending vibration of C−H in aromatic rings at approximately 1430 cm^{-1}, along with the C−O group of polyols, such as hydroxy-flavonoids, have been noticed at the region approximately 1230 cm^{-1}.[65] A comparable FTIR analysis was performed on peaks in a study carried out by Khalil et al.[66] The above-presented FTIR results demonstrated the effective characterization of the date palm leaf extract.

3.9. Calculation of the α-Cellulose Content. The obtained alpha-cellulose values for Cellulose 1, Cellulose 2, Cellulose 3, and Cellulose 4 of 0.03 g samples were 71.0, 78.6, 75.6, and 77.0%, respectively. According to some studies, the alpha-cellulose content in the cellulose prepared from date palm tree leaves can range from 39.9 to 87.9%, depending on the method of extraction and purification.[24] In a study by Galiwango et al.,[24] cellulose was extracted from date palm tree, and the resulting cellulose had an alpha-cellulose content of 78.63, 75.64, and 70.40%, obtained from rachis, leaflet, and fiber, respectively. It is worth noting that different methods of extraction and purification can lead to different alpha-cellulose values; so, the specific method used in a given study can influence the results. Additionally, the alpha-cellulose content can vary depending on factors such as the age and condition of the date palm leaves, as well as the season in which they were harvested.

4. CONCLUSIONS

In this study, a sustainable, ecofriendly, and simple process of isolating cellulose from date palm leaves was examined. Here, the best mechanical approach (ball milling, grinding, or its combination) for changing the leaves into powder form, as well as sustainable and simple chemical extraction of cellulose from those date palm leaves, is analyzed. According to the results obtained from the XRD, FTIR, SEM, DSC, TGA, EDX, and Raman analyses, it can be concluded that cellulose obtained from method 2, using the grinding technique, has the most potential in terms of cellulose structural formation, as well as the fact that the obtained yield of Cellulose 2 was 42%. From a future perspective, cellulose from the grinding technique could be used to produce more advanced and specialized forms of products, such as nanocellulose and cellulose triacetate (CTA), which have distinct and promising properties and could be generated from conventional cellulose in the future.

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Notes

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