SEMARAK ILMU PUBLISHING

Journal of Advanced Research in Micro and Nano Engineering

Journal homepage:
https://semarakilmu.com.my/journals/index.php/micro_nano_engineering/index
ISSN: 2756-8210

Isolation and Characterization of Cellulose Nanocrystals from Solid Seaweed Wastes

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ARTICLE INFO

Article history:
Received 7 September 2024
Received in revised form 11 October 2024
Accepted 19 November 2024
Available online 31 December 2024

Keywords:
Nanocellulose; lignocellulosic biomass; acid hydrolysis; carrageenan extraction; reinforcing filler

ABSTRACT

Cellulose nanocrystals (CNCs) are an intriguing class of bio-based nanoscale materials, desirable for a wide range of applications due to their remarkable physicochemical properties which include biocompatibility, biodegradability, renewability, low density, tuneable surface chemistry, optical transparency and improved mechanical properties. In this study, the solid seaweed wastes (SSW) obtained from carrageenan production were utilized to isolate CNCs using a modified one-pot oxidative-hydrolysis treatment. The physicochemical properties of the CNCs were further characterized using various methods including Fourier Transform Infrared Spectroscopy (FTIR), Energy Dispersive X-ray (EDX) analysis, X-ray Diffraction (XRD), Scanning Electron Microscopy (SEM), Transmission Electron Microscope (TEM) and Atomic Force Microscopy (AFM). The spectra analysis from FTIR and EDX reveals the removal of non-cellulosic components from the SSW after hydrolysis, along with high percentages of carbon and oxygen, implying the cellulose-rich content of the CNCs. The SEM, TEM and AFM analysis depicts the needle-like morphologies of CNCs with sizes ranging from 6 to 35 nm. The XRD analysis also reveals the high crystallinity of CNCs, which benefits its mechanical properties. This study demonstrates the potential for exploiting waste products from carrageenan extraction of red seaweed to obtain valuable CNCs which hold great potential for applications in areas such as flexible electronics and food packaging.

1. Introduction

Seaweed has emerged as one of the most promising resources due to its remarkable adaptability, rapid growth and resource sustainability. From 2000 to 2019 [1], global seaweed production increased almost three times, from 118,000 tonnes to 358,200 tonnes. By 2030, the market is expected to generate $1,512 million, growing at a Compound Annual Growth Rate (CAGR) of 11.6%. Seaweed is primarily exploited for its hydrocolloids, such as agar, alginate and carrageenan, which

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https://doi.org/10.37934/armne.28.1.4759

are used as thickening, stabilizing and gelling agents in the food industry as well as bioactive materials in the medical and pharmaceutical fields [2].

Carrageenan is a sulphated linear polysaccharide extracted from red seaweed that is made up of d-galactose residues linked by (1→3)-linked β-d-galactopyranose and (1→4)-linked α-d-galactopyranose [3]. A massive number of solid residues or also known as seaweed solid waste (SSW) are produced in the carrageenan processing industry, resulting in the loss of valuable substances, environmental pollution, economic burden and health risks [4,5]. It is estimated that at least 72,000 dry tons of the remaining seaweed wastes might be recovered from seaweed hydrocolloid producers, given that the extraction yield of hydrocolloid varies from 17% to 52% [6]. The fact that one-third of SSW is cellulose with a trace of lignin and hemicellulose makes it a promising feedstock for nanocellulose production. An average cellulose content of 6.31% was found in various brown and red seaweed species, the main contributor to hydrocolloid production [7].

Nanocellulose currently has enormous potential in a wide range of applications, from plastic packaging to therapeutic excipients [8]. This is due to its intrinsic properties, such as abundant in nature, biodegradable and renewable. The nano-dimensional features including high specific surface area with abundant surface hydroxyl groups and high aspect ratio endows them with superior mechanical and thermal properties [9]. Nanocellulose can be isolated from various lignocellulosic biomass of terrestrial origin including wood, cotton and agricultural residues such as sugarcane bagasse, rice husk and fruit peels [10-16]. Compared to terrestrial lignocellulosic sources, seaweed offers several advantages for nanocellulose production including rapid growth, ease of cultivation and the possibility of a mild extraction method due to a low content of recalcitrant non-cellulosic components [7].

Several studies have been conducted on the utilization of seaweed from various species such as *Ulva fasciata* [17], *Gelidiella aceroso* [18], *Caulerpa corynophera* and *Sargassum siliquosum* [19] for nanocellulose production. However, research on exploiting SSW as a source of nanocellulose is still in its infancy. Therefore, the current study aims to explore the potential of SSW obtained from carrageenan extraction of *Kappaphycus alvarezii* seaweed as feedstock for nanocellulose production through a mild one-pot oxidative hydrolysis method. Physicochemical properties of the SSW nanocellulose were investigated using FTIR, EDX, XRD, SEM, TEM and AFM. Findings from this study may stimulate the development of sustainable nanotechnology practices for nanocellulose production, which can be utilized in various sectors, such as in healthcare products, enhancing packaging materials, fortifying composite materials used in construction, automotive and electronics, as well as improving the quality of paper and textiles and even refining food products.

2. Methodology

2.1 Materials

Kappaphycus alvarezii (*K. alvarezii*) seaweed was purchased from a local supplier in Semporna, Sabah, Malaysia. Hydrogen peroxide (H_2O_2, 30%) and sulfuric acid (H_2SO_4, 98%) were a product of Merck, Germany. A cellulose standard, Avicel® PH-101 (microcrystalline), was supplied by Sigma-Aldrich, Ireland. Deionized water (Millipore) was used throughout this work. All the reagents used were of analytical grade.

2.2 Isolation of Nanocellulose from Seaweed Solid Waste (SSW)

SSW was obtained by drying the leftover solid material after carrageenan extraction using a 60°C oven for 24 hours and kept at room temperature until further use. Subsequently, the SSW underwent

an acid hydrolysis treatment following the reported method with slight modifications for nanocellulose production [20]. Briefly, 15 g of the dried SSW was soaked overnight in deionized water. The soaked SSW was then centrifuged and filtered to remove excess water from the sample. Bleaching was performed by adding 100 mL of 20% H_2O_2 to the same pot, followed by five-hour incubation at 80℃ with mechanical agitation using a water bath shaker (Daihan Digital Precise Shaking Water Bath MaXturdy™, DaihanSci, South Korea). After that, the mixture was allowed to cool to room temperature, filtered and washed three times with distilled water to remove excess of H_2O_2. Then, acid hydrolysis was performed by adding 8% H_2SO_4 into the same pot containing the suspension and heated to 90℃ for one hour using the same water bath shaker. The suspension was then ultrasonicated in a pulsing mode (15s on and 5s off) for 5 min to ensure homogeneity. Successive washing with deionized water and centrifugation at 12 000 rpm (25℃) were conducted until a constant pH was reached. The white suspension was then kept in a vial at 4℃ until further use and freeze-dried before further analysis.

2.3 Characterizations of Nanocellulose

2.3.1 Fourier-transform infrared (FT-IR) spectroscopy

Fourier-transform Infrared (FTIR) spectroscopy analysis investigated the chemical functionality of isolated nanocellulose, with reference to standard microcrystalline cellulose. The infrared spectrum between 600 and 4000 cm^{-1} was analysed at 25 °C using an Agilent Cary 630 FTIR Spectrometer (Agilent Technologies Inc., USA) with 32 scans and a spectral resolution of 4 cm^{-1}.

2.3.2 Energy dispersive x-ray (EDX) spectroscopy

The elemental analysis was conducted using an energy-dispersive X-ray (EDX) device (Bruker Nano GmbH Berlin, Germany) linked to the SEM at 15 keV and equipped with an XFlash 5010 detector.

2.3.3 X-ray diffraction (XRD)

The crystallographic data of the nanocellulose were obtained using an X-ray diffractometer (Rigaku SmartLab, Rigaku Corporation, Tokyo, Japan) operated at 40 kV and 50 mA. The resulting nanocellulose was scanned over the 3° to 80° diffraction angle (2θ) range using a Cu-K radiation source with a wavelength of 1.54 at 25 °C. Continuous analysis was conducted with a 4.00 °/min scan speed. From the XRD data, the crystallinity index (I_{CR}) was calculated using the Eq. (1) [21,22].

$$I_{CR}\ (\%) = [(I_{200} - I_{am})/ I_{200}] \times 100 \quad (1)$$

where I_{200} is the maximum intensity of the diffraction peak from the (200) plane at $2\theta \approx 22°$ and I_{am} is the intensity of the amorphous region between the (110) and (200) planes ($2\theta \approx 16°$).

2.3.4 Scanning electron microscopy (SEM)

A scanning electron microscope (SEM, Hitachi SU3800) was used to view the surface of nanocellulose to obtain its morphological information. The sample was placed on a stub using double-sided black conducting tape and observed using SEM under vacuum at an accelerating voltage of 5 kV.

2.3.5 Transmission electron microscope (TEM)

The size and shape of the nanocellulose were seen using a Tecnai G2 Spirit BioTWIN transmission electron microscopy (TEM) (FEI, USA) operating at an 80 kV voltage. Before analysis, the diluted nanocellulose suspension was kept in an ultrasonic bath for an hour. After applying a drop of the suspension, TEDPELLA support films (Formvar/Carbon 300 mesh, Copper, approximately 63 m grid hole size) were dried at 60 °C for 20 minutes. The acquired image with a minimum of 120 particles detected was then analysed using Image J analysis software (NIH, USA) to ascertain the size and size distribution of the nanocellulose particles.

2.3.6 Atomic force microscopy (AFM)

The topographical imaging was performed using a Dimension Icon AFM instrument (Bruker, Santa Barbara, CA, USA) that scanned at 0.6 lines per second. After being pulse-sonicated for five minutes, the nanocellulose suspension was spread out on a glass substrate and allowed to dry at room temperature before analysis.

3. Results and Discussion

3.1 Isolation of Nanocellulose from SSW

The SSW was successfully converted into nanocellulose by using a one-pot process. Figure 1 shows the products obtained after each treatment. The slight yellow brownish colour of the SSW is due to the presence of non-cellulosic components including lignin and hemicellulose [23].

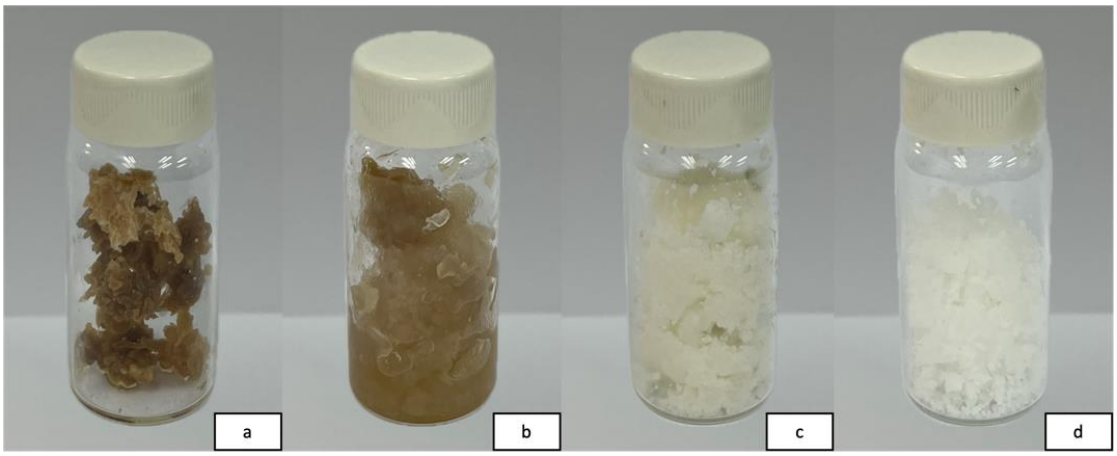

Fig. 1. Products of (a) SSW (b) 24h soaked SSW (c) H_2O_2-treated SSW (d) H_2SO_4-treated SSW

Throughout these processes, these non-cellulosic components were removed, leaving a white appearance of nanocellulose after filtering. The mechanism of nanocellulose production through the one-pot process is depicted in Figure 2 [11,24-26]. Initially, the SSW was subjected to overnight soaking to loosen the lignocellulosic structure by penetrating water molecules. H_2O_2 was used as an oxidizing agent that facilitated the degradation and discoloration of lignin and hemicellulose through the formation of highly reactive hydroxyl (HO•) and superoxide (O_2−•) radicals [27]. Further treatment with H_2SO_4 produces H^+ ions that can penetrate the amorphous domain in the cellulose chain and enhance the hydrolytic cleavage of β-1,4 glycosidic linkages to produce nanocellulose [28]. This is due to the nature of cellulose's low density amorphous regions that are more susceptible to

hydrolytic action and acid exposure, which causes them to break down and release the individual cellulose crystallites [29].

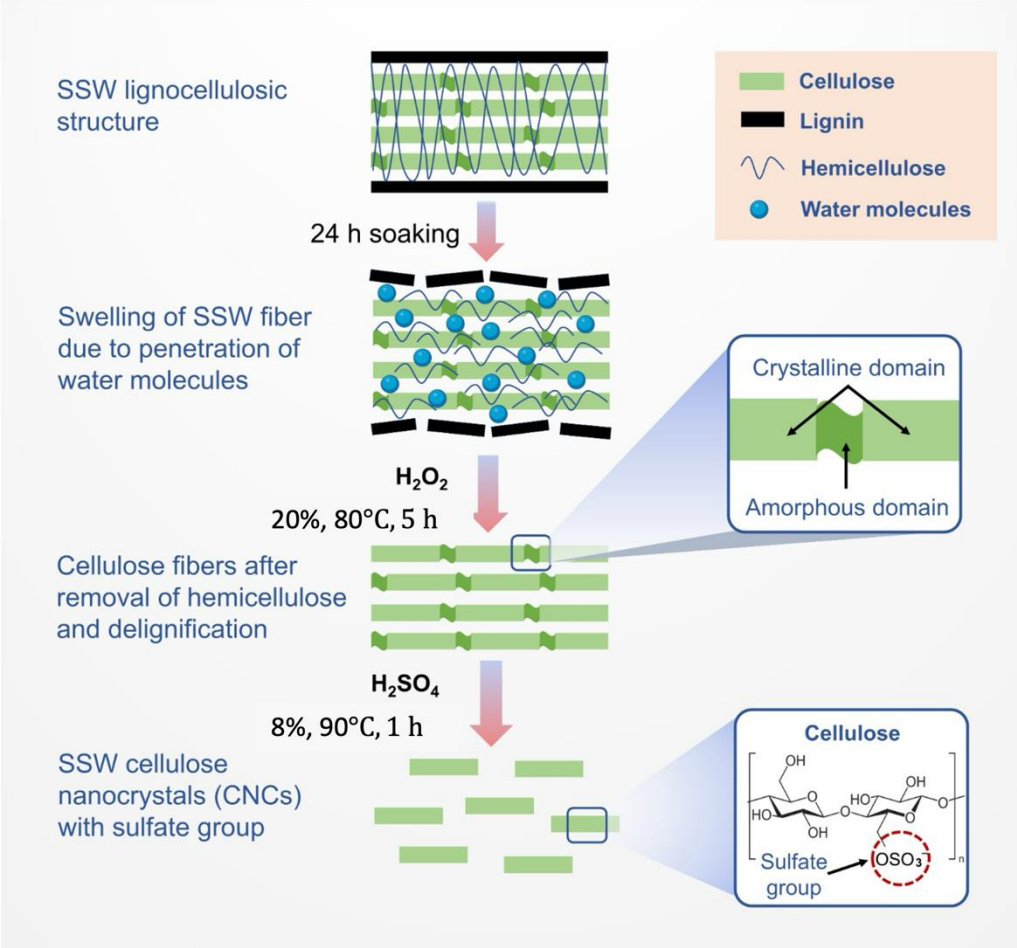

Fig. 2. Mechanism of nanocellulose production from SSW through one-pot oxidative hydrolysis (adapted figure from Mohd Jamil *et al.,* [11])

3.2 Characterizations of Nanocellulose

Figure 3 depicts the FTIR spectra of SSW and nanocellulose obtained after treatment with H_2O_2 and H_2SO_4 with reference to standard microcrystalline cellulose. All samples exhibit a similar characteristic peak, suggesting that the isolation process from SSW does not involve any chemical structure changes or damage during the preparation process and it retained the basic cellulose molecular structure [30]. Broad peaks at 3700–2900 cm^{-1} wavenumber is the most prominent, indicating OH groups of cellulose, bounded by intermolecular hydrogen bonding [31]. The bands between 2896–2885 cm^{-1} corresponded to the asymmetrical stretching vibration of the C–H groups in the polysaccharides. The peak intensity became less apparent after acid hydrolysis due to the decrease in bond strength and polarity [31].

Similarly, the peak at 1640 cm^{-1}, which is a characteristic peak from the stretching of the C=C and C=O group of the lignin aromatic ring, gradually reduced after H_2O_2 and H_2SO_4 treatments, indicating that the lignin from SSW has partially been eliminated [32]. The success of nanocellulose isolation from SSW was further confirmed by the absorption peak in the range of 1400 cm^{-1} and 1338 cm^{-1} associated with CH_2 symmetric bending and C-H bending of cellulose, compared to the untreated SSW sample that exhibited a less prominent peak in the similar region. Such an observation is

obtained as most of the cellulose in the untreated SSW was incorporated by the agar and other non-cellulosic components [33]. Besides, the depicted spectra of nanocellulose obtained after acid hydrolysis were comparable to that of the standard microcrystalline cellulose in Figure 3(d), suggesting the successful isolation of cellulose fibrils to CNCs.

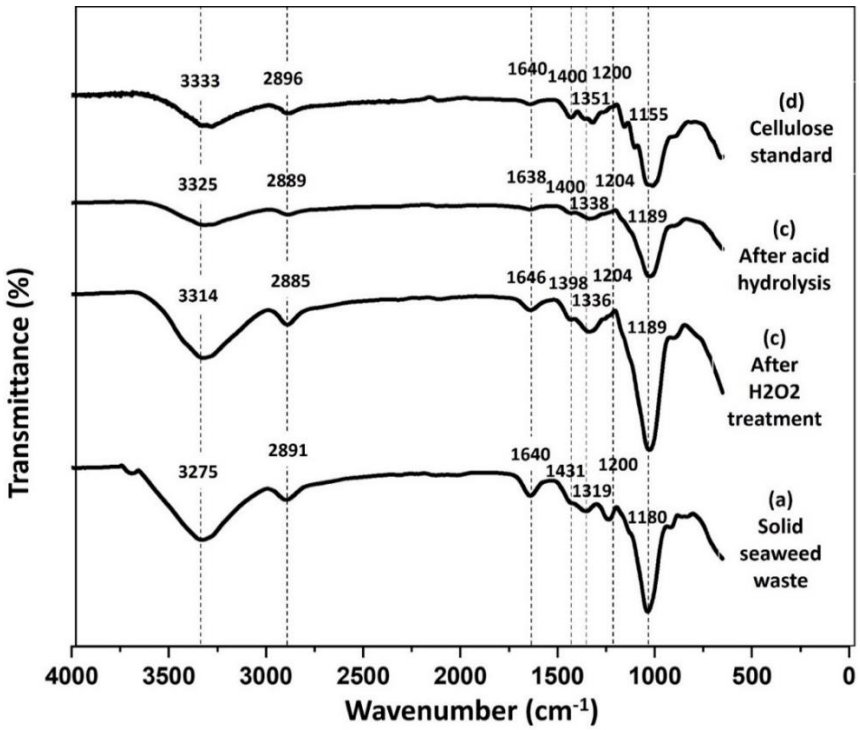

Fig. 3. FTIR spectra of (a) SSW (b) Freeze-dried H_2O_2-treated SSW (c) Freeze-dried H_2SO_4-treated SSW) (d) Microcrystalline nanocellulose

The EDX spectrum in Figure 4 shows the peaks for carbon, oxygen and other elements corresponding to their binding energies. Carbon and oxygen are the significant elements for all samples, implying the cellulose-rich content [34]. The presence of gold is due to the gold sputtering applied to the samples before analysis. For SSW (Figure 4(a)), trace amounts of impurities from sodium (5.13 wt%), magnesium (5.94 wt%) and calcium (0.36 wt%) were detected which were eliminated during the H_2O_2 treatment (Figure 4(b)). The presence of alkaline earth metal species such as sodium, magnesium and calcium are common in algae biomass, due to its high mineral content [35,36]. Sulphur element in the spectrum of nanocellulose obtained after hydrolysis evidenced (Figure 4(c)) the formation of nanocellulose with sulphate group due to the esterification between H_2SO_4 and the hydroxyl group of cellulose.

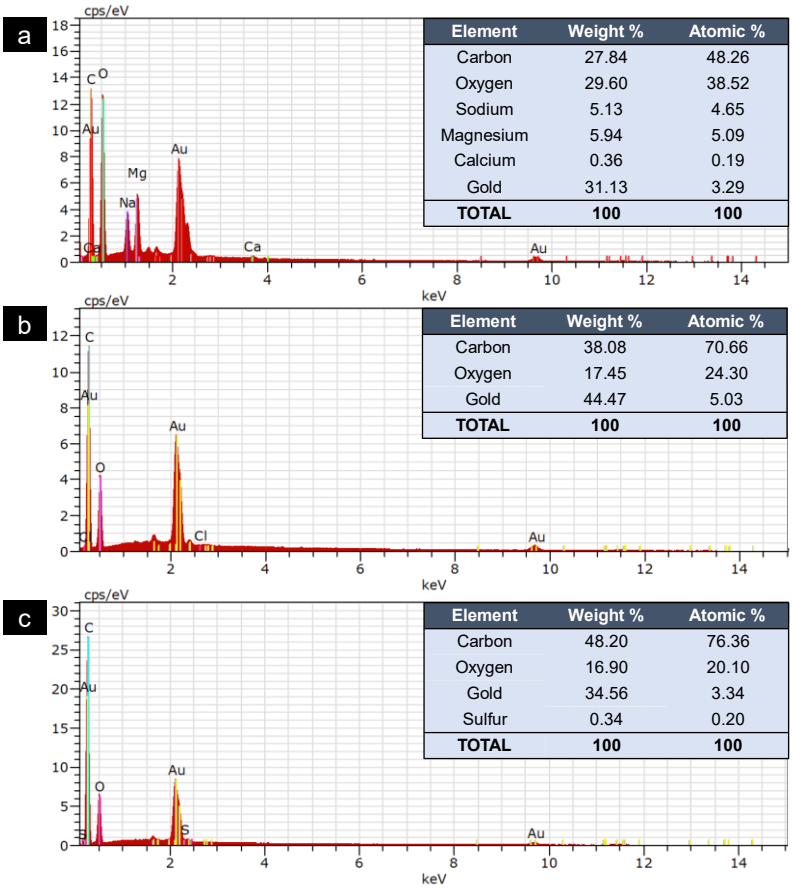

Fig. 4. EDX spectra and the corresponding elemental composition of (a) SSW, (b) H_2O_2-treated SSW and (c) H_2O_2/H_2SO_4-treated SSW

Crystallinity is one of the main factors determining the mechanical properties of nanocellulose [37]. XRD analysis obtained crystallographic information on the SSW before and after oxidative hydrolysis. As shown in Figure 5, all samples exhibit prominent peaks at 2θ = 16°, 22° and 34° corresponding to the (110), (200) and (004) lattice planes of cellulose [38]. The findings are consistent with other research [13,39,40] and confirm that the original structure of native seaweed cellulose was retained after the isolation process [39]. An additional peak at 2θ = 18° in the spectra of the SSW indicates the presence of impurities from inorganic substances, in line with the EDX data presented in Figure 4(a).

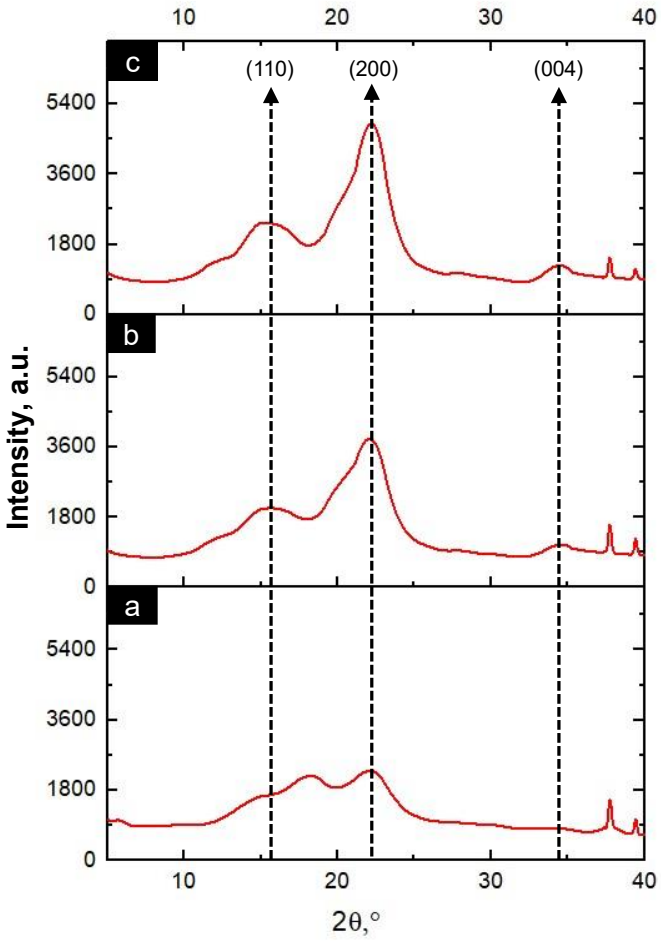

Fig. 5. XRD pattern of (a) SSW (b) H_2O_2-treated SSW (c) H_2O_2/H_2SO_4-treated SSW

As shown in Table 1, the SSW has low crystallinity (18.68%) due to the high content of amorphous components. A remarkable increase in crystallinity index was observed after the H_2O_2 treatment (54.16%) due to the elimination of hemicellulose and the delignification process and the removal of amorphous domains of cellulose [40,41]. Nanocellulose obtained after acid hydrolysis has considerably high crystallinity (63.65%), comparable to that of the nanocellulose isolated from *Gelidiella aceroso* seaweed (60%) and other biomass such as Spanish poplar (65%) [42] and cocoa pod husk (67.60%) [16]. The high crystallinity and sharp diffraction peak indicate stronger hydrogen bonding interaction between the nanocellulose chains, which create a highly crystalline and compact structure [43]. High crystallinity is a significant characteristic of nanocellulose, where the amorphous regions of cellulose are hydrolysed by acid treatment to produce nanocellulose [7].

Table 1
Crystallinity index of SSW before and treatment with H_2O_2 and H_2SO_4

Sample	Crystallinity index, %
SSW	18.68
H_2O_2-treated SSW	54.16
H_2O_2/ H_2SO_4-treated SSW	63.65

The morphological structures of freeze-dried SSW and suspensions after each treatment were observed using SEM. As presented in Figure 6, freeze-dried SSW forms a sheet-like structure [44,45],

suggesting the cellulose-rich content in the seaweed cell wall [19]. The microfibrillar structure of cellulose could be seen after the H_2O_2 treatment (Figure 6(b)) due to the removal of non-cellulosic components from the SSW [18]. Further treatment with H_2SO_4 causes defragmentation of the microfibril into nanocrystals (Figure 6(c)) [46]. During hydrolysis, amorphous areas decay and crystalline areas remain intact. The acid treatment allows the amorphous portions of microfibrils that are farther apart, have a lower density and are more accessible for hydrogen bonding with other molecules like water to hydrolyse, leaving behind a highly crystalline residue [7,47]. Moreover, the observed typical honeycomb structure and porous cellulose network after freeze-drying signifies the successful isolation of nanocellulose from SSW [18].

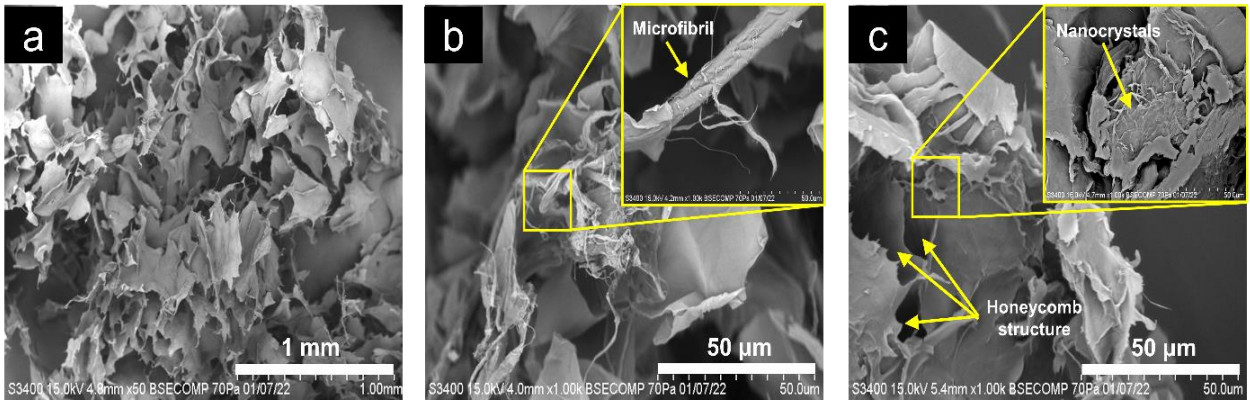

Fig. 6. SEM images of (a) SSW (b) Freeze-dried H_2O_2-treated SSW (c) Freeze-dried H_2SO_4-treated SSW

The TEM image in Figure 7(a) visualized the needle-like morphologies of nanocellulose, commonly known as cellulose nanocrystals (CNCs). This is a typical structure of nanocellulose produced through acid hydrolysis, as reported in several studies [48-50]. The image was processed further to obtain the size distribution and mean diameter.

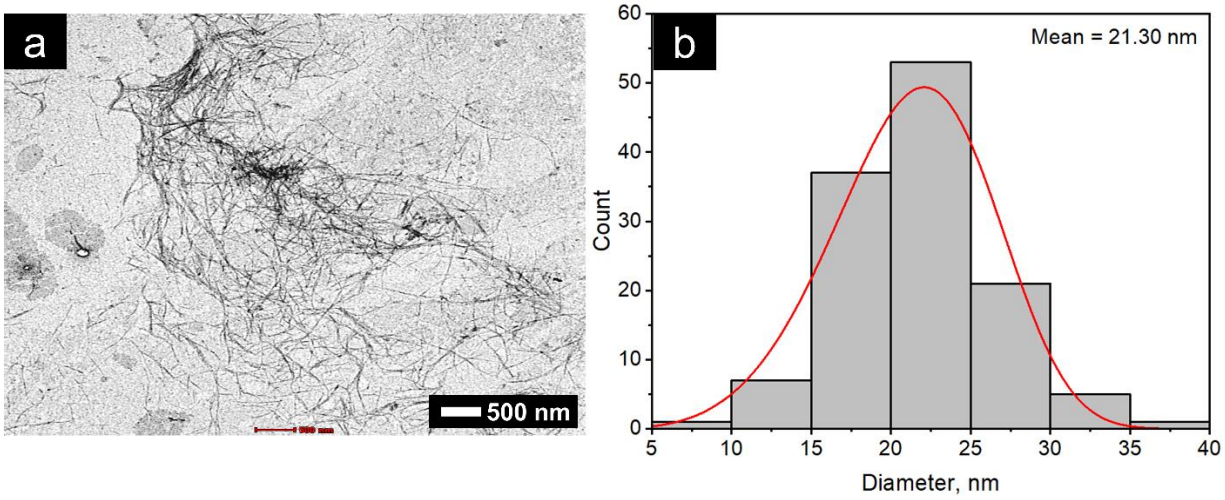

Fig. 7. (a) TEM image (b) the corresponding size distribution of the nanocellulose obtained from SSW after sulfuric acid hydrolysis and ultrasonication

From Figure 7(b), the nanocellulose formed a normal distribution curve with a narrow size distribution in the 6 to 35 nm range, with an average diameter of 21.30 ± 4.62 nm, comparable to a similar reported study on the production of CNCs from different seaweed species as shown in Table 2.

Table 2
Diameter of CNCs obtained from different seaweed sources

Source	Diameter (nm)	References
K.alvarezii (SSW)	21.30 ± 4.62	Present study
Gelidium elegans (SSW)	21.8 ± 11.1	[33]
Unpurified algae waste	9.1 ± 3.1	[12]
S. fluitans	43.06 ± 8.94	[31]

Furthermore, AFM analysis was used to examine the isolated nanocellulose's size, distribution and dispersion. The needle-like morphologies [18,51] and distribution of the CNCs, as demonstrated by the sample's two-dimensional (2D) topographical AFM scanning in Figure 8(a), are consistent with the findings from the TEM and SEM. The height analysis of the nanocellulose surface, as shown in the 3D structure of Figure 8(b) can be used to deduce the roughness of the sample based on Ra and Rq values of 10.6 nm and 12.7 nm, respectively, revealing the low surface roughness of the produced CNCs. The low surface roughness in soft material like nanocellulose is expected as they provide better surface qualities due to their hydrophilicity [52] and thus are more desirable in optoelectronics, biomedical and film packaging application [33].

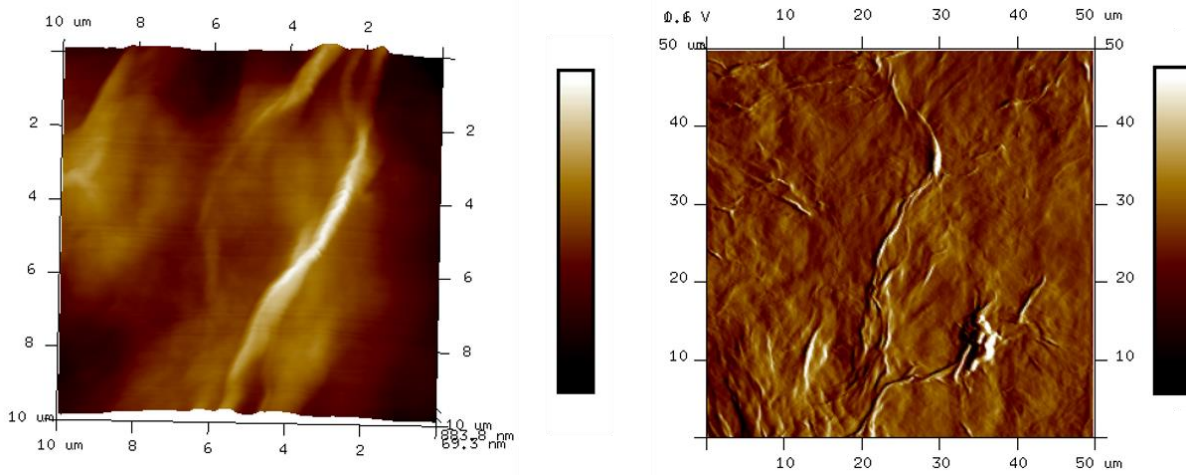

Fig. 8. AFM images (a) in 2D (b) in 3D

4. Conclusion

The present study provides insight into the potential of underutilized solid seaweed waste (SSW), a residue obtained after carrageenan extraction from K. alvarezii, as a promising source of cellulose nanocrystals. Applying one-pot oxidative hydrolysis using H_2O_2 and mild sulfuric H_2SO_4 hydrolysis provides simpler alternatives for nanocellulose production, as evidenced by the FTIR, EDX, XRD, SEM, TEM and AFM data. The SSW nanocellulose exhibits a needle-like structure with an average diameter of 21.30 ± 4.62 nm. The crystalline properties of the SSW nanocellulose are beneficial for application in food packaging and flexible electronics.

Acknowledgements
We would like to thank Malai Roziah Syafiqah Malai Rozlan and Nurul Athirah Binti Mohd Kamarulzaman from Kulliyyah of Science, International Islamic University Malaysia for their helpful and valuable contributions during the early stage of this project. This work was supported by Universiti Malaysia Sabah (UMSGreat Grant No. GUG0550).

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